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Detritus - Multidisciplinary Journal for Waste Resources and Residues - is aimed at extending the "waste" concept by opening up the field to other waste-related disciplines (e.g. earth science, applied microbiology, environmental science, architecture, art, law, etc.) welcoming strategic, review and opinion papers. **Detritus is indexed in Emerging Sources Citation Index (ESCI) Web of Science, Scopus, Elsevier, DOAJ Directory of Open Access Journals and Google Scholar**. Detritus is an official journal of IWWG (International Waste Working Group), a non-profit organisation established in 2002 to serve as a forum for the scientific and professional community and to respond to a need for the international promotion and dissemination of new developments in the waste management industry.

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Editorial

PUBLISHING: ARE WE ON THE RIGHT TRACK?

Publication is an essential part of scientific work. The dissemination of results represents a fundamental step in expanding basic knowledge to foster the development of innovative, more effective, less costly and/or more environmentally friendly technologies or processes. This was the initial intention underlying the publication of research studies and remains valid today. Publication nowadays represents an integral part of education, and whilst it is not expected that a master or PhD thesis will necessarily result in the identification of new inventions or fundamental processes, a series of stepping stones may be added to our specific knowledge. When considering the vast number of theses produced worldwide, the more innovative could indeed contribute towards furthering knowledge and producing new technologies or developments.

In order to progress in our scientific careers, there is a general "requirement" to produce innovative results on a yearly basis. A certain number of publications, preferably in high impact factor scientific journals, are mandatory prior to gaining authorization to proceed with a PhD viva or in achieving qualifications for an academic career. Considerable pressure is therefore placed on students and academics at Universities and research institutions. Of course, the idea behind this approach is to encourage researchers and students in undertaking their studies and publishing the results obtained. However, research is not predictable, it is time-consuming and failures do occur. Indeed, I still have the – perhaps romantic idea - that researchers are very much devoted to working under their own incentive…

This impelling need for publications resembles the sword of Damocles hanging over the academic community. If, for a series of reasons, adequate material for publication is not available, an alternative means of "survival" will need to be identified. For this reason, research results suitable for presentation in a single paper may be divided into a series of potential publications. Moreover, increasingly lengthy lists of authors may be acknowledged on the paper although their input is not always clear; at times it is almost as if names are included merely to boost the individual's number of publications. This practice has been manifested as the result of the stringent requirements relating to number of required publications per year.

Indeed, when evaluating the scientific merit of an academic, the number of publications may at times be held in higher esteem than quality or relevance. A recognised thorny issue, but greater care and attention should be placed on validating publication rather than restricting the focus to a mere counting of numbers.

I should also like to highlight another important aspect. To open up research to new areas and gain more progress in waste management, an interdisciplinary research field is required. This approach is indeed already required for larger joint research projects, but implementation is not an easy task. First of all, appropriate partners from other disciplines need to be identified; although we may all have colleagues who study other disciplines and would be willing to cooperate, in my view interdisciplinary research is complicated by the fact that each discipline uses its own "language". Communicate therefore becomes an issue. As an example, a cooperation between social scientists and environmental engineers is rendered problematic due to the difficulty for an engineer to understand the "language" social scientists use, and probably vice versa. The reason as to why each discipline uses a specific "language" and how important this may be falls beyond the scope of this article, but I confess that this situation is a barrier not only for mutual learning but also for distributing results to the public. At times I feel that scientific work that is easy to grasp is not "real science", - science needs to be cleverly packaged to be accepted in an academic circle, sometimes it seems to me a bit as "l´art pour l´art".

I strongly believe that we should present ideas, processes and scientific results in publications in a "simpler", interdisciplinary, more understandable, but of course, correct way. This is not easy. On the other hand, when it comes to the acceptance of highly specific and detailed research by colleagues working in the same field, to enhance communication use of the field-specific "language" may be necessary. We must however strive to find ways to communicate our specific messages in an easy understandable way to other disciplines and the public. This aspect has become even more important following the increasing use of electronic media in the dissemination of results from research, reflections and investigations – we should not entrust this task to non-experts.

Over a period of 12 years I chaired a joint project in which natural scientists and engineers - both from different disciplines –worked together on the treatment of contaminated soils. In the first year we had to learn to communicate and mutually understand each other, at times being required to grasp different approaches used in dealing with specific issues. At the outset, the presentation of intermediate research reports was complex, but by continuously asking questions, we were able to overcome this difficulty to work successfully together and we all learnt a lot from each other; I think that my biggest gain in knowledge was

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FIGURE 1: The Great Transformatiom, Climate – can we ban the heat? (Anonymous, 2014).

obtained through cooperating in this project.

An interesting example of the presentation of scientifically correct facts and processes is given in a cartoon well- prepared by the German Advisory Council Germany (WBGU) (Anonymous, 2014). This is addressed to the general public and is available free of charge. We need more of these approaches.

I should also like to draw your attention to another point. This relates to the frequently stringent rules that papers submitted for publication have to meet. The presentation of strategy papers, visions or overviews may not be allowed as they do not meet the set requirements. However, at times, the above papers may actually have a higher scientific and public impact than research on a highly specific topic. I suggest that these regulations should be reviewed. Of course, these strategy or position papers would need to be sent out for peer review whilst bearing in mind potentially amended acceptance criteria.

During my career as a reviewer, I have also noticed issues relating to citation. This is not a huge issue, but in my view, particularly with regard to publications received from young researchers, an abundance of citations may be present. Although an exaggeration, levels of citations are at times along the lines of “there is less rainfall in the desert than in tropical areas”. This is common knowledge and needs no citations. The authors’ own thoughts and ideas would be more relevant may not necessarily need extensive use of citations. We should be more creative and self - confident of our own ideas rather than relying heavily on the work of others. I agree that it is essential to refer to the results and experiences of other colleagues in a research project, this is not the point, but when presenting your own ideas citation may not be needed. Of course, to a certain extent this is the result of the widely adopted publication policies: extensive citations are seen to add to the value of the paper; this may at times be true, but there is ample room for manipulation.

To continue - if authors are cited are we sure that the messages and/or results conveyed are correct? Do we know all the circumstances and conditions as to how the cited results have been elaborated? Do we always demonstrate the correctness of mathematical modelling presented? Citations are ultimately the responsibility of the authors, who have used citations that were a best fit with their own research. Particularly in view of the enormous number of new journals, frequently open access, it has become more difficult to judge the correctness of published papers.

Just to clarify, literature review for scientific work is of course fundamental, and correct citation is naturally part of this scientific work; it is however imperative that the content should be critically reviewed prior to citing it – never make the mistake of merely citing a paper on the grounds of it being published in a peer reviewed journal (which of course should provide more confidence). My advice is to undertake a more extensive search of publications relating to the specific subject of interest. Critical judgement should always be applied.

It may take a long time before we succeed in modifying the world of scientific publication, but perhaps from time to time we should rethink and discuss procedures that have developed over the years and that produce such a significant influence on scientific work.

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A SPATIAL-AND-SCALE-DEPENDANT MODEL FOR PREDICTING MSW GENERATION, DIVERSION AND COLLECTION COST BASED ON DWELLING-TYPE DISTRIBUTION

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ABSTRACT

Comprehensive models were developed to predict waste generation for different collection streams. Taking into account the dwelling-type distribution encountered during the different waste collections, it was possible to better capture the waste generation variability. Using the same approach, collection and transportation cost models were also developed. This series of models were validated using data from the Urban Agglomeration of Montreal (UAM), which is composed of 33 districts with widely different scales of population and dwelling characteristics. The unknown parameters of the models were identified through mean square regressions applied on the real data available for the case-study. For example, values of 1.364, 1.019 and 0.500 t/(dwelling.yr) were identified for the total quantity of wastes generated in single-family, duplex and other dwelling, respectively. Using the same approach, it was possible to determine collection time as a function of the dwelling-type distribution along the collection route. Values of 28.7 s, 11.4 s and 5.22 s were identified as the collection time per dwelling for single-family, duplex and other dwelling, respectively. Equipped with a combination of fitted parameters and reported values from the literature, the models were used as predictive tools. Three features are illustrated in this paper: 1) the simulation of various scales for the generation, diversion and specific collection cost; 2) the effect of adding a new collection stream; 3) the effect of an increase of the citizen participation to a specific collection stream. Predicted results enable decision-makers to have access to very useful information.

1. INTRODUCTION

In the context of municipal solid waste management, it is frequently useful or necessary to estimate the generation and diversion of wastes at different spatial scales (e.g. country, county, city, borough), including at very small scale (e.g. neighbourhood, dwelling, household, individual). Activities where such information becomes essential include the development of future policies (e.g. new collection stream, financial incentive) or public information campaigns adapted to local context and citizen behaviour, as well as waste collection planification or route optimization, in terms of budget, quantities or dwelling-type distribution. All these planning activities are known to be strongly influenced by how MSW are generated and diverted. The case of collection cost is a good example. It is mainly defined by the available or planned collection structure, by the urbanistic characteristics of the collected area (e.g. dwelling-type distribution), by the amount of waste generated in total, and by the diversion of the waste in the collection streams. When the participation of citizens to a particular collection stream increases, this will not only directly influence the collection cost related to this stream, but also the collection cost for other streams.

Usually, data needed to quantify generation and diversion are available only at large scales, and information about collection cost, when publicly available, are difficult to use in different contexts and scales. In the literature, several studies have highlighted the relations between generation, diversion or collection costs and various socio-demographic indicators. Some mathematical models are also described and allow a fair estimation of those parameters. Goel et al. (2017) reviewed the different modeling approaches used in order to forecast solid waste generation rates. For example, some models are integrating geographical information system (GIS) information to predict waste generation rates as a function of household size

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and other socioeconomic conditions (Purcell and Magette, 2009, Akther et al., 2016). Application of GIS-based methods to link waste generation rate to a multiplicity of factors is increasing as the GIS-linked information becomes more readily available. Although the type of dwellings found in a neighborhood is directly linked to demographic and socioeconomic factors, only one study was found where the waste generation rate was correlated to the dwelling unit size (Grossman et al., 1974). Dwellings types are known to be correlated to the household size, demographic profiles and income level. Since the dwelling-type distribution along a waste collection route can be easily obtained using GIS information, this approach seems to offer a very promising basis for the development of a simple waste prediction tools.

The Chaire de Recherche sur la Valorisation des Matières Résiduelles (CRVMR, Research Chair on Advanced Waste Recovery) at Polytechnique Montreal, is currently developing a methodology to assess the sustainability of Waste Management Systems (WMS), based on the integration of three distinct VMR (for Valorisation des Matières Résiduelles) tools:

VMR-Gen: Agent-Based Model (ABM) to predict the behaviour of the waste generator, providing the MSW flows and compositions of the source-sorted waste streams;

VMR-Sys: Material flow analysis (MFA) based framework to calculate waste and product flows and stocks throughout the WMS. Comprehensive process modules, one for each waste treatment technology, are developed and integrated into this framework;

VMR-Imp: Waste LCA modelling to evaluate the WMS impacts.

To accompany the development of VMR-Gen, we developed a series of predictive models, adaptable to variable scales, to predict the MSW generation, diversion and specific collection costs (in $/dwelling and $/t). The main parameters of the model are related to the dwelling-type distribution (e.g. single-family house, duplex, triplex, apartment buildings), which has shown to have a key influence on generation, diversion and collection cost.

In this paper, the models will be described briefly, and key results will be presented and discussed in order to illustrate three features: 1) the simulation of various scales for the generation, diversion and specific collection cost; 2) the effect of adding a new collection stream; 3) the effect of an increase of the citizen participation to a specific collection stream.

2. MATERIALS AND METHODS

The following section describes in the first part the waste generation model, which is developed based on the dwelling-type distribution. In the second part, the dwelling-type distribution approach is used to develop a collection and transportation cost model.

The proposed models are developed in order to represent the waste generation, diversion and collection costs on a given territory. The territory used as case-study for this work is that of the Urban Agglomeration of Montreal (UAM), which is constituted by the 19 boroughs of the City of Montreal and 14 linked municipalities. The UAM occupies the entire Island of Montreal, Quebec (Canada).

2.1 System description

A territory is described as a spatial zone with the following properties: population, number of dwellings, dwelling-type distribution, distance travelled during one collection, average distances to the waste treatment facilities, annual quantity of waste generated, and annual quantities of waste collected in source-separated streams.

2.2 Waste stream models

It is assumed that the generation of waste and the sorting of waste between available source-separated collection streams is constant for a given dwelling-type on a territory. Then, the annual quantity of waste generated on a territory (M_{tot}) is expressed as:

$$M_{\text{tot}} = \sum_{i \in D} n_i \, m_{\text{tot}}^i \quad (1)$$

where:
D is the set of dwelling-types (e.g. single-family houses, duplex, other)
n_i is the number of dwellings of type i on the territory
m_{tot}^i is the quantity of waste generated annually in a dwelling of type i

Let C be the set of source-separated collection streams available on the territory, then, for each c in C, the annual quantity of waste collected in the stream c (M_c) is expressed as:

$$M_c = \sum_{i \in D} n_i \, m_{\text{tot}}^i \, x_c^i \, \tau_c^i \, \frac{1}{\pi_c^i} \, \sigma_c^i \quad (2)$$

where:
x_c^i is the mass fraction of the waste stream generated that should be placed in collection stream c, for a dwelling of type i
τ_c^i is the recovery rate of collection stream c, for a dwelling of type i
π_c^i is the purity of collection stream c, for a dwelling of type i
σ_c^i is the fraction of dwelling of type i having access to collection stream c

The residual quantity of waste (M_{RES}) is expressed as:

$$M_{\text{RES}} = M_{\text{tot}} - \sum_{c \in C} M_c \quad (3)$$

Finally, the global waste diversion on a territory (Y) is defined as:

$$Y = \frac{\sum_{c \in C} M_c}{M_{\text{tot}}} \quad (4)$$

2.3 Collection and transportation models

Based on the modelling approach described by Tanguy, Villot, Glaus, Laforest, & Hausler (2017), the models for collection and transportation costs are defined in similar ways, considering two main contributions: cost of the fuel and the hourly operating cost for the truck.

For the collection cost model, taking into account the dwelling-type distribution along the route in order to estimate the average collection speed will be considered. It is reasonable to suppose that the required time to pick up wastes from one dwelling is a function of the dwelling-type. For example, a single-family dwelling will require a longer collection time than a multiplex dwelling, since in the latter case multiple dwellings are collected during a single stop, hence yielding a shorter collection time per dwelling.

For the transportation costs model, one may assume that these costs are simply related to the distance travelled between the territory where the waste is collected and the final destination. The dwelling-type distribution should not have an effect on these costs.

2.3.1 Collection cost

The annual collection cost for a stream s ($s \in \{C, RES\}$, meaning source-separated streams or residual stream) is defined as:

$$k_{\mathrm{col}}^{s} = \left(k_{\mathrm{fuel,col}} + \frac{k_{h,\mathrm{col}}}{v_{\mathrm{col}}^{s}}\right) d_{\mathrm{col}}^{s} N_{col}^{s} \tag{5}$$

where:

$k_{fuel,col}$ is the fuel cost per km for the collection
$k_{h,col}$ is the hourly cost of the truck for the collection
v_{col}^{s} is the average collection speed for the stream s
d_{col}^{s} is the distance travelled during one collection of stream s
N_{col}^{s} is the number of collections of stream s in a year

The average collection speed for the stream s is defined as:

$$v_{\mathrm{col}}^{s} = \frac{d_{\mathrm{col}}^{s}}{t_{\mathrm{col}}^{s}} \tag{6}$$

where:

t_{col}^{s} is the average collection time for the stream s expressed as a function of the dwelling-type distribution:

$$t_{\mathrm{col}}^{s} = \sum_{i \in D} n_i \, \alpha_s^i \tag{7}$$

where:

α_s^i is the average collection time for the stream s in the case of a dwelling of type i. For this averaging process, possible covariance terms were not considered. This could be revised in a future improvement of the model.

2.3.2 Transportation cost

The annual transportation cost for a stream s ($s \in \{C, RES\}$) is defined as:

$$k_{\mathrm{tran}}^{s} = \left(k_{\mathrm{fuel,tran}} + \frac{k_{h,\mathrm{tran}}}{v_{\mathrm{tran}}^{s}}\right) 2 d_{\mathrm{tran}}^{s} M_s \frac{1}{L} \tag{8}$$

where:

$k_{fuel,tran}$ is the fuel cost per km for the transportation
$k_{h,tran}$ is the hourly cost of the truck for the transportation
v_{tran}^{s} is the average transportation speed for the stream s
d_{tran}^{s} is the transportation distance from the centroid of the territory to the destination of stream s
M_s is the annual quantity of waste collected in the stream c
L is the mass capacity of the transportation truck

2.3.3 Mass and dwelling specific collection and transportation costs

The mass specific collection (K_{col}^{s}) and transportation (K_{tran}^{s}) costs for a stream s are defined as:

$$K_{\mathrm{col}}^{s} = \frac{k_{\mathrm{col}}^{s}}{M_s} \tag{9}$$

$$K_{\mathrm{tran}}^{s} = \frac{k_{\mathrm{tran}}^{s}}{M_s} \tag{10}$$

The dwelling specific collection ($\tilde{K}_{\mathrm{col}}^{s}$) and transportation ($\tilde{K}_{\mathrm{tran}}^{s}$) costs for a stream s are defined as:

$$\tilde{K}_{\mathrm{col}}^{s} = \frac{k_{\mathrm{col}}^{s}}{\sum_{i \in D} n_i} \tag{11}$$

$$\tilde{K}_{\mathrm{tran}}^{s} = \frac{k_{\mathrm{tran}}^{s}}{\sum_{i \in D} n_i} \tag{12}$$

2.4 Case-study

Data collected for the Urban Agglomeration of Montreal (UAM) for the year 2016 were used for this case-study. Characteristics of the 33 individual districts (the 19 boroughs of the City of Montreal and 14 linked municipalities) are presented in Table 1. Data presented include the population, the number of dwellings regrouped under three different types: single-family, duplex and other (3 or more apartments), the annual collected weight of total wastes and recyclables. The UAM territory was selected for this case-study because it comprises a wide variety of districts having different scales in terms of population, numbers of dwellings and relative fraction of dwelling-type habitations.

In Figure 1, the number of dwellings and the fraction of single-family houses are presented as a function of the population for the 34 districts of the UAM. For the most populous districts, we observe that the fractions of single-family households are much smaller than for those with lesser population. Trend lines giving the total number of dwellings in a district considering occupancies between 1.5 and 3.0 persons/dwelling are also shown in Figure 1. For districts where there is a high fraction of single-family households, the points are close to the 3.0 occupancy trend line, while the points for districts with high number of other dwellings (multiplex) are close to the 1.5 occupancy trend line. This is consistent with reported data on the average size of household in different types of dwelling in Quebec (2.7 for single-family, 2.2 for duplex and 1.9 for multiplex) (Lagneau, 2018).

2.5 Model parameters

Some of the model parameters presented above may be obtained from published waste characterization results. RECYC-QUEBEC publishes a comprehensive report every two years, where it presents the wastes collected in different streams, including household wastes, recyclable materials (REC) and organic wastes (ORG) collected in the Province of Quebec. Since the UAM data are part of those results, the reported values from RECYC-QUEBEC were used to estimate some parameters in our generation models. These parameters and the reported values are shown in Table 2.

The fuel costs per km and the hourly costs are considered constant for both collection and transportation and equal to 0.89 \$/km and 72 \$/km, respectively. The mass capacity of the transportation truck is fixed to 10 t.

3. RESULTS AND DISCUSSION

The unknown parameters of the model (m_{tot}^{i} and $\alpha_{\mathrm{RES}}^{i}$) were identified through mean square regressions applied on the real data available for the case study. The fitted pa-

TABLE 1: Characteristics of the 33 districts of the UAM in 2016 (Montréal, 2017).

District	Population	n^{single}	n^{duplex}	n^{other}	M_{tot} [t/yr]	M_{REC} [t/yr]
AC	136461	8372	15781	38346	46479	8762
AJ	44567	3152	6039	10288	14775	2666
BF	19801	6264	8	642	9870	2268
BU	3900	1324	2	46	2096	401
BV	4980	989	293	850	2251	558
CL	33847	4113	1334	9044	11819	2169
CN	172961	6483	19160	57941	56038	11698
DO	50789	12461	202	4919	20809	3717
DV	19431	4334	372	4291	7822	1912
HS	7279	1480	529	658	2641	639
IS	19123	4885	471	1536	8576	1766
KL	21270	6325	14	463	9230	2184
LC	45003	4802	4370	12610	17598	3403
LN	81777	2933	9065	19947	27990	5183
LR	101530	9412	5843	24921	31539	6479
LS	79651	3724	16681	16322	26365	6309
ME	3846	510	408	924	1916	300
MH	139612	6477	16840	47944	49656	10844
MN	89145	4547	10759	21318	30048	6000
MO	5212	1218	579	174	2192	454
MR	20869	4156	739	3043	10323	2126
OM	25043	1985	1144	6694	8920	2182
PC	31898	8572	229	3756	13770	3617
PM	105139	1865	5973	57432	38712	8967
PR	72399	16073	1185	9044	26974	5499
RO	142578	3578	14404	60138	49614	12523
RP	111617	21556	4663	16743	40832	8717
SO	78027	2804	7528	32595	28179	6356
SV	929	346	10	7	577	127
VD	70527	2933	4169	29457	24122	6626
VM	88799	1548	1762	59209	30719	8633
VS	149075	3819	18858	47162	49562	7931
WM	20621	3563	579	4794	8968	1917

rameters together with the parameters presented in Table 2 are then used to solve the model in order to simulate the waste generation and collection for each district of the case-study. The results of the simulations are presented and discussed in this section.

3.1 Generated stream

The values of 1.364, 1.019 and 0.500 t/(dwelling.yr) have been identified for m_{tot}^{single}, m_{tot}^{duplex} and m_{tot}^{other}, respectively. The comparison of the predicted generated quantities with the real quantities is presented in Figure 2. A very good agreement between predicted and real values is observed. Figure 2 shows how the predictions of MSW generation are improved when the distribution of dwelling-types along the collection route is taken into account (R^2 = 99%), compared to predictions using a model based solely on the total number of dwellings being collected and a unique average value for m_{tot} equal to 0.698 t/(dwelling.yr) (R^2 = 92%). This figure clearly shows that the dwelling-type distribution has a real influence on the quantities of MSW being generated along a collection route, as quantified by the values of m_{tot}^i, and that averaging the generation per collected dwelling does not capture this difference.

3.2 Collected stream

In the case of the source-separated collection stream of recyclables (REC), the comparison between predicted and real quantities of collected recyclables is presented in Figure 3. It is worth mentioning that in this case, we only used values reported in the literature for the model param-

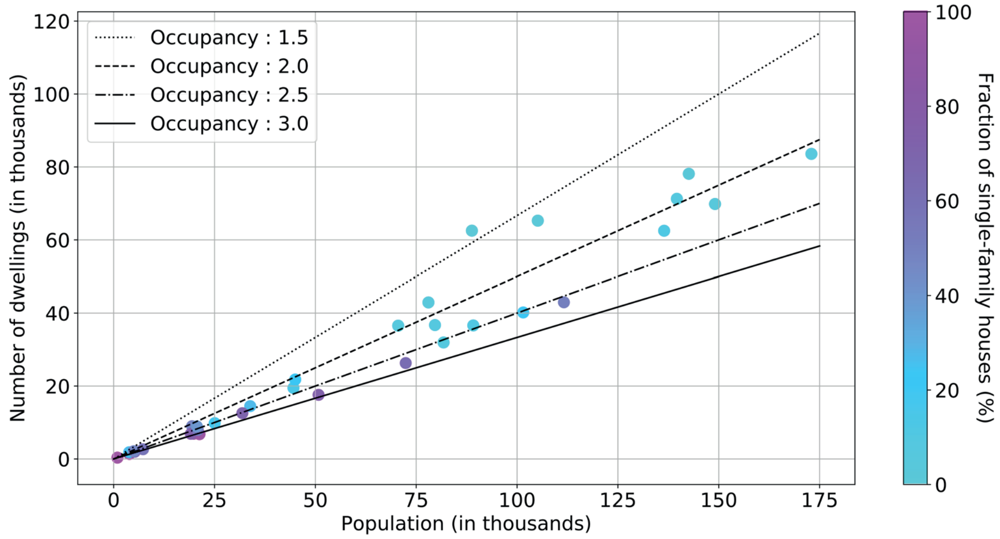

FIGURE 1: Number of dwellings and fraction of single-family dwellings as a function of the population for the 33 districts of the UAM (trend lines show the number of dwellings as a function of the population in the districts, for different occupancies).

eters. Again, a very good agreement between the predicted and the real values is observed, although a greater dispersion in the resulting fit is seen.

Since the deployment of the source-separated collection stream for the organics (ORG) is still recent and in progress, it is not possible to compare the model predictions for this stream with real data as they are not available for all districts. Consequently, this stream is combined with the residual stream. The comparison of the predicted collected quantities with the real quantities, in the case of the combination of the residual stream with the source-separated organics collection, is presented in Figure 4.

The fitted parameters for each collection type and dwelling type are in reasonable agreement with published results from RECYC-QUEBEC. From the generation model, it is possible to estimate the fraction of total waste that is being diverted in the other source-separated collection streams (recyclables and organic wastes where available). A global diversion rate of 22% on average is observed for all UAM districts. Results presented so far confirm that using an approach that takes into account the types of dwellings encountered along the collection route provides a more accurate predictive model of waste generation and diversion.

3.3 Collection and transportation costs

Since the objective here is to estimate the collection and transportation costs without including profit margin and other fixed costs that would be charged by a private waste collector, it is not possible to compare these cost estimates with the actual contractual costs. Only the variations from one district to another and the influence of the source-sorting behaviour of the citizens on the trends and orders of magnitudes of these predicted costs will be discussed.

As demonstrated above, the use of the dwelling-type distribution as the basis of our generation predictive model gave good results. The same approach was used to determine collection time as a function of the dwelling-type distribution along the collection route. Using data from a number of collection sectors where the residual waste collection trucks were equipped with GPS (see Table 3), we were able to identify the values of 0.00797 h (28.7 s),

TABLE 2: Model parameters obtained from reported values in the literature (Lagneau, 2018).

Parameter	Single-family	Duplex	Other
x^i_{REC}	32.3%	33.1%	36.8%
x^i_{ORG}	43.3%	32.3%	33.6%
τ^i_{REC}	65%	60%	48%
π^i_{REC}	90%	90%	85%
σ^i_{REC}	100%	100%	100%

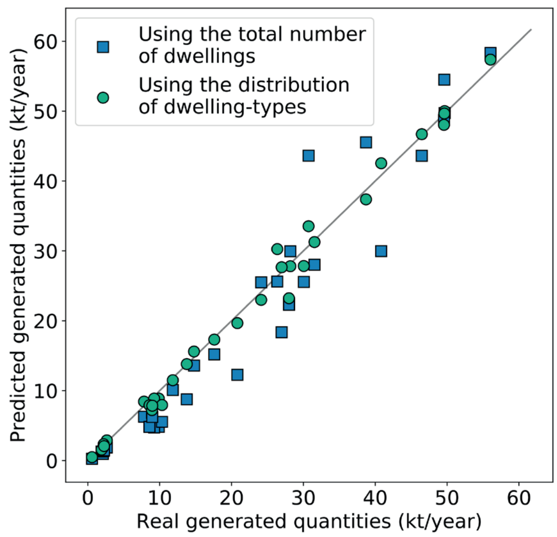

FIGURE 2: Comparison between predicted and real quantities of total generated wastes (Mtot) for the UAM districts based on two approaches: (green circles) using the dwelling-type distribution; (blue squares) using the total number of dwellings.

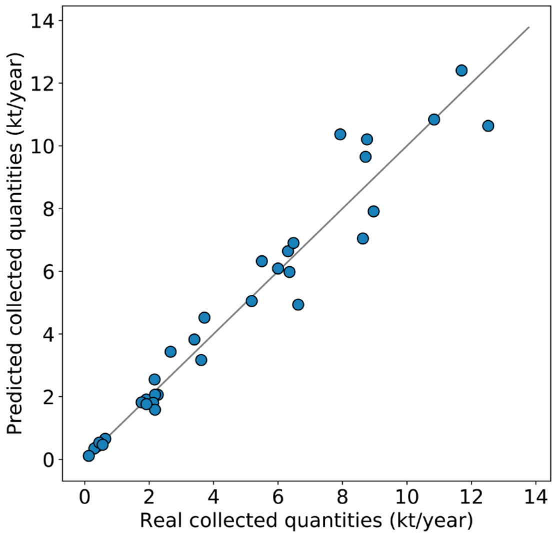

FIGURE 3: Comparison between predicted and real quantities of collected recyclables (MREC) for the UAM districts, based on the dwelling-type distribution.

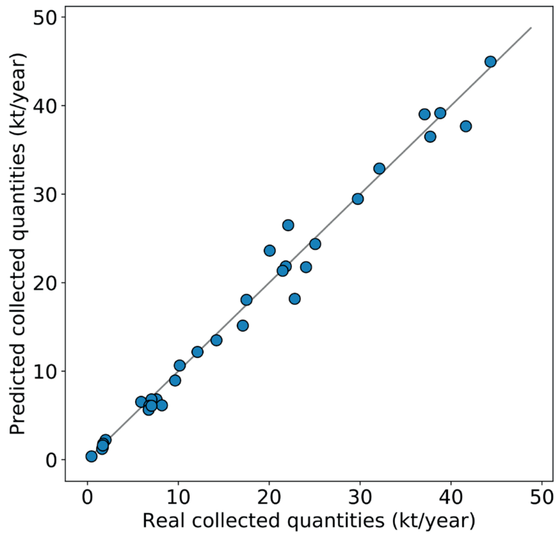

FIGURE 4: Comparison between predicted and real quantities of residual wastes (MRES + MORG) for the UAM districts, based on the dwelling-type distribution.

0.00317 h (11.4 s) and 0.00145 h (5.22 s) for α_{RES}^{single}, α_{RES}^{duplex}, and α_{RES}^{other}, respectively.

The comparison between predicted and real collection times is presented in Figure 5. In this Figure, fitted results using the total number of dwellings, with a unique average collection time per dwelling equal to 0.00272 h (9.8 s), are also presented. It is clear once again that the predictions using a model based on the dwelling-type distribution are more accurate (R^2 = 94.6%) than when only the number of dwellings is considered (R^2 = -14.6%). In this case, the influence is captured through the collection time, which is specific to each type of dwelling configuration. The parameters used in the models were adjusted for the Province of Quebec, Canada, but they could easily be adapted to other regional contexts.

The values of α_{RES}^{i} were then used to estimate residual waste collection times for 18 selected districts of the UAM for which data were available. Knowing the total annual collection distances and the annual numbers of residual waste collections for each district (see Table 4), it was possible, using the model developed in section 2.3, to calculate the average collection velocity and the collection cost for each of them. In a similar manner, knowing the transportation distance between each of the selected districts and the final destination, together with the average transportation velocity (see Table 4), it was possible to estimate the transportation cost. These costs are presented in Figure 6 and are reported on two specific bases: \$/dwelling and \$/t.

TABLE 3: Dwelling-type distribution and residual waste (RES) collection times for 12 collection sectors of the UAM.

Collection sector	n^{single}	n^{duplex}	n^{other}	[h]
LC-1	2835	1015	4295	32.6
LS-1	1215	4060	1925	25.0
LS-2	770	4985	1380	21.6
LS-3	865	3205	4230	24.1
LS-4	1100	5250	3070	31.3
MN-1	1115	1925	5640	25.2
MN-2	1895	1395	6135	27.0
MN-3	420	405	7150	15.5
LN-1	1970	690	9540	31.0
LN-2	535	2195	10850	26.1
SO-1	375	2245	5145	18.1
SO-2	690	3335	2350	20.0

Since the districts have a wide range of population and distances being covered for collection and transportation of the residual wastes, the specific costs vary greatly from one district to another. The collection costs vary between 8 \$/dwelling and 24 \$/dwelling (12 \$/t to 44 \$/t) and transportation costs vary between 2 \$/dwelling and 11 \$/dwelling (4 \$/t to 16 \$/t). These wide ranges of values confirm the importance of taking into account not only the distances for collection and transportation, but also the distribution of dwelling-types encountered during the collection. Overall, transportation costs are much lower than collection costs with a ratio of about 3 between the two in terms of \$/t.

3.4 Addition of a new collection and variation of the participation

So far, predictive models to estimate generated quantities of wastes for different collection streams and to estimate collection and transportation costs were presented. The model parameters were determined through a combination of fitted variables and reported values in the literature. Predictions from these models were compared to actual data from 33 districts having very different profiles (see Table 1) and results are very promising.

In this section, the models will be used as prospective tools to determine the impact of adding a new collection stream, a source-separated organic waste collection

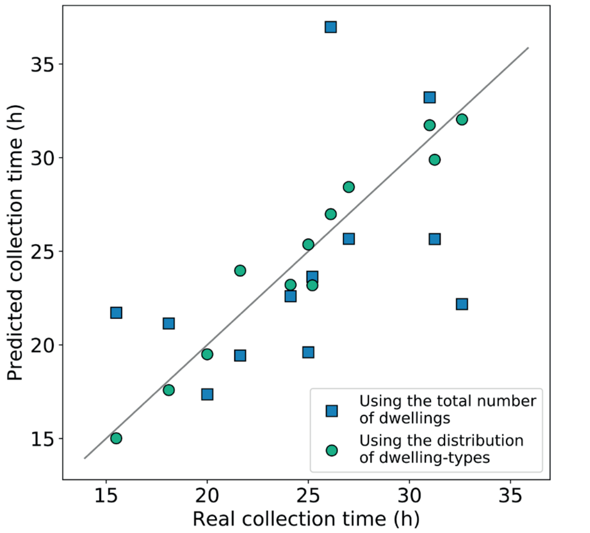

FIGURE 5: Comparison between predicted and real residual waste (RES) collection time for the UAM districts based on two approaches: (green circles) using the dwelling-type distribution; (blue squares) using the total number of dwellings.

(ORG), on the global diversion rate and collection cost. In order to simulate this new addition, the recovery rate τ^i_{ORG} was varied, as well as the percentage of dwelling having access to this new collection.

The simulation is based on the following assumptions:

- A source-separated organic waste collection stream is added in the 33 districts;
- The mass fraction of the total waste stream generated that should be placed in the organic waste collection stream (x^i_{ORG} is taken as reported values from the literature (see Table 2);
- The purity of this organic waste collected stream (π^i_{ORG}) is 100%;
- The recovery rate (τ^i_{ORG}), equivalent to the participation rate of the citizens to the new collection, is varied from 0 to 100%;
- The fraction of dwellings having access to this new collection stream (σ^i_{ORG} is modulated according to two scenarios: 1. all dwellings have access to the new collection (σ^i_{ORG} = 100% for all the dwellings); 2. Only single-family and duplex dwellings have access to the new collection and not the others (σ^i_{ORG} = 100% for single-family and duplex, σ^i_{ORG} = 0% for other).

TABLE 4: Characteristics of the 18 selected districts regarding the collection and transportation of the residual stream.

Territory	d^{RES}_{col} [km]	N^{RES}_{col} [-]	d^{RES}_{tran} [km]	v^{RES}_{tran} [km/h]
AC	696	104	14	51
AJ	304	104	19	68
CN	560	104	23	52
IS	237	52	26	51
LC	298	52	25	62
LN	386	78	9	60
LR	773	52	17	61
LS	403	104	27	44
MH	340	52	12	71
MN	372	104	24	64
MO	95	104	34	57
PM	309	104	18	46
PR	565	52	22	56
RP	484	52	15	65
SO	259	52	29	53
VD	239	52	30	51
VM	397	104	33	51
VS	509	104	12	60

Results are presented in Figure 7 in terms of the global diversion rate (Y, see equation 4) as a function of the participation rate for the two scenarios.

In Figure 7, the average value for each scenario is shown as a trend line, while the variability associated with the different districts is shown as a colored area. When all dwellings have access to the new organic waste collection, the global diversion rate is directly related to the participation rate with little variability amongst the different districts (blue area in Figure 7 is quite narrow). This is not the case when only single-family houses and duplexes have access to the new collection, in which case there is much wider differences in diversion rate between the different districts (wider orange area).

The cost models presented above were used to estimate the specific collection costs for the residual waste (RES) collection resulting from the addition of the new organic stream collection. The results are presented in Figure 8, where the specific costs in $/dwelling and $/t for the residual waste collection stream are plotted as a function of participation rate for the same two scenarios.

Figure 8 shows the impact of implementing a new collection stream (ORG) on the collection and transportation costs of an existing collection (RES). In this case, the specific costs for the residual collection, in $/dwelling and in $/t, are presented as a function of participation rate in the new organic waste collection. As expected, the collection costs of residual waste in $/dwelling is not affected by the presence of the new collection for both scenarios, since the trucks collecting the waste must travel the same distance as before (blue and orange lines on left graph). However, when the collection costs are expressed in $/t, since some of the waste is now diverted into the new organic collection, there is a strong dependency on the participation rate (blue and orange curves on right graph). For the transportation costs, they go down slightly in all cases since there are less waste to transport. It is interesting to note that the trend is reverse between the two scenarios, with the transportation cost in $/dwelling being more affected by the participation rate when all dwellings have access to the new collection, while the transportation cost in $/t is more affected by the participation rate in the case where only single-family houses and duplexes have access to the new collection. This type of behaviour is certainly very valuable information to take

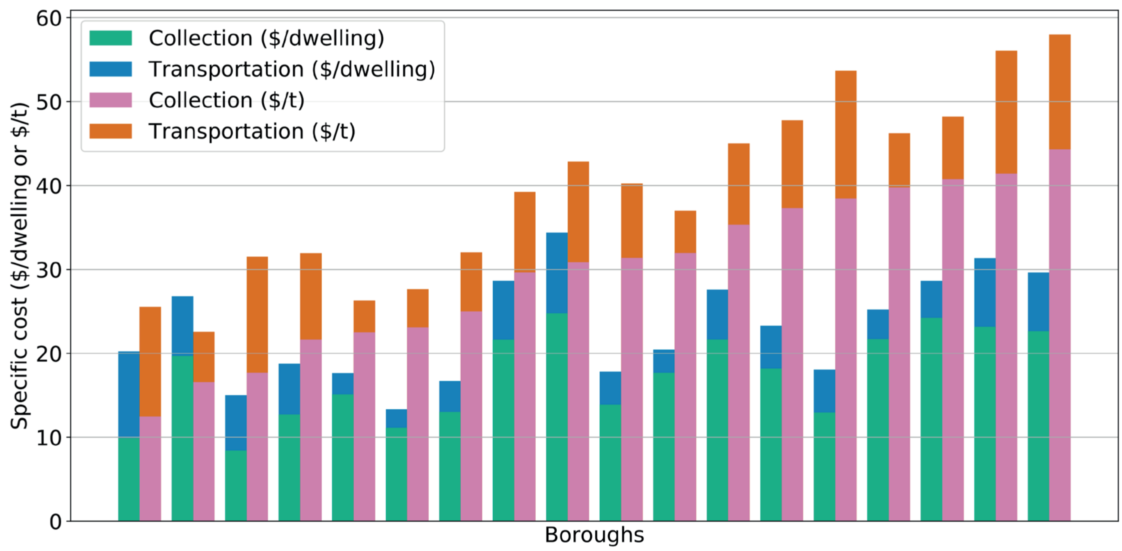

FIGURE 6: Specific collection and transportation costs per dwelling and per tonne as predicted by the model for the 18 selected districts of UAM.

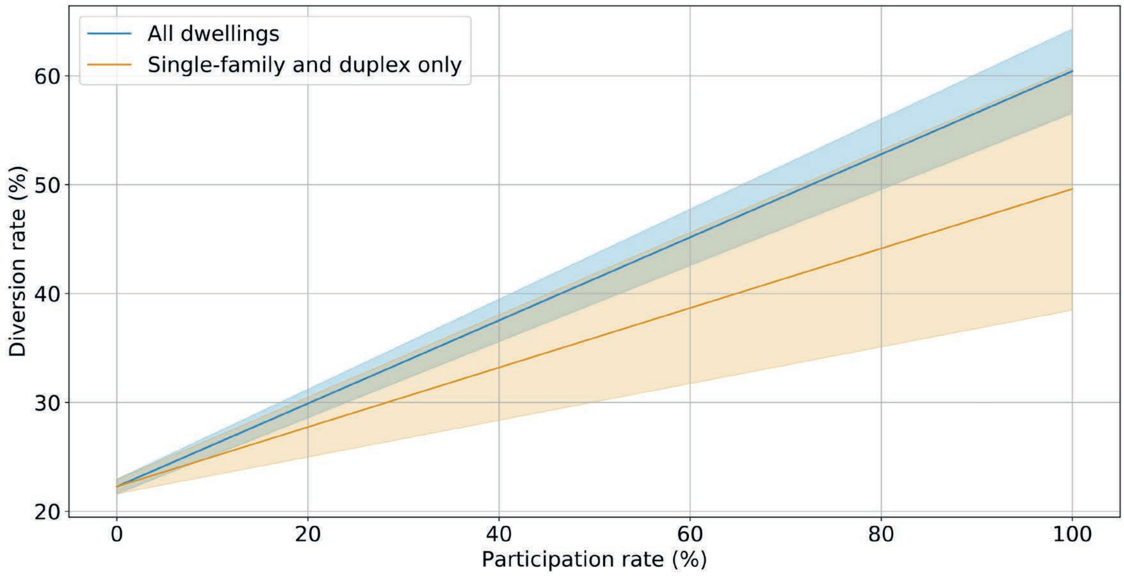

FIGURE 7: Global diversion rate as a function of the participation rate in a new collection stream of organic wastes (in blue: all the dwellings have access; in orange: only the single-family and the duplex dwellings have access).

into account when new collection streams are being evaluated.

4. CONCLUSIONS

As part of the development of decision-making tools by the CRVMR, a series of comprehensive models were developed to estimate waste generation and diversion, as well as collection/transportation costs. These models make use of the dwelling-type distribution encountered during the collection of different source-separated waste streams. They were calibrated and validated for widely different profiles of districts in terms of scales of population and dwelling characteristics. Using this approach, we were able to capture the effect of the dwelling-type distribution on the generation and diversion of wastes. This led to better predictive models and in turn, to better estimates of collection and transportation costs. The models were then used to simulate the effect of adding a new organic waste collection, with different participation rate of the population to this new collection. Prediction results revealed complex interactions. The model results give decision-makers very useful information in several of their tasks, such as allocation of collection contracts, estimation of scale for waste treatment facility and implementation of waste-related incentives.

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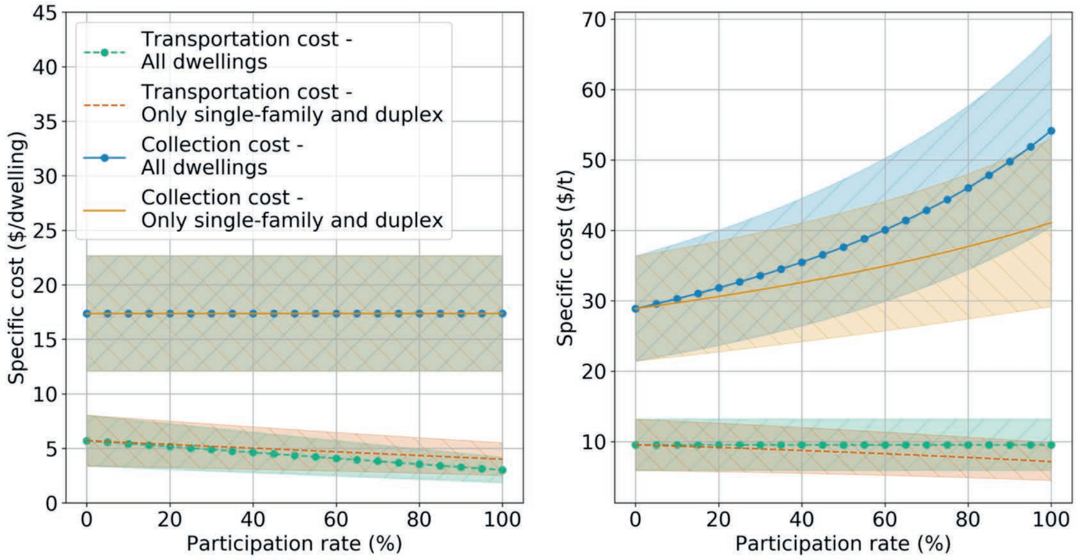

FIGURE 8: Specific costs of the residual waste (RES) collection as a function of the participation rate in a new collection of organic wastes (in blue: all the dwellings have access; in orange: only the single-family and the duplex dwellings have access).

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THE CIRCULAR ECONOMY PACKAGE OF THE EUROPEAN UNION: ARE NEW PATHS BEING TAKEN OR IS IT AN OLD STORY?

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ABSTRACT

The European Commission is in the process of improving its waste management and as a result, the so-called Circular Economy Package (CEP) has been launched. As a matter of fact, only recently several directives in the field of waste management have been amended and in the next years targets for re-use and recycling of waste will be significantly tightened. However, the CEP aims to go beyond recycling and issues such as easy-to-repair design or new business models are put in the foreground. It is striking that some ideas of the CEP are already more than four decades old. Nevertheless, the CEP has to be welcomed under the motto “better late than not at all”.

1. INTRODUCTION

In the European Union, waste management has changed dramatically in recent years (Pomberger, Sarc, & Lorber, 2017). In the year 1995, only 25·10^3 t of municipal solid waste (MSW) have been recycled in the EU which corresponded to about 11 % of the total quantity of 227·103 t (eurostat, 2019). The most current dataset of 2017 shows that the total amount of MSW increased by 10 % to 249·10^3 t but the amount of recycled waste has almost quadrupled (115·10^3 t) which means that the recycling rate is as high as 46 % (eurostat, 2019).

In order to further promote recycling and waste prevention the European commission has presented a so-called circular economy package (European Commission, 2015). The intention is not only to save the environment but to create new jobs, boost the economy and reduce the dependency on scarce resources. The project seems almost ingenious as the interests of environmental, the economic and social aspects can be aligned. The question is whether the plan is realistic or whether it is not worth the paper it is written on.

2. THE CIRCULAR ECONOMY

2.1 The genesis of the circular economy

Circular Economy is a concept that has spread virally in recent years. Based on a search in the database SciFinder® (ACS, 2019) the term is quite new as its first evidence in the scientific literature was not before 2003 (Xue et al., 2003). In the “early stage” circular economy was dominated by China. In the period from 2003 to 2007 SciFinder® reports 29 entries of which 26 are in Chinese language (ACS, 2019). The only paper in English including a Digital Object Identifier (DOI) was published in 2007 (Peters et al., 2007), but it also deals with China. Basically, Circular Economy seems to be a Chinese “invention”.

The first paper on Waste Prevention as important goal in modern waste management has already been mentioned much earlier in 1906 (Phelps, 1906) followed by Resource Management in 1965 (Fitzpatrick & Heller, 1965) and Zero Waste in 1975 (Milios, 1975; Wang & Yang, 1975) according to SciFinder®.

The chart in Figure 1 shows that in 2008 the terms Circular Economy (19 entries), Zero Waste (18 entries) and Waste Prevention (5 entries) lag far behind Resource Management (212 entries). In the last decade the usage of Circular Economy tremendously increased, in particular since the year 2015. As a result, in the year 2018 the usage of Resource Management (503 entries) and Circular Economy (443 entries) is almost equal. In the year 2019 (on the reporting date 13 November 2019) Circular Economy could take the lead with 662 entries over Resource Management with 479 entries. Even though Circular Economy was a latecomer, the term has become indispensable in today’s waste management.

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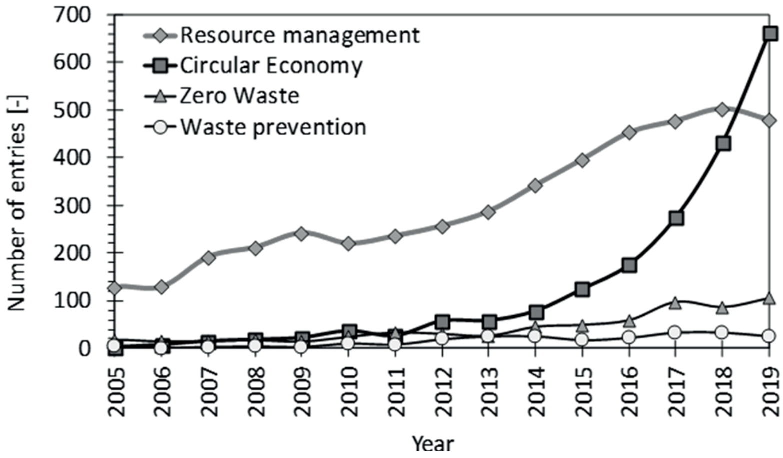

FIGURE 1: Number of entries in the database SciFinder® for the research topic "Resource Management", "Circular Economy", "Zero Waste" and "Waste Prevention" between 2005 and 2019; the entries of 2019 are based on the reporting date 13 November 2019.

2.2 The EU perspective on the circular economy

It was not until the 1970's before sound waste management became an issue in the European Union. As a reaction on environmental problems associated with waste disposal the first directive on waste was enacted in 1975 (European Communities, 1975). It was amended in 1991 (European Communities, 1991), 1996 (European Communities, 1996) and 2006 (European Parliament, 2006) and replaced by the Waste Framework Directive (WFD) in 2008 (European Parliament, 2008). The WFD introduced the waste hierarchy with waste prevention as upmost goal.

On 2 July 2014 the European Commission presented a proposal to amend some directives in the field of waste management (European Commission, 2014). Later, on 16 December 2014 the proposal was withdrawn by the new Commission (FUSIONS, 2014) and replaced by a new, "even more ambitious", suggestion which was released on 2 December 2015 (European Commission, 2015). Finally, the Directive (EU) 2018/851 (European Parliament, 2018a) has been passed in 2018 amending the WFD. As a matter of fact the "circular economy" must be implemented in all EU member states by 2020.

2.3 The issue of growth

It is the goal of most governments to create framework conditions that enable a high growth of the economy. Economists claim that the gross domestic product (GDP) should show a growth ideally between 2 and 3 % (Amadeo, 2019). Below or above this interval, economy is not healthy and problems will occur.

Our current economic system is dependent on growth. As shown by Strunz and Schindler (2018) there exist barriers to reach a post-growth economy. In particular, the authors investigated the following case studies:

- Economic growth is essential to keep unemployment on a low level. Up to now policies of reducing working time to translating only future productivity gains into leisure have not be successful.
- Alternative indicators to GDP have not succeeded in replacing GDP even if alternative measures such as "green GDP" are available (Stjepanović et al. 2017). Only recently the disadvantages of the indicator GDP have been clearly demonstrated (Brynjolfsson & Collis, 2019; Kapoor & Debroy, 2019).
- Pension systems depend on economic growth to compensate for demographic change. In an elaborate study Alda (2017) shows that population growth is essential that pension funds show a positive effect on stock markets independent on the age of the population.

The growth and the doubling rate are logarithmically related and the doubling rate can be calculated by the formulae shown in Equation 1 and Equation 2. This relationship is further demonstrated in Figure 2. Even though a growth of 2 or 3 % seems quite moderate it means that the doubling time is 23 or 35 year, respectively.

The situation is further outlined in Figure 3. Starting at a value of 1 (e.g. GDP, population, waste generation, etc.) an exponential growth occurs. After a period of 50 years, which lies within the time frame of human life, the value has increased 2.7 times (2 % growth) or 4.4 times (3 % growth). The situation is worse when considering a growth rate of 5 % which means that after 50 years the starting value has increased 11.5 times. Compared to China a growth of 5 % is very moderate as its economy has shown an even higher increase (IMF, 2019). Figure 4 plots the growth rates of China's GDP (from 2011 to forecasts until 2023) which averages at 6.88 %. A growth of 6.88 % means a doubling time of 10.4 years only (Figure 2) and will result in a 28-fold increase over 50 years (Figure 3). While many people will welcome such an economic growth, it is clear that a 28-fold generation of waste, GHG emissions or energy consumption is unacceptable for the environment.

$$q=1+P/100 \tag{1}$$

where:
q Growth rate [-]; P Growth [%], e.g. GDP, waste, etc.

$$t_d= \ln(2)/\ln(q) \tag{2}$$

where:
t_d Doubling time [a]

$$v = q^t \quad (3)$$

where:
v value [a.u.], e.g. GDP, waste, etc.; t time [a]

A tree is a perennial plant which can reach considerable heights even up to 120 m (Koch et al., 2004). It is reported that a Karri tree (Eucalyptus diversicolor) can grow quite fast and reaches a height up to 60 m after 60 years (Rayner, 1991). However, it will not grow up to the sky. The growth speed will significantly slow down and finally it will stop growing at an age of about 100 years (Rayner, 1991) even if it can get much older.

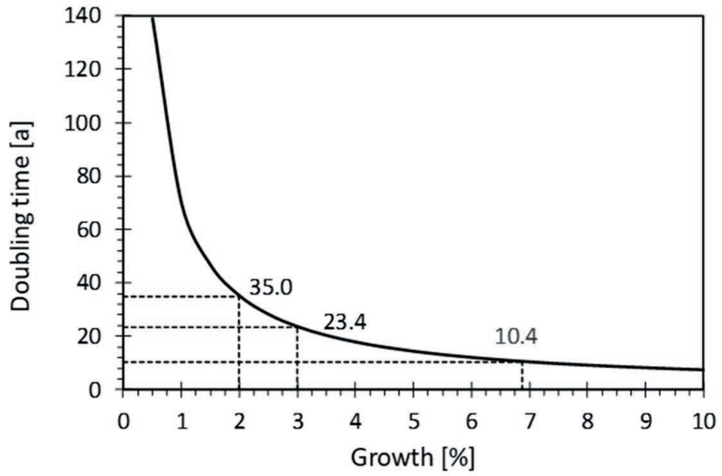

FIGURE 2: Doubling time as a function of growth (e.g. GDP, waste, GHG emissions, etc.) as calculated by Equation 1 and Equation 2.

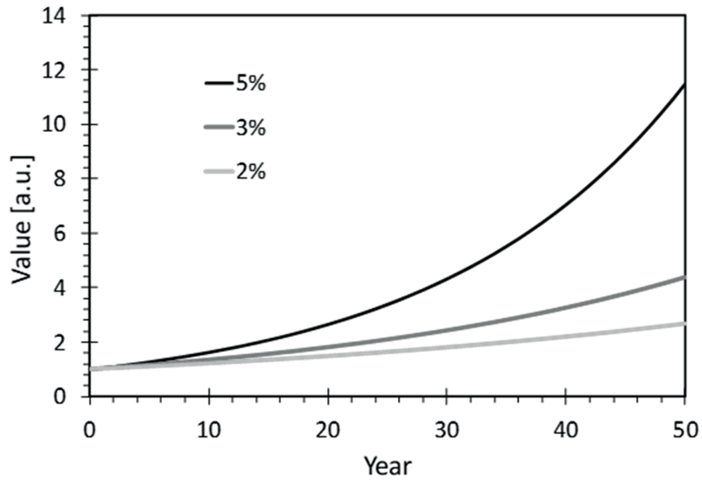

FIGURE 3: Exponential growth with 2, 3, 5 and 6.88 % over 50 years; starting at 1 in the year 0 calculated according to Equation 3.

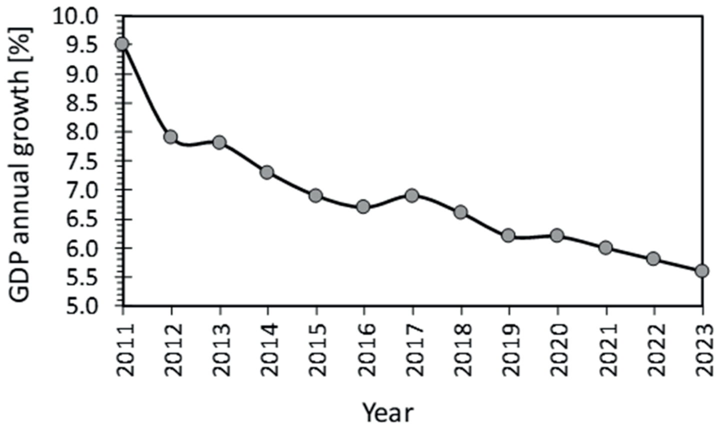

FIGURE 4: Annual growth rate of China's GDP from 2011 to 2017 and forecasts until 2023 (IMF, 2019).

The question is justified, why our economy is condemned to always grow. A sustainable system will grow, but only to a certain point. As exemplarily shown for a tree, ecosystems on our globe move towards an equilibrium state.

2.4 The limits to growth

The problem of exponential growth (Figure 2) was already addressed in 1972 by the famous book "The Limits to Growth" (Meadows et al., 1972). The authors concluded that the limits to growth on earth would become evident by 2072 if business was continued as usual. Without appropriate measures, in 2072 a "*sudden and uncontrollable decline in both population and industrial capacity will*" (Meadows et al., 1972) occur. The authors demanded that economy and population must reach an equilibrium state and any growth must be prohibited. Basically, the growth of population and economy must follow the example of a tree which stops growth at a certain height.

3. CIRCULAR ECONOMY PACKAGE VS. THE LIMITS TO GROWTH

It is striking that some of the key points of the circular economy package are already anticipated in "The Limits to Growth". Meadows et al. (1972) mention in Chapter "V The State of Global Equilibrium" some examples of technological advance that are required even if an equilibrium state (i.e. no growth of population or economy) is achieved.

A. "new methods of waste collection, to decrease pollution and make discarded material available for recycling"
B. "more efficient techniques of recycling, to reduce rates of resource depletion"
C. "better product design to increase product lifetime and promote easy repair, so that the capital depreciation rate would be minimized"

The sentence (A) sounds very current. Indeed, in Chapter 3. "Waste management" of the Circular Economy Package (European Commission, 2015) the importance of waste collection as an enabler for recycling is pointed out:

- […] The way we collect and manage our waste can lead either to high rates of recycling and to valuable materials finding their way back into the economy, or to an inefficient system where most recyclable waste ends in landfills or is incinerated, with potentially harmful environmental impacts and significant economic losses. […]

The importance of waste collection is further emphasized in the new Directive (EU) 2018/852 (European Parliament, 2018b), which was enacted as a results of the CEP and is amending the Directive 94/62/EC (European Parliament, 1994) on packaging and packaging waste. The revised directive introduces stringent targets for re-use and recycling packaging waste. However, in the introduction in paragraph 20 and and in the revised article 7 under paragraph 1(a) one can find the following sentences:

- […] Effective extended producer responsibility schemes can have a positive environmental impact by reducing the generation of packaging waste and increasing its separate collection and recycling […].
- […] the return and/or collection of used packaging and/or packaging waste from the consumer, other final user, or from the waste stream in order to channel it to the most appropriate waste management alternatives […].

The ideas of Meadows et al. (Meadows et al., 1972) and the Directive (EU) 2018/851 (European Parliament, 2018) are strikingly similar as both define waste collection as a key element for recycling. In 1972 the separate collection of waste was still in its infancy and it was not clear what "*new methods of waste collection*" will be. The "*new methods of waste collection*" have not yet been known, but today in many countries extended produced responsibility (EPR) is the core of legislation and policy to deal with end-of-life products, in particular packaging (Gupt & Sahay, 2015). EPR is the main driver for a separate collection of waste and seems to be the "new method" as demanded by Meadows et al. (1972). However, in many EU countries even today separate collection and recycling is on a very low level. In 5 EU countries, Malta, Greece, Cyprus, Croatia and Romania, more than 70 % of municipal waste is disposed of (landfilling or incineration D10). It seems that the proposals of Meadows et al. (Meadows et al., 1972) have gone unheard for four decades.

Also suggestion (B) can be found in the Circular Economy Package (European Commission, 2015) in a quite similar way. In chapter 4 - From waste to resources: boosting the market for secondary raw materials and water reuse, 1st paragraph and in chater 6 - Innovation, investment, and other horizontal measures, 2nd paragraph one can read:

- In a circular economy, materials that can be recycled are injected back into the economy as new raw materials thus increasing the security of supply;
- We will need new technologies, processes, services and business models.

The message of both is quite similar. Recycling is assigned an important role and it is the key element to reduce the demand for primary resources. According to Meadows et al. (Meadows et al., 1972) the main reason for recycling is the environment and the reduction of the depletion of limited resources. In contrast, the CEP sees the advantage of recycling in securing the supply of raw materials and boosting the economy (European Commission, 2015).

Finally, suggestion (C) can be found in a similar way in the CEP (European Commission, 2015) in Chapter 1.1 – Product design, first paragraph, in Chapter 2 – Consumption 5th paragraph and 1 Chapter 5.3. - Critical raw materials, 2nd paragraph:

- Better design can make products more durable or easier to repair;
- Once a product has been purchased, its lifetime can be extended through reuse and repair, hence avoiding wastage;
- Improve the recyclability of electronic devices through product design.

The above-mentioned principles represent the core of the CEP. It is not about targets for re-use and recycling but it addresses the minimization of the quantity of waste. It is striking that the ideas are more than 40 years old, but still today there are no appropriate measures to get waste prevention off the ground. In contrast, products such as electronic devices or apparel are getting increasingly cheaper. The useful life is more and more reduced and, even worse, items are becoming disposable goods. It has to be welcomed that the EU commission has addressed the right targets but it has to be questioned how our society can get there. Producers and retailers are interested to increase sales and turnaround. Policies such as a long service life or easy-to-repair design are often a mere lip service and do not take place in practice.

4. CONCLUSIONS

The CEP (European Commission, 2015) does indeed seem to be an "old hat". Some of the most important elements were formulated more than four decades ago by Meadows et al. (1972). Nonetheless, the CEP is still up to date because there is still a lack of substantial and consistent implementation of these ideas. In particular, the proposals of the CEP that go beyond the mere fixing of quotas are to be welcomed.

However, it is not clearly stated in the CEP how some of these requirements can be implemented in practice. Even if an increase of product lifetime and an easy-to-repair design are very efficient measures to reduce waste, manufacturers and retailers will continue to increase the sales. However, it is the first step in the right direction and it is to be hoped that the time is now ripe to put the concepts of waste prevention into practice.

Even though the concepts of the CEP (European Commission, 2015) and "the limits to growth" (Meadows et al., 1972) are similar to a certain extent, a clear difference is discernible. According to Meadows et al. (1972) it is absolutely necessary to achieve an equilibrium state in which growth has come to a standstill which is, in particular, outlined for population and economy. However, the European commission sees the CEP as a tool to boost the European economy and increase growth. According to Meadows et al. economic growth would wipe out all technological advances.

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WHY DO (W)EEE HOARD? THE EFFECT OF CONSUMER BEHAVIOUR ON THE RELEASE OF HOME ENTERTAINMENT PRODUCTS INTO THE CIRCULAR ECONOMY

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ABSTRACT

Waste electrical and electronic equipment (WEEE) continues to grow due to rising consumer demand for technologically advanced and affordable electronic products. Home entertainment (HE) products are particularly rich in metals and plastics and thus have enormous potential as a source of materials from the anthroposphere, especially from within Distinct Urban Mines (DUMs). Consumers' end-of-life (EoL) management decisions (i.e. stockpiling, hoarding, reusing, discarding of WEEE) strongly influence the exploitation potential of a DUM. This study aimed to assess the effect of consumer behaviour on the release of HE (W)EEE into the circular economy. A survey was undertaken in Southampton (Hampshire, UK) to assess perceptions and behaviours relating to the EoL management of HE (W)EEE. The study provides previously unavailable data and critical evaluation on the ownership, use and hoarding levels of HE EEE in a typical city DUM, and the reasons behind their hoarding. Results indicated that ownership levels were very high, with an average of 12 home entertainment items owned per household. This makes urban areas extremely plausible as DUMs; we estimate that there are over 1 million HE devices owned and ~440,000 HE devices hoarded in Southampton and >150 million HE EEE owned and ~61 million HE devices hoarded in UK households. Hoarding is common, especially for smaller or older equipment, due to their perceived residual value. HE product lifecycles averaged 4-5 years. The most common EoL routes were donating to relatives, friends or charities; hoarding; recycling via Household Waste Recycling Centres; or discarding items in general refuse. To encourage the recovery of EoL HE equipment in a DUM: i) convenient and accessible WEEE collection points should be established for regular (periodic) harvesting and ii) promoted via awareness campaigns and incentives.

1. INTRODUCTION

1.1 Hoarding of WEEE

The rapid technological advancements of the past decades, a growing market of affordable Electrical and Electronic Equipment (EEE) and shorter product lifespans have resulted in huge quantities of electronic waste (e-waste) being produced globally (Imran et al., 2017; Ongondo & Williams, 2012). At present, e-waste (also known as waste electrical and electronic equipment, WEEE) is one of the fastest-growing waste streams in the world, with 40-50 million tonnes being produced globally every year at an annual growth rate of 4-5% (Golev et al., 2016). WEEE poses an unavoidable waste management challenge for both developed and developing countries. Its huge global quantities, impacts on natural resources, as well as the potential human health, environmental and ethical concerns associated with its end-of-life (EoL) management if treated through rudimentary means (i.e. 'backyard recycling'), make the sustainable management of WEEE a global issue (Baldé et al., 2015; Hursthouse et al., 2017; Lodhia et al., 2017). EEE come to their EoL when they cease to function or be of any value to their owners, at which point they become WEEE (Ongondo & Williams, 2011a).

Market forces (e.g. technological advances or fashion), consumer behaviour and product features such as their material composition, condition, or reusability stimulate the generation of WEEE in throwaway societies (van Barneveld et al., 2016; Benton, 2015). Promoting a slower rate of consumption and the reuse of EEE purchases can

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help alleviate current challenges associated with WEEE production. Indeed, reuse of products has come into fashion again (Williams & Shaw, 2017) with the emergence of the so-called 'sharing economy', in which underutilized possessions are shared (or re-sold) through peer-to-peer exchanges, enabling more efficient use of products alongside economic and social benefits (Fletcher & Dunk, 2018; Cherry and Pidgeon, 2018; Martin, 2016). In the United Kingdom, the charity retail sector is becoming an increasingly significant player in terms of demonstrating the benefits of reuse and how it can be practically realized (Osterley & Williams, 2019). Under a sustainable closed-loop circular economy, resources within WEEE such as plastics, glass and metals are recovered, ultimately reducing the need for further extraction of raw materials (Schluep et al., 2009; Ongondo & Williams, 2012). Modern high-tech EEE such as home entertainment equipment are especially rich in metals and critical raw materials (e.g. platinum group metals and rare earth elements) and make up a large proportion of anthropogenic stocks (Massari & Ruberti, 2013; Golev et al., 2016; Williams, 2016). These secondary resources may be exploited via urban mining, helping the transition towards a circular economy (Izzat et al., 2014; Ongondo et al., 2015; Simon & Holm, 2018). Ideal locations for urban mining – known as Distinct Urban Mines (DUMs) - include urban hubs such as cities and universities, which have clear geographical boundaries, localised populations and quantifiable anthropogenic (W)EEE stocks as well as material flows (Pierron et al., 2017). Material stocks can be associated with in-use and hibernating (hoarded or stockpiled) (W)EEE in society, while material flows involve the reuse, recycling and discarding of EoL electronics (Pierron et al., 2017). To ensure the recovery of material stocks within a DUM, it is important to recognise potential hindrances to the circular flow of products and resources.

Poor recovery efforts for (W)EEE currently dictate that the majority of this waste stream is improperly disposed of, stockpiled or hoarded in society, or illegally exported to developing countries (van Barneveld et al., 2016; Imran et al., 2017; Pierron et al., 2017; Osibanjo & Nnorom, 2007). These factors generate serious environmental concerns, as they prevent the exploitation of a large proportion of material stocks, resulting in the extraction of additional natural resources to meet product demand (Ongondo & Williams, 2011a). In the EU alone, it is reported that from the 12 million tonnes of WEEE disposed of annually, only 30% is properly collected and recycled (Williams, 2016); with most small WEEE such as Universal Serial Bus (USB) sticks, mobile phones, electric toothbrushes, or lamps ending up in the general waste (Baldé et al., 2015). Consumer behaviours associated with their intent to sell, reuse, recycle, discard, or hoard (W)EEE determines the extent of an urban mine's exploitation.

Hoarding occurs when consumers indefinitely store obsolete EEE that are no longer used or wanted. This is a major barrier to accessing an urban mine and releasing exploitable materials into the CE. The average household in the United States of America (USA) hoards 4.1 small (>4.5kg) and 2.4 large electronic items in their attic or basement, mainly due to ample storage space, preventing their recycling or reuse (Saphores et al., 2009). In Europe and the United Kingdom (UK), hoarding is common for (W)EEE with a perceived residual value (e.g. monetary, functional, sentimental), such as mobile phones (Ongondo & Williams, 2011a). Access to an urban mine primarily depends on the availability of resources for a given type of (W)EEE. Hoarded items that are currently unavailable (hibernating stocks) greatly reduce the exploitation potential of anthropogenic resources since they are stored in homes indefinitely. Ideally, (W)EEE should be stockpiled/hoarded with the intent of releasing it into the CE instead; which will ensure future access to a DUM's unexploited resource stocks. To gain insight on a DUM and facilitate a closed loop system for resource and material flows, it is essential to quantify the ownership and use levels for (W)EEE as well as the EoL practices and dispositions of consumers.

The primary aim of this study was to assess the effect of EoL consumer behaviours on the release of home entertainment (HE) (W)EEE into the circular economy. The objectives were to: i) determine the technological advances in HE EEE through time by producing evolutionary timelines for audio, visual and interactive electronic devices; ii) identify and evaluate the types and quantities of HE (W)EEE that consumers own, use and hoard; iii) establish and analyse the reasons behind consumer hoarding; and iv) evaluate and critically discuss consumer purchasing, hoarding, gifting, selling and disposing behaviours for HE (W)EEE, to establish how they might be addressed to achieve a CE.

We critically discuss findings in the context of the technological and fashion-related evolution of HE electronic products, whilst considering urban areas as a potential DUM under the framework discussed by Pierron et al. (2017), using the city of Southampton as an exemplar.

1.2 Evolution of home entertainment technologies

Since their invention, HE systems in the form of audio devices (speakers and radios), televisions, video players and games consoles have revolutionised the market economy globally. Through the decades, the growing demand for the production of electronic goods within developed economies has been gradually reinforcing the principles of a "throwaway society", which are associated with rapid product replacement and high market competition (Williams, 2016). An analysis of the evolution of HE EEE from 1861 until the present day is depicted in Appendix 1, which documents and illustrates how audio speakers, televisions and video players, and game consoles have evolved from inception to the present day. Insight on the technological advancements and fashion-related evolution of such products contributes to the understanding of current HE EEE, the relationship between manufacturers and consumers and EoL behaviours.

2. METHODS

2.1 Postal survey

Primary data was collected from residents of the city of Southampton (Hampshire, UK) via a postal survey, during the course of 3 months in January-March 2018. The design of the survey built upon the approach used in previous studies (e.g. Bergland and Matti, 2006; Timlett and Williams, 2009, Ongondo & Williams, 2011b). The self-administered questionnaire (Appendix 2) incorporated questions to: (a) assess community perceptions, behaviours, and dispositions related to the EoL management of HE (W)EEE; (b) establish the types of HE (W)EEE consumers own, use, and stockpile/hoard in their homes; and (c) determine the reasons behind these tendencies. The questionnaire sought information on respondents' demographic characteristics, household size and storage space. Respondents' dispositions and behavioural tendencies were determined to distinguish between respondents' intent to act and past actions. The survey was permitted via the University of Southampton's standard procedures for risk assessment (code FEERA 15034) and ethical approval (code ERGO II 40143).

The questionnaire was piloted among twelve participants from the target population prior to data collection, to ensure ease-of-understanding, ease-of-use, and content validity. Questions were modified as a consequence of feedback received from the pilot survey. A participant information sheet was attached to the final questionnaire to provide simple instructions and to answer potential participant queries.

Seven hundred and twenty questionnaires were hand-delivered across Southampton households within four pre-selected wards. These locations were selected to ensure a representative sample incorporating all levels of household affluence, as determined by the UK's Index of Multiple Deprivation (IMD) of 2015 (OpenDataCommunities.org, 2015). The four wards represented levels of high (Redbridge ward: 10% most deprived), average-high (Swaythling ward: 30-40% most deprived), average-low (Bargate ward: 50% least deprived) and low (Upper Shirley ward: 10-20% least deprived) deprivation within the city.

One hundred and eighty questionnaires were posted in each ward across randomly selected households and collected in person a week after delivery. To encourage a higher response rate, three collection attempts were made per household. Questionnaires were coded by ward, and household numbers were documented separately to enable future collection attempts.

2.2 Statistical analysis

The results were analysed using SPSS 20 (IBM Ltd). Non-parametric tests were applied to examine hoarding habits and reasons behind such behaviour, and compare between hoarding reasons, home entertainment (W)EEE gifting, selling and disposal routes, as well as according to respondent demographics.

A chi-square test was carried out for association between age, gender, educational level, deprivation zone, household size, household storage space and hoarding habits (whether respondents hoarded or not). Z-scores from the chi-square cross-tabulation table were compared to examine the relationship of the significant associations.

Kruskal-Wallis tests were performed to assess for a significant difference between respondent demographic variables (age, gender, educational level, deprivation zone, household size, household storage space) and hoarding reasons (functional, social, sentimental, monetary and lack-of-awareness reasons). A series of post-hoc Mann-Whitney U tests were performed on the significant results, to examine where the significance lies within each demographic variable and hoarding reason.

A Friedman's analysis of variance test was performed to examine whether hoarding reasons differ significantly from one another. Wilcoxon signed-rank tests were applied to rank hoarding reasons in order of their influence on respondents' hoarding dispositions. A Friedman's analysis of variance test was also carried out to examine the significance between home entertainment (W)EEE gifting, selling and disposal routes. Wilcoxon signed-rank tests were subsequently applied to rank the gifting, selling and disposal routes in order of likelihood of opting for each route, as indicated by survey respondents.

3. RESULTS

A total of 139 useable questionnaires were collected (19% response rate). The results of the survey are presented in the following sections. Respondent behaviours (previous past actions; sections 3.1 and 3.2) and dispositions (intent to act; section 3.3) have been analysed separately, to differentiate between past consumer hoarding practices and behavioural tendencies. The results were broadly representative for each deprivation zone and gender, although more respondents from the younger age groups completed the survey than from the older (61% of respondents were 18-24 and 25-44 years, whereas only 12% of respondents were 65+ years).

3.1 Behaviours and practices

All respondents owned and used at least one HE EEE. Of those, approximately 75% hoarded home entertainment electronic products that they no longer wanted or used, 60% gifted, sold or disposed (GSD) of at least one HE electronic product since 2012, and 42% GSD of a HE electronic product since 2015.

Respondents owned more HE electronics than they used in every product category (Figure 1). Most respondents owned at least one TV (96%), audio device (89%), video player (62%), and Nintendo game console (62%). Of those, the most commonly owned products were Plasma TVs (81%), portable speakers (65%), Smart TVs (60%), radios (54%), DVD players (50%) and the Nintendo Wii (42%). Smart TVs had the highest use-to-ownership ratio. No respondents used their black and white TV. Overall, products that accompanied TV sets (e.g. video players, TiVo & set-top boxes, or audio devices) had higher ownership and use rates than most game consoles. The Nintendo Wii and DS had the highest game console ownership rates, yet they also had the smallest use rates. The Sony PlayStation 4 had the highest use-to-ownership ratio for game consoles. The HE categories with the lowest ownership rate were black and white TVs and virtual reality (VR) sets.

On average respondents owned 2.8 TV sets (1.6 Plasma TVs, 1 Smart TV), 1.4 video players, 1.1 TiVo and set-top boxes, 2.6 audio entertainment devices (1.1 portable speakers) and 0.2 game consoles, with a total of 11.8

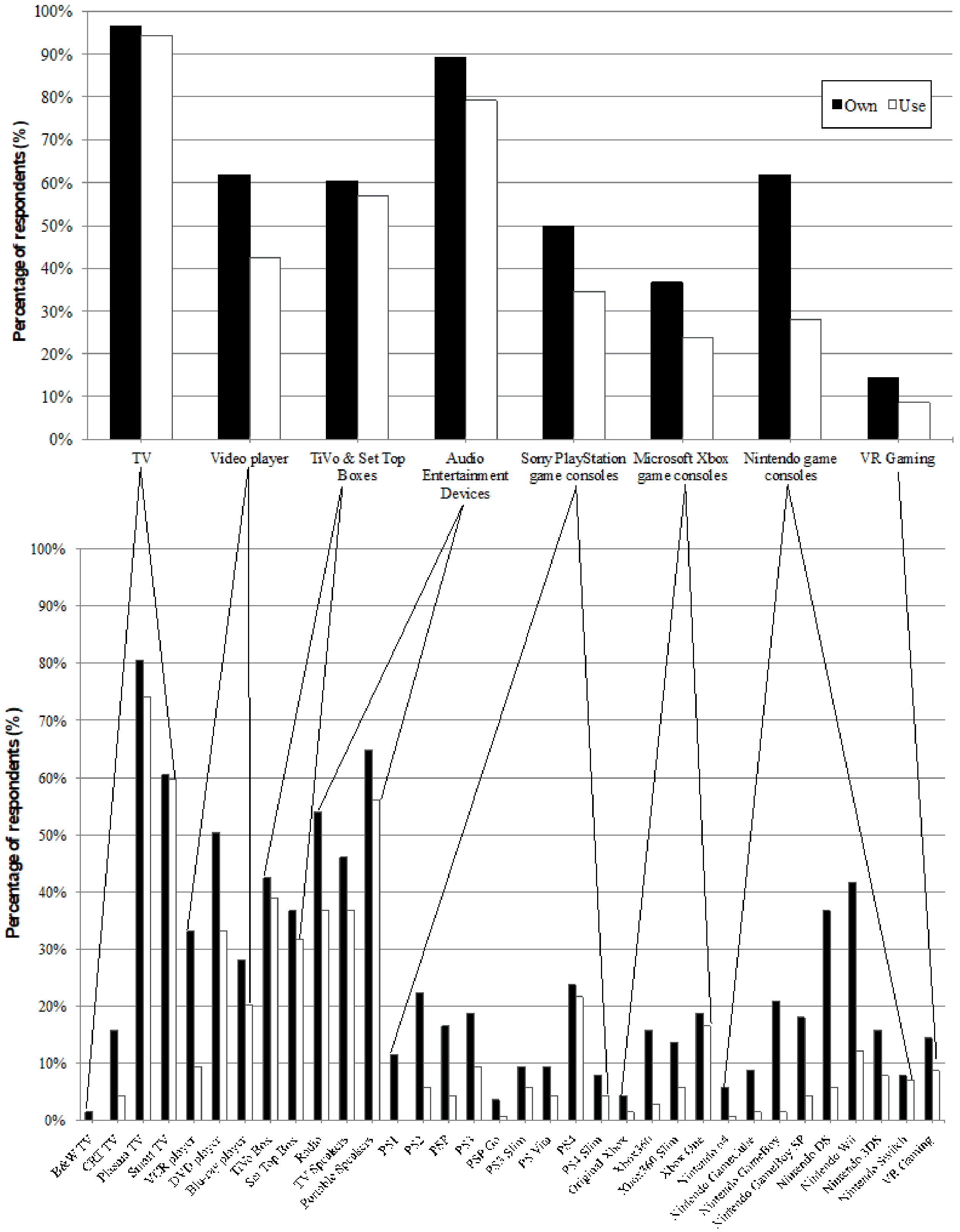

FIGURE 1: Proportion of respondents owning and using at least one home entertainment electronic product in Southampton in 2018 (n=139).

HE devices owned per household. The products with the highest proportion of multiple devices were plasma TVs (43%), portable speakers (27%), radios (20%) and smart TVs (19%). The game console with the highest proportion of multiple devices was the Nintendo DS (10%).

Almost all respondents (97%) purchased their HE EEE through an electronics retailer as brand new products, and approximately a quarter of respondents were gifted HE products. Only a small minority of respondents opted to purchase second-hand EEE (i.e. from eBay, CeX shop, second-hand shop - Table 1).

Since 2012, 81% of respondents had purchased new HE products in addition to what they already owned (not as replacements - Table 2), and 19% had only purchased new products to replace their old ones. The most common additionally purchased products were TVs (60%) and specifically smart TVs (37%), followed by Sony PlayStation game consoles (see Appendix 3 for a complete breakdown of additionally purchased products). Almost no respondents had purchased additional audio entertainment devices.

Most respondents (60%) reported not planning any HE EEE purchases for 2018. The rest indicated plans of purchasing new TVs (mostly smart TVs), Sony PlayStations (mostly PlayStation 5), VR gaming sets and Set-Top Boxes (Table 3).

Approximately 60% of respondents had gifted, sold or disposed (GSD) of at least one HE electronic item since 2012 (5 years ago) and 42% had GSD of items since 2015 (2 years ago). TVs were the most commonly GSD of devices in both instances, followed by audio entertainment devices and Nintendo game consoles (Figure 2).

Specifically, plasma TVs, radios, and Nintendo Wii consoles were the most frequently GSD of HE electronic products. The majority of respondents had gifted products to family and friends, taken them to Household Waste Recycling Centres (HWRCs), or thrown them out via general waste bin (Table 4). It was common for relatively newer models of EEE such as plasma TVs or PlayStation 3 consoles to be gifted to family or friends, whereas older products such black and white TVs, CRT TVs and VCR players to be taken to HWRCs.

The majority of respondents (39%) reported gifting, selling or disposing of new/relatively new HE EEE because they wanted a more up-to-date model (Table 5). Respond-

TABLE 1: Respondents' purchasing options for home entertainment electronics in Southampton in 2018 (n=139).

Purchasing Options	Number of respondents (N)	Proportion of respondents (%)
Electronics retailer	134	**96.4**
Second-hand shop	5	3.6
CeX shop	4	2.9
Gifted from family and friends	35	25.2
Bought from family and friends	13	9.4
eBay	1	0.7

TABLE 2: Home entertainment (HE) electronic products additionally purchased (not purchased as replacement products) by respondents since 2012 (n=138).

HE EEE Categories	Number of respondents (N)	Proportion of respondents (%)
TV	82	**59.4**
Video Player	11	8.0
TiVo & Set-Top Boxes	25	18.1
Audio Entertainment Devices	42	0.1
Sony PlayStation game consoles	28	20.3
Microsoft Xbox game consoles	15	10.9
Nintendo game consoles	16	11.6
VR Gaming	5	3.6
Other	3	2.2
No additional items bought (only replacements)	26	18.8

TABLE 3: Respondents' planned purchases for home entertainment (HE) electronic products in 2018 (n=139)products) by respondents since 2012 (n=138).

HE EEE Categories	Number of respondents (N)	Proportion of respondents (%)
TV	27	19.4
Video player	0	0.0
TiVo & Set-Top Boxes	11	7.9
Audio Entertainment	9	6
Game consoles & VR	26	18.7
Other	5	3.6
Not planning on purchasing anything	83	59.7

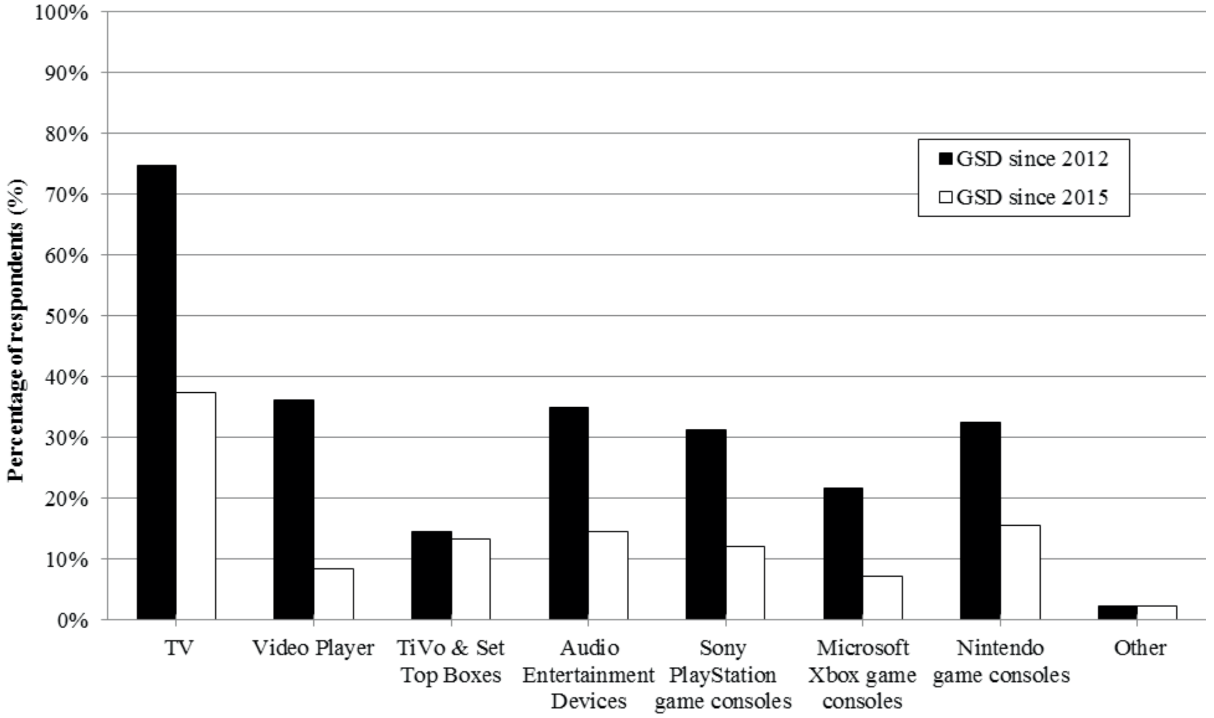

FIGURE 2: Proportion of respondents who gifted, sold or disposed (GSD) of home entertainment electronics of since 2012 and 2015.

ents also commonly disposed of HE products because they did not need them anymore.

3.2 Hoarding practices

Overall, 75% of respondents reported hoarding HE EEE, of which 55% and 32% (respectively) were also storing videotape and DVD products that they no longer used or wanted (Table 6). A high proportion of respondents hoarded game consoles and audio entertainment devices (Figure 3). The most commonly hoarded items were videotape products (42% of respondents hoarded at least one), Nintendo DS consoles (31%), Nintendo Wii consoles (29%), VCR Play-

TABLE 4: Gifting, selling and disposing routes for end-of-life (EoL) home entertainment (HE) electronics (n=83).

EoL routes of HE EEE	Number of respondents (N)	Proportion of respondents (%)
Sold online	6	7.2
Gave to family or friends	57	68.7
Sold to CeX shop	7	8.4
Threw out via general waste bin	14	16.9
Sold to family or friends	3	3.6
Gave to charities	10	12.0
Dumped away from house	1	1.2
Gave away via online free recycling	0	0.0
Took to HWRC	24	28.9
Left outside house	8	9.6
Threw out via recycling bin	12	14.5

TABLE 5: Reasons for gifting, selling and disposing of end-of-life home entertainment electronics (n=83).

Reasons	Number of respondents (N)	Proportion of respondents (%)
The item was dated and broken and needed replacing	17	20.5
The item was working properly, but I wanted a more up-to-date model	32	38.6
The item was new/relatively new but had stopped working and needed replacing	2	2.4
I was gifted a more up-to-date model	9	10.8
I decided I did not need it nor did I need a replacement	23	27.7

TABLE 6: Respondent responses on the ownership and use of videotape and DVD products (n=104).

Responses	Videotape products		DVD products	
	No. of respondents (N)	Proportion of respondents (%)	No. of respondents (N)	Proportion of respondents (%)
Yes, and I still use them	15	13.9	64	59.3
Yes, and I no longer want them	59	54.6	34	31.5
No, I have given them all away	34	31.5	10	9.3
Total	108	100.0	108	100.0

ers (24%), Nintendo GameBoy consoles (19%), DVD players (17%), PlayStation 2 consoles (17%) and radios (17%) (Figure 2). On average respondents hoarded 1.5 Nintendo consoles, 0.8 Sony PlayStation consoles and audio devices, and 0.6 TVs and video players, with a total of 4.8 HE devices hoarded per household. The products with the highest proportion of multiple hoarded devices were Nintendo DS consoles (9%), portable speakers (7%) and radios (7%).

The functional value ('I may need it someday') placed on HE EEE is the most important factor influencing respondent hoarding habits (Figure 4). Both sentimental ('I'm too attached to the item') and social ('One day I'll find someone to give it to, who will really want it') reasons for hoarding ranked as second most important influencing factors. Approximately 45% of respondents agreed that lack-of-awareness (LoA) of what else to do with the item was an important influencing factor for hoarding of HE electronics. Monetary reasons for hoarding ('I paid too much money just to get rid of it') were ranked the least important hoarding factor.

3.3 Dispositions, tendencies and opinions

The functional value ('I may need it someday') placed on HE EEE was the most important influencing factor for respondent hoarding tendencies (Figure 5). For hoarding tendencies, social reasons to hoard ('One day I'll find someone to give it to, who will really want it') were ranked higher than sentimental reasons ('I'm too attached to the item'). Lack-of-Awareness ('I don't know what else to do with it, so I keep it') was ranked third most important influencing factor alongside sentimental hoarding reasons. Monetary reasons were ranked as the least important influencing factor for respondent hoarding tendencies.

When gifted a new HE electronic product, the majority of respondents indicated that they would keep both the old and newly gifted products, signifying a tendency to hoard (Figure 6). More than half of respondents also reported that they would get rid of the old product once gifted a newer model. Hardly any respondents reported that they would keep the old product and dispose of the newly gifted model.

Similar to hoarding practices vs. tendencies, the gifting, selling and disposing tendencies of HE electronics were also examined to ensure responses from all participants (n=139), including the ones who hadn't previously gifted, sold or disposed of products (n=53). When examining respondent tendencies on the EoL management routes, the majority of respondents (86%) reported that they would opt

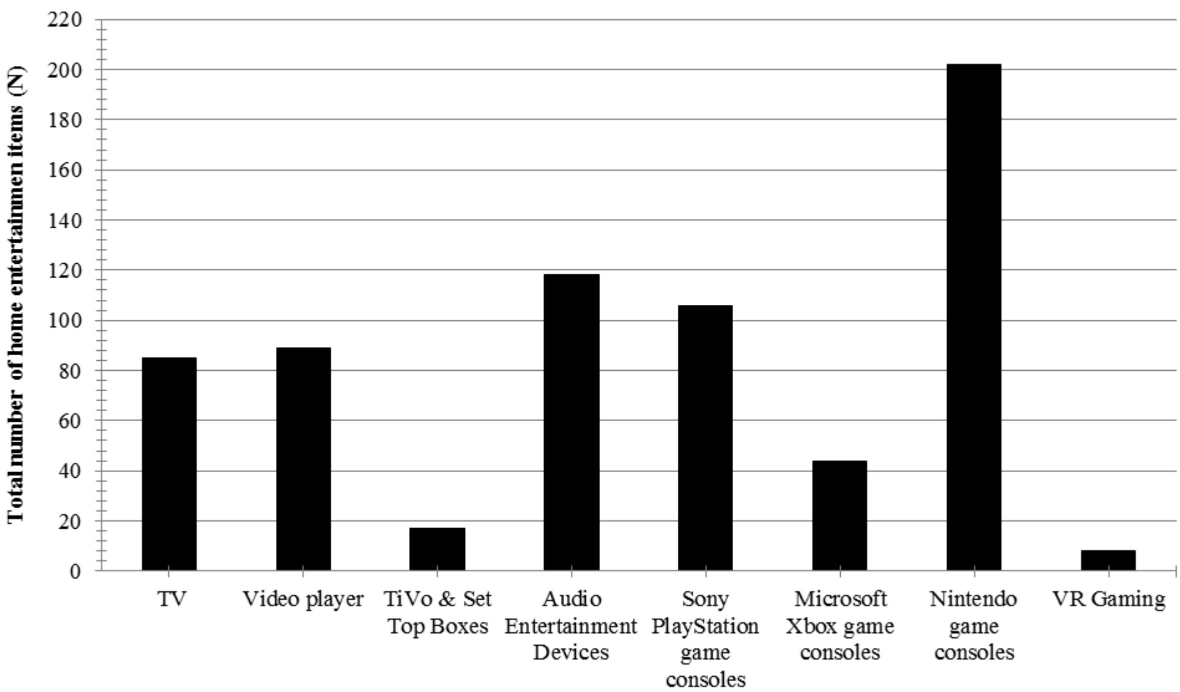

FIGURE 3: Total number of home entertainment items hoarded by respondents (n=104).

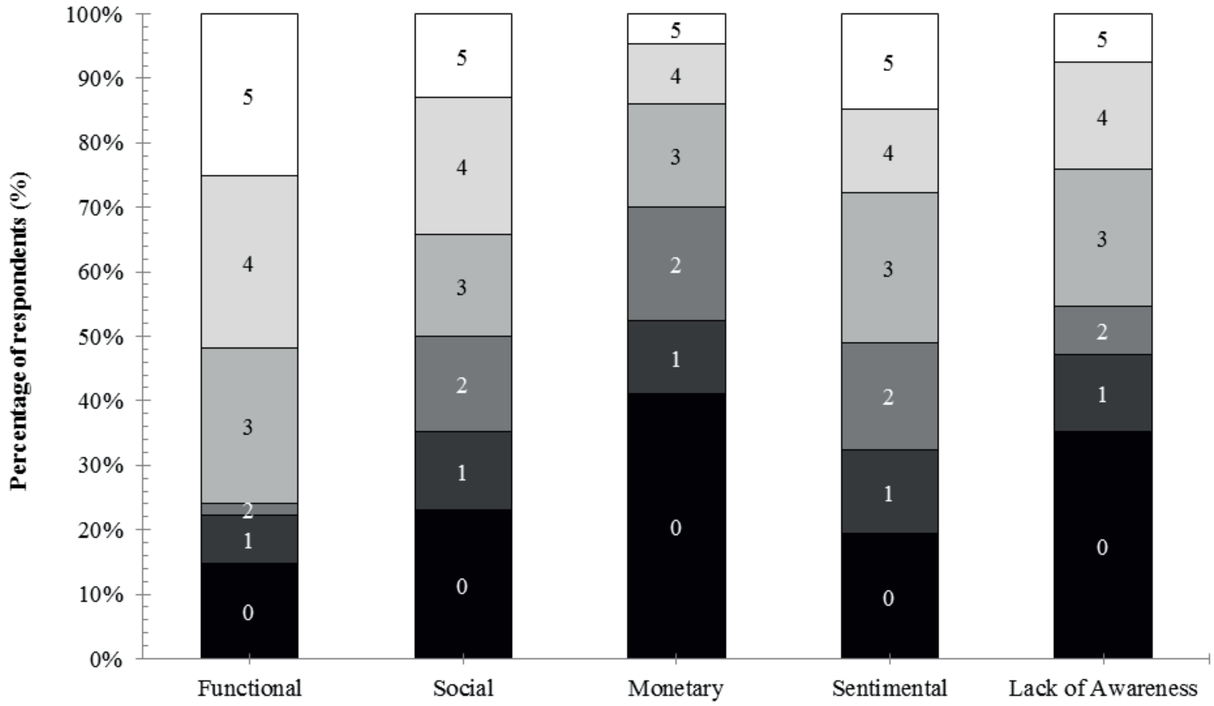

FIGURE 4: Reasons influencing respondent hoarding practices (Proportion of respondents who disagree: 0, to completely agree: 5, n=104).

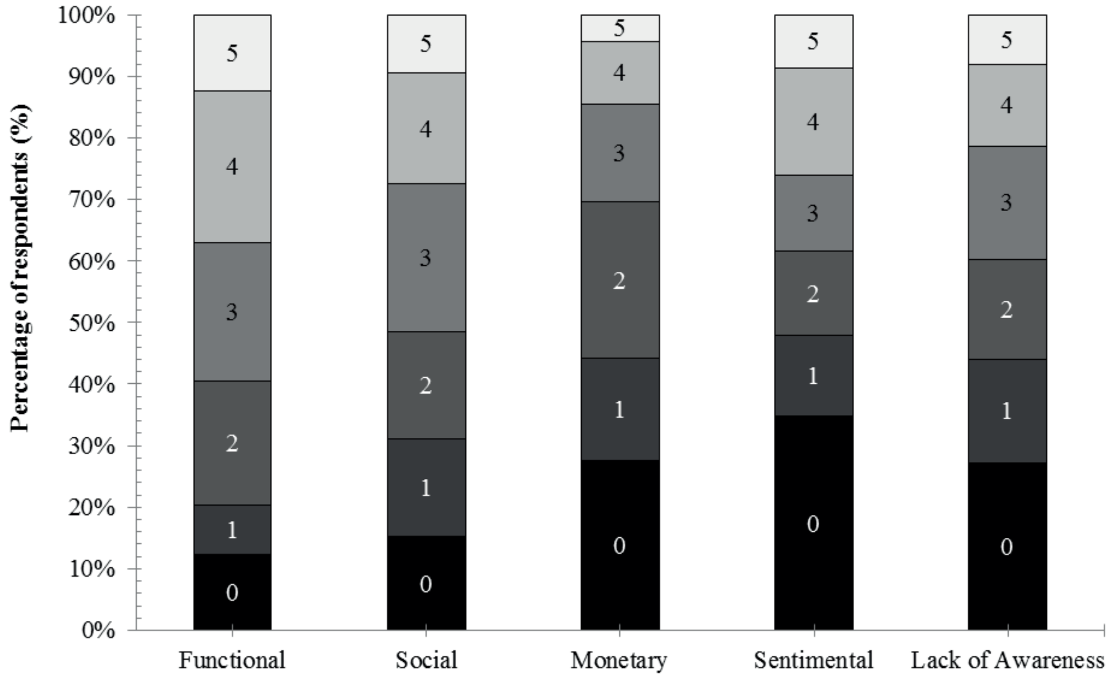

FIGURE 5: Reasons influencing respondent hoarding tendencies (Proportion of respondents who disagree: 0, to completely agree: 5, n=139).

to 'give away' their products, first to family and friends, and then to charities (Figure 7). Respondents would sell their unwanted EEE as their second most common EoL management option, with 49% opting to sell items online, 30% to friends and family and 23% to a reuse shop (e.g. CeX).

The third most common route for respondents' EoL management tendencies was recycling of WEEE, with approximately 38% opting to take them to a HWRC, 25% using online recycling and 20% throwing them out via recycling bin (Figure 7). Fewer respondents reported tendencies of throwing WEEE out via general waste bin, or leaving them outside/away from their house.

When it came to purchasing HE electronics, 74% of respondents agreed that they would wait for their EEE to stop working before purchasing a newer model; 61% agreed that they would wait for their EEE to become dated before mak-

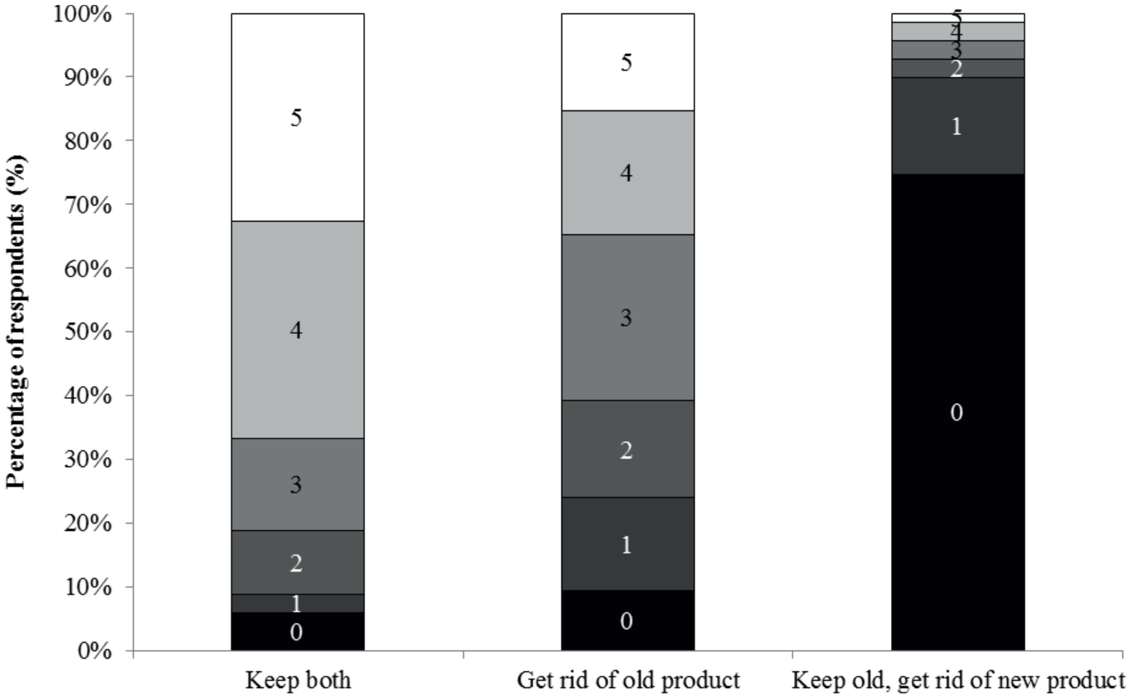

FIGURE 6: Respondent tendencies for the end-of-life management of their old home entertainment electronic products when gifted a newer model (Proportion of respondents who disagree: 0, to completely agree: 5, n=139).

ing a new purchase; and 37% agreed that they like to purchase the newest model of HE equipment that is currently on the market. Around 16% of respondents agreed that if they kept their HE EEE for long enough, their monetary value would increase. The majority of respondents (89%) would not dispose of their TV and only use their phone or tablet, even though 30% of them claimed to watch television via their phone or tablet more often than via their TV.

Based on replacement cycles, the most lifecycle of HE products was 4-5 years (Figure 8). No respondents believed that TVs and game consoles should be replaced every year, and just a small proportion (2.2%) would replace their products only once they stop working.

3.4 Demographic characteristics and hoarding

A statistically significant association was found between respondent age groups and hoarding (whether someone hoarded or no - Table 7) at the 0.05 level with a p-value of 0.019. Statistically fewer respondents than expected from the age group of 25-44 years hoarded, and statistically more respondents than expected from the age group of 45-65 years hoarded HE WEEE. No association was found between all other demographic variables (gender, deprivation level, educational level, household size and storage space) and hoarding habits.

A statistically significant difference was determined between hoarding reasons (i.e. functional, social, sentimental, monetary and lack of awareness) at the 0.05 level (DF=4, p-value<0.0005). It was determined that significantly more respondents considered functional reasons ("I may need it someday") as important influencing factors for hoarding when compared to all other hoarding reasons (Table 8). Fewer respondents rated monetary reasons as important influencing factors for the hoarding of HE EEE, habits when compared to functional, social and sentimental reasons.

A statistically significant difference was found between respondent's age group and two of the five hoarding reasons, namely monetary and lack-of-awareness (LoA) reasons (Table 9). Significantly more respondents from the youngest age group (18-24 years) considered LoA as an important influencing factor for hoarding, compared to all other age groups (Table 10). Similarly, significantly more respondents from the younger age groups (18-24 and 25-44 years) agreed that monetary reasons influenced their hoarding of HE (W)EEE when compared to the 65+ age group. No significant difference was found between all other demographic variables and hoarding reasons.

3.5 End-of-life routes

A significant difference was found between the EoL management routes of gifting, selling and disposing at the 0.05 level (DF=10, p<0.0005). Significantly more respondents would opt for giving to family and friends as their primary EoL management route. Similarly, more respondents would choose for giving to charities, selling online and taking HE (W)EEE to the HWRC as their second, third and fourth EoL management routes. Overall, no significant difference was found between all other routes (e.g. selling to CeX, throwing out via recycling bin, etc.).

4. DISCUSSION

4.1 Evolution of HE electronics

HE electronics have evolved from analogue to digital, to 'smart' and interactive technologies. The timelines (Appendix 1) show that although consumer-based audio and

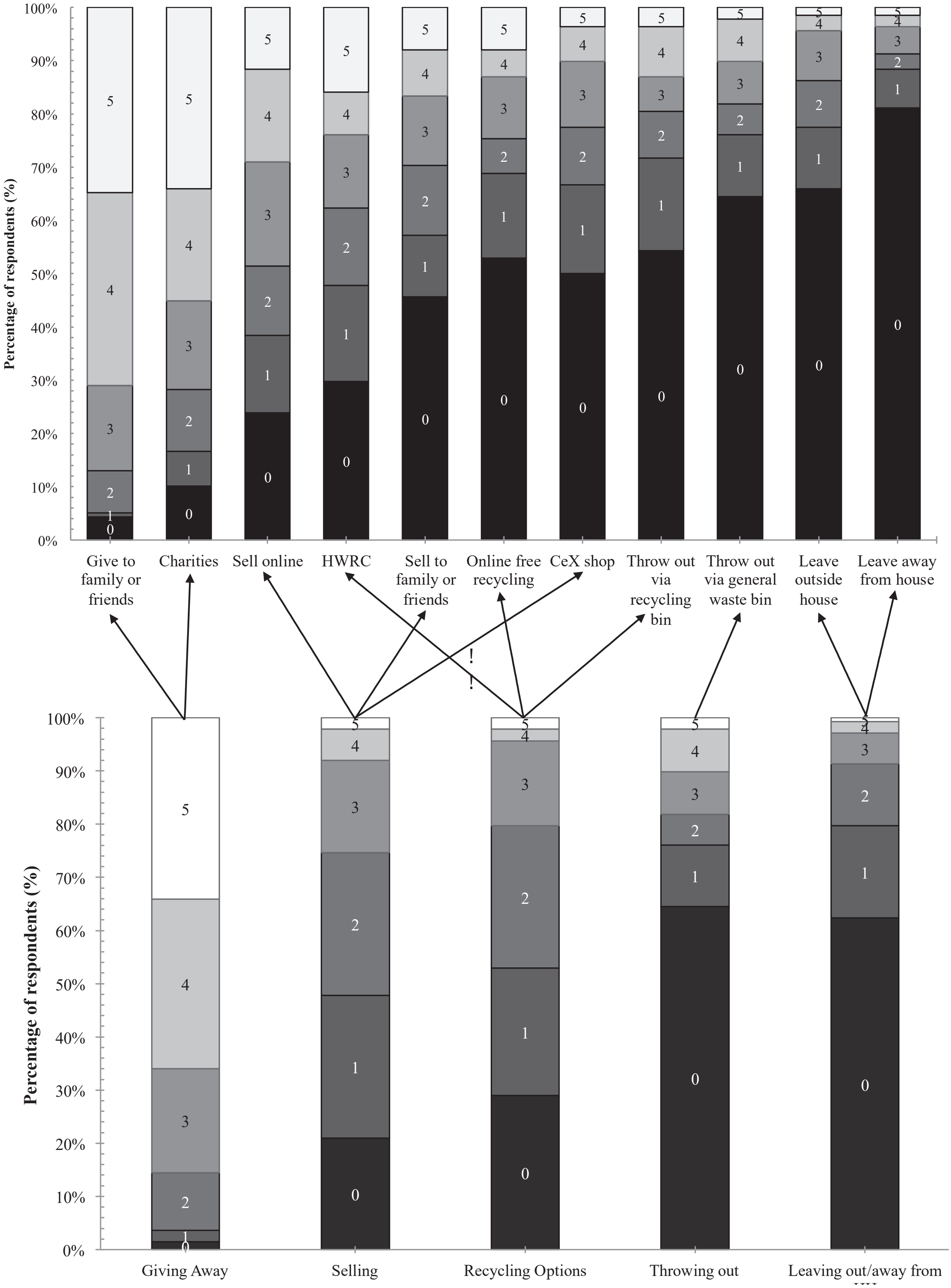

FIGURE 7: Respondent thoughts on the GSD routes of end-of-life management of home entertainment EEE (Proportion of respondents who disagree: 0, to completely agree: 5, n=139).

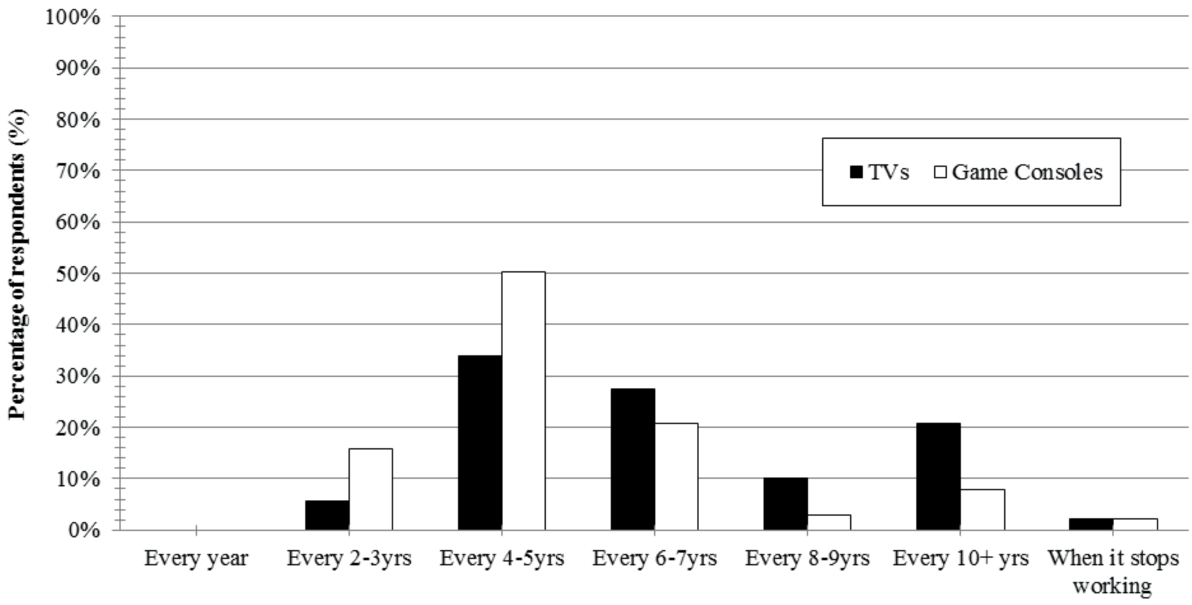

FIGURE 8: Appropriate replacement time for home entertainment electronics (years) (n=139).

television technologies were emerging in the early 1900s, their production flourished after WWII when HE products became a staple in consumer households. Technological advancements were accompanied by growing consumer demand and competition between manufacturers for market dominance. It wasn't until the 1980s when the industry properly manifested itself into a consumer-oriented economy, following the onset of the first compact disc players, video recorders and games consoles, which sold countless of units worldwide (Kang & Schoenung, 2005). At that time, many companies that previously dominated the market like RCA or Atari were replaced by emerging companies such as Sony, Nintendo and Samsung. These new companies transformed HE EEE into an essential part of consumers' lives. Since the 1980s, features and capabilities of EEE have evolved using cheaper materials, while manufactured under higher technological standards and complexity (Kang & Schoenung, 2005). Shorter product lifespans and quicker adoptions of new technologies have triggered subsequent upsurges in electronic waste (McCollough, 2009; Ongondo & Williams, 2011a). Similar to the replacement of analogue technologies by digital devices during the digital switchover of 2012, it can be predicted that the increase in purchases of affordable 'smart' technologies that begun in 2015 will result in the replacement of digital devices like plasma/LCD TVs or DVD players in the near future (Ongondo et al., 2011b).

TABLE 7: Chi-square results for association between age groups and hoarding habits for home entertainment WEEE (n=139).

	Hoarded	Not hoarded	Total
18-24 years	38	9	47
	35.2	11.8	
	1.2	-1.2	
25-44 years	22	16	38
	28.4	9.6	
	-2.8	2.8	
45-64 years	33	5	38
	28.4	9.6	
	2.0	-2.0	
65+ years	11	5	16
	12	4	
	-0.6	0.6	
Total	35	104	139

Chi-square= 9.914, DF=3, p-value= 0.019
For all age groups, top row figures are observed counts, middle row are expected counts, and bottom row are residual Z-scores. For Z-scores a value of >1.96 indicates statistical significance.

In current 'throwaway societies' consumers often replace EEE because they are no longer attracted to/satisfied by them (fashion obsolescence), or because a technologically superior product is introduced on the market (technological obsolescence) (McCollough, 2009). The concept of obsolescence is reflected in this study's results since the majority of respondents discarded HE EEE that were working properly because they wanted a more up-to-date model (Table 5). The concept of planned product obsolescence driven by manufacturers who knowingly produce 'disposable goods' to promote repetitive consumption is often discussed in current literature (Williams, 2016; McCollough, 2009; Ongondo & Williams, 2011a). However, findings from this study show that less than 3% of respondents discarded items that were new but had stopped working (Table 5), indicating that fashion and technological obsolescence were more prevalent across the surveyed sample instead. On average, respondents replaced game consoles more frequently than TVs, suggesting that planned obsolescence is more common amongst smaller HE devices (i.e. game consoles) rather than larger products such as TVs.

To achieve sustainable consumption under a CE and enhance resource efficiency, product lifespans must be

TABLE 8: Chi-square results for association between age groups and hoarding habits for home entertainment WEEE (n=139).

	Social	Sentimental	Monetary	Lack of Awareness
Functional	52	56	72	67
	24	23	13	23
	32	29	22	18
	-3.771	-3.334	-6.165	-4.310
	0.000	0.001	0.000	0.000
Social		45	48	50
		40	20	32
		23	39	26
		-0.104	-4.169	-1.797
		0.917	0.000	0.072
Sentimental			56	47
			21	31
			30	30
			-3.912	-1.819
			0.000	0.069
Monetary				27
				46
				34
				-1.859
				0.063

Series of Wilcoxon tests assessing significant differences between all hoarding reasons at the 0.005 level.
For each hoarding reason, the top-row figures are negative ranks (times that value of column < row). The second-row figures are positive ranks (times that value of column > row). The third-row figures are the rank ties (times that value of column = row). The fourth-row figures are the Z-scores (based on positive ranks) for each Wilcoxon test. The bottom row figures are the p-values for each Wilcoxon test; figures of 0.000 indicate a p value <0.0005.

extended through the use of more durable materials, while the standardisation of product size, shape and location of components will aid dismantling, prolong use and reuse, and ensure material recovery (Long et al., 2016). Current manufacturing techniques are driven by consumer demands and changes in fashion, making modern electronics challenging to disassemble and recover. To encourage the repair and reconditioning of HE EEE rather than their replacement, the cost of repair should be made cheaper than the purchase of new products (McCollough, 2009). Consumer perceptions on the stigma around the 'poorer quality' of reused, repaired or recycled products compared to new electronics should also be broken (Lin & Huang, 2012; Long et al., 2016).

TABLE 9: Results of Kruskal-Wallis test for statistical significance between age groups and hoarding reasons (n=139).

Functional	Social	Sentimental	Monetary	Lack of Awareness
3.537	0.667	4.348	9.284	18.524
0.316	0.881	0.226	0.026	0.000

DF=3, a= 0.05
Top row figures show the Kruskal-Wallis H, and the bottom row figures are the p-values; figures of 0.000 indicate a p value <0.0005.

TABLE 10: Results of Mann-Whitney U tests comparing between age groups and the monetary and lack-of-awareness hoarding factors (n=139).

	Monetary	Lack of Awareness
18-24 and 25-44 years	418.000	246.000
	-0.656	-3.282
	0.512	0.001
18-24 and 45-64 years	544.500	359.000
	-1.012	-3.320
	0.312	0.002
18-24 and 65+ years	97.500	85.000
	-2.719	-3.026
	0.007	0.002
25-44 and 45-64 years	336.000	422.500
	-1.422	747.500
	0.155	0.967
25-44 and 65+ years	62.000	120.500
	-2.728	-0.620
	0.006	0.535
45-64 and 65+ years	121.500	164.000
	-1.886	-0.656
	0.059	0.512
Total	35	104

For each age group pair, the top row figures show the Mann Whitney-U H value, the middle rows are the Z-scores, and the bottom rows are the p-values; figures of 0.000 indicate a p value <0.0005.

4.2 Ownership and hoarding of HE electronics

4.2.1 Ownership levels

Ownership levels for EEE in economically developed nations have increased considerably over the past few decades, transforming cities into hugely exploitable potential DUMs (Pierron et al., 2017). The availability of EEE from in-use and hibernating stocks will affect the exploitability of a mine (Ongondo et al., 2015; Pierron et al., 2017). Ownership levels for HE EEE in the surveyed zones of Southampton were high; with an average of 12 and a maximum of 45 devices found per household. These results are translated into estimates for the exploitation potential of DUMs at local and national levels in Table 11. Overall, we estimate that there are over 1 million HE devices owned and ~440,000 HE devices hoarded in Southampton; scaling up these data suggests that there are >150 million HE EEE owned and ~61 million HE devices hoarded in UK households. The estimations reported in Table 11 may not be entirely representative at a national level due to the survey's small sample size but they provide a realistic order of magnitude value that shows the potential country-wide opportunity for circular economy resource mining. The results are broadly representative of Southampton since the city was randomly sampled across four different deprivation zones.

Almost all survey respondents owned at least one TV, TV-related item (i.e. video player, TiVo, set-top box) and an audio entertainment device (Figure 1). On average, each surveyed household owned 2.8 TV sets, 2.6 audio enter-

TABLE 11: Total number of home entertainment items owned and hoarded on average in Southampton and the United Kingdom (UK) (estimated from survey results).

Home Entertainment EEE	Southampton*		UK**	
	Owned	Hoarded	Owned	Hoarded
TVs	254,620	55,780	35,450,360	7,766,187
Video players	124,685	58,405	17,359,712	8,131,655
TiVo & Set Top Boxes	97,123	11,156	13,522,302	1,553,237
Audio Entertainment Devices	240,839	77,436	33,531,655	10,781,295
Sony PlayStation game consoles	121,404	69,561	16,902,878	9,684,892
Microsoft Xbox game consoles	54,468	28,874	7,583,453	4,020,144
Nintendo game consoles	173,247	132,560	24,120,863	18,456,115
VR Gaming	13,781	5,250	1,918,705	730,935
Total	1,080,167	439,023	150,389,928	61,124,460

**HCC (2008) **ONS (2016)*

tainment devices, 1.4 video players, and 1.1 TiVo/set-top boxes. These findings broadly correspond with those reported by BARB (2018) and the Energy Saving Trust (2012), where 95.4% of households were considered 'digital/interactive TV homes' in 2018, with the average UK household owning 2.0-2.3 TV sets. Overall, ownership of game consoles was less common among respondents compared to TV-related equipment, although more common amongst the younger age groups. The most frequently owned game consoles were the Nintendo Wii, DS and PlayStation 4 (Figure 1). A UK-wide survey by Statista (2018) determined that 36% of consumers claimed to own at least one game console in 2017, of which the majority reported owning a Nintendo Wii and an Xbox360. The slight higher value of game console ownership from this study is probably due to the higher proportion of younger demographic that completed the survey. The study's findings indicated that the Nintendo Wii was the most frequently owned game console, which is in line with the Statista's survey results.

Determining ownership levels and use patterns is essential when evaluating the potential of DUMs. Knowledge gained can also assist in estimating the scale and types of soon-exploitable resources at both local and national levels. This way, waste-processing infrastructure can adapt their technological capacities to efficiently recover the soon-available EEE types (Ongondo et al., 2015). The findings of this study can be used to give a broadly representative idea of the extent of ownership of HE EEE across Southampton and UK households (e.g. over 12 million UK households own at least one TV, and almost 8 million own at least one Nintendo game console; See Table 12).

4.2.2 Hoarding levels

Stockpiled and hoarded HE electronics represent currently inaccessible, high-grade, hibernating stocks, especially rich in metals (Golev et al., 2016). Similar to ownership, hoarding levels of HE EEE provide information on the scope of urban stocks and help map out the complete exploitation potential of a DUM. Hoarding of HE electronics was clearly evident in the surveyed zones of Southampton, with 75% of respondents claiming to have hoarded at least one type of HE device. Tables 11 and 12 translate hoarding levels into an estimate of the 'hibernating' stocks available locally and nationally, assuming that the survey results are representative. We predict that an average of 450,000 HE electronic devices are hoarded in Southampton and that over 61 million are hoarded across the UK (Table 11). We estimate that 71,530 Southampton households and almost 10 million UK households hoard at least one HE item, with at least 5 million households hoarding at least one TV,

TABLE 12: Total number of households owning and hoarding at least one home entertainment item across surveyed zones, including estimations for Southampton and the UK.

Home Entertainment EEE	Surveyed Zones		Southampton*		UK**	
	Owning	Hoarding	Owning	Hoarding	Owning	Hoarding
TVs	134	52	87,936	34,124	12,243,165	4,751,079
Video players	86	53	56,437	34,781	7,857,554	4,842,446
TiVo & set top boxes	84	16	55,124	10,500	7,674,820	1,461,871
Audio entertainment devices	124	58	81,374	38,062	11,329,496	5,299,281
Sony PlayStation consoles	69	47	45,281	30,843	6,304,317	4,294,245
Microsoft Xbox consoles	51	28	33,468	18,375	4,659,712	2,558,273
Nintendo game consoles	86	71	56,437	46,593	7,857,554	6,487,050
Virtual reality gaming	20	8	13,125	5,250	1,827,338	730,935
Total	139	109	91,217	71,530	12,700,000	9,958,993

**HCC (2008) **ONS (2016)*

video player and game consoles (Table 12). The hoarding levels reported in Table 12 assume that each household only hoards one HE item from each product category, yet the survey results indicate that 20-36% of respondents actually hoarded more than one audio entertainment device, PlayStation and Nintendo game console, and 14% hoarded more than one TV. This suggests that the number of devices hoarded in reality across UK households could be closer to the estimates shown in Table 11.

The survey indicated that the most commonly hoarded electronics were videotapes (VHS, Betamax), DVDs, Nintendo game consoles (DS, Wii and GameBoy), VCR players, DVD players and radios (Figure 3). The results show that game consoles were owned less frequently than TV sets, yet hoarded more. The most commonly hoarded items were smaller HE equipment (i.e. videotapes, DVDs, Nintendo DS etc.). This corresponds to other research that also determined the higher likelihood of storing small EEE (Darby & Obara, 2005; Ongondo et al., 2011b). The hoarding of non-digital devices (i.e. videotape products; VCR players) could potentially be explained by the lower level of consumer awareness on the effect of the digital switchover on VCR players, which led to their stockpile once the switchover was complete (Ongondo et al., 2011b). The electronic items hoarded the least (i.e. smart TVs, Nintendo Switch, Xbox One and PlayStation 4 game consoles) were relatively new models of HE electronics (less than five years old) that may yet be hoarded in the future.

Understanding the reasons behind consumer hoarding of HE EEE is very important when considering their EoL management, to tailor collection and take-back facilities in a way that addresses and alleviates hoarding barriers. In this study most respondents illustrated a significant tendency to hoard, mainly due to the attributed residual functional value they placed on HE EEE ("I may need it someday"), followed by their perceived social value ("One day I'll find someone to give it to, who will really want it") (Figure 5). This is reflected in other studies, where respondents stockpiled old (W)EEE mainly as 'back-ups' or with the aim of giving them to someone else (Ongondo et al., 2015; MFE, 2006; Ongondo & Williams, 2011a). Sentimental reasons to hoard were ranked more highly when respondents recalled past behaviours than when they reported on their hoarding dispositions, suggesting that consumers become attached to electronic products more often than they think they do. A significantly higher proportion of the younger age groups (18-24 years) considered lack-of-awareness (LoA) and product cost as important reasons to hoard their unwanted EEE (Table 10). Other studies reflected similar results, where the price of EEE and LoA were important influencing factors for stockpiling and EoL decisions of younger consumers (see Ongondo & Williams, 2011a; MFE, 2006; Ylä-Mella et al., 2015; Li et al., 2012; Pierron et al., 2017).

4.3 Current end-of-life EEE management practices

In the case of purchasing HE electronics, the majority of respondents would wait for a product to become dated/stop working before they purchased a newer model. More than a third of respondents were influenced by fashion and technological obsolescence, as they liked to purchase the newest models on the market. Almost all respondents purchased their new products from electronic retailers, rather than second-hand shops, reinforcing the concept discussed by Long et al. (2016) on the perceived inferior quality of reused, repaired or recycled products. Recent purchasing habits commonly involved acquiring new smart TVs, portable speakers or PlayStation 4 consoles. These findings are in line with 2017 sales figures, where 82%, 74% and 76% of the market-share was attributed to smart TVs, PlayStation 4 consoles and portable speakers, respectively (J'son & Partners, 2017; D'Angelo, 2017; Business Wire, 2017).

The study indicated that when HE products were in working condition but of no use to their owners, they were most frequently given to relatives and friends or donated to charities. This decision can be attributed to the residual functional value assigned to HE products, which can be beneficial in prolonging the products' lifetime and encouraging reuse. Respondents also reported a disposition to sell their unwanted EEE online, or take them to HWRCs. The disposition to recycle EEE was evident among respondents, yet their disposal habits reflected a higher probability of discarding items via general refuse. Ongondo & Williams (2011a) determined that consumers tend to send bulkier items to HWRCs, while medium-sized items with residual value (such as HE electronics) are donated to charities or given to friends and family, which match this study's findings. Broken equipment or smaller items such as remote controls or game console handsets usually do not warrant a trip to the HWRC and are most often hoarded or disposed of in the general waste (Ongondo & Williams, 2011a; Pierron et al., 2017). It was also evident that respondents' perceived residual value of (W)EEE would cease once products became very dated or were broken.

The average replacement time for HE electronics was 4-5 years, with game consoles being replaced slightly sooner than TVs. Unless hoarded, this makes the majority of HE EEE purchased since 2012 soon-exploitable resources within a DUM. HE equipment lifecycles are also illustrated through respondents' purchasing and disposal habits. The majority of respondents had purchased additional HE products in the last 5 years, as well as disposed of old electronics, demonstrating a clear average 5-year lifecycle. These findings match the current literature, which suggests that most EEE are discarded within 3 to 6 years (Hursthouse et al., 2017; Ongondo et al., 2015; Cooper, 2002).

4.4 Distinct Urban Mine potential

The high ownership levels for HE electronics in Southampton make it an extremely plausible metal-specific DUM, abundant in ferrous metals, aluminium, copper and plastics (Haig et al., 2012). Its potential as a DUM is strongly influenced by the public's decision to hoard or dispose of their (W)EEE. Reuse and recycling are positive decisions for accessing the DUM whereas discarding and hoarding prevent the recovery of resources. It is essential to incorporate information on the EoL decisions of consumers into waste management efforts. Access to an urban mine is closely related to the prevention of stockpiling and thus the willingness of consumers to give up their products (Ongondo et al., 2015; Li et al., 2012). Promoting the collection and recycling of (W)EEE through awareness campaigns could help consumers associate EoL electronics with material recovery, and deter from their perceived residual value. Awareness campaigns can also be applied to improve public knowledge on the disposal methods for HE (W)EEE, by providing information on collection centres and schemes. To encourage recycling behaviour, recovery schemes must incorporate more accessible and convenient collection points, with proximity to commonly visited urban hubs (e.g. transport hubs or shopping precincts) (Ylä-Mella et al., 2015; Morris & Metternicht, 2016). Additionally, collection services could incorporate monetary incentives (e.g. cash payments or vouchers), to encourage participation of younger age groups who frequently hoard electronics due to monetary reasons (Ongondo & Williams, 2011b; Li et al., 2012). Such incentives could also promote the faster release of (W)EEE from consumers' homes, ensuring access to recoverable materials in a shorter timeframe. A study by Hursthouse et al. (2017) on the efficient recovery of critical raw material under a closed-loop economy, determined that working closely with local social enterprises and schools to collect (W)EEE might encourage higher recovery rates, due to their local dimension and trusted relationship with the public. Under the same concept, Ongondo et al. (2015) suggested that universities could be utilised as recovery centres for (W)EEE. Targeting the recovery of larger items (e.g. plasma TVs) or devices with a higher proportion of multiple hoarded devices (e.g. game consoles) might be an easier way to influence consumer hoarding due storage space constraints. This approach could gradually stimulate consumers to view the EoL of all (W)EEE differently (Hursthouse et al., 2017).

5. CONCLUSIONS

This study has assessed the effect of consumer behaviour on the release of HE (W)EEE into the circular economy. It provides original contributions to the current literature by establishing, presenting and analysing previously unavailable data on the ownership, use and hoarding levels of HE EEE in a typical city DUM, and the reasons behind their hoarding. In a city with a population of ~240,000 in the UK, ownership levels were very high, with an average of 12 home entertainment items owned per household. This makes urban areas extremely plausible as DUMs; we estimate that there are over 1 million HE devices owned and ~440,000 HE devices hoarded in Southampton and >150 million HE EEE owned and ~61 million HE devices hoarded in UK households. Hoarding is therefore common, especially for smaller or older equipment, due to their perceived residual value. HE product lifecycles averaged 4-5 years. The most common EoL routes were donating to relatives, friends or charities; hoarding; recycling via Household Waste Recycling Centres; or discarding items in general refuse. To encourage the recovery of EoL HE equipment in a DUM, convenient and accessible collection points should be promoted via awareness campaigns and incentives.

The most common respondent behaviours involved hoarding of small (W)EEE, the recycling of larger products via HWRCs, and the gifting/donating of working EEE with perceived residual value. EoL HE equipment was often hoarded as a back-up (i.e. stockpiling) or with the aim of giving them away in the future. More respondents from younger age groups considered lack-of-awareness of disposal options and the steep price of purchased items as important influencing factors for hoarding. The study also quantified soon-exploitable and hibernating stocks within a large urban area's surveyed zones, to predict product and material yields that could be made available to a DUM in the near future. To ensure recovery of used EEE and WEEE, waste management efforts should:

- Promote the recycling and reuse through awareness campaigns on collection schemes for consumer electronics.
- Establish convenient and accessible used EEE and WEEE collection points to encourage regular (periodic) harvesting and incentives.to encourage reuse / recycling behaviour, with potential monetary incentives.
- Target the recovery of items that take up more space in consumer households (larger equipment or with a higher proportion of multiple devices).
- Encourage faster reuse of EEE via donations to charities, relatives or friends in order to gradually stimulate consumers to view EoL WEEE differently.

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CONCENTRATIONS OF BROMINATED FLAME RETARDANTS IN PLASTICS OF ELECTRICAL AND ELECTRONIC EQUIPMENT, VEHICLES, CONSTRUCTION, TEXTILES AND NON-FOOD PACKAGING: A REVIEW OF OCCURRENCE AND MANAGEMENT

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ABSTRACT

A synthesis of 4000 published data from 37 references of brominated flame retardants (BFRs) concentrations, in plastics of electrical and electronic equipment, vehicles, construction products, textiles and non-food packaging is presented. For POP decabromodiphenylether, a median concentration of 50 mg/kg in plastics of electrical and electronic equipment (n=276), is reported, as well as 31 mg/kg in plastics of vehicles (n=80), 0 mg/kg in plastics of construction (n=81), and 0 mg/kg in plastics of textiles equipment and upholstery (n=75). The mean concentrations are 5200, 3100, 8700 and 6500 mg/kg, respectively. In non-food packaging (expanded or extruded polystyrene), hexabromocyclododecane is present in some samples. All these plastics always have at least some samples with one BFR with a mean concentration above the EU regulatory concentration limit for substances, products or hazardous waste. The distribution of all reported concentrations of PBDEs is skewed, with for instance, in plastics of vehicles, 84% of the data lower than 1000 mg decaBDE /kg, and some large values up to 150 000 mg/kg. The sorting and the up-to-date management technologies are for these categories of plastics (estimated to be 40% of the plastic use in the EU, the brominated fraction of them being a few percent) necessary to weed out banned substances in the circular economy.

1. INTRODUCTION

The EU is developing a strategy on plastics in a circular economy (EC 2017). Some plastic flows have a low rate of recycling and reuse of plastics due to, among other reasons, quality issues (presence of additives or mixing of different types of polymers). On the other hand, scientific progress in knowledge of substances results in the restriction or banning of elements or substances in products, and restricted management options when these substances are present in waste. This happens frequently as a legacy of previous practices. Research is active in this field. Human biomonitoring of global populations has identified an exposure to a range of plastic additives, detectable in some cases in most people (Galloway et al. 2019). A database of chemicals associated with (food) plastic packaging found 906 chemicals identified as likely, 3377 chemicals as possibly associated; the 148 most hazardous of them being classified in the CLP the United Nations' Globally Harmonized System (GHS). Some are classified as persistent, bioaccumulative and toxic (PBT) or very persistent and very bioaccumulative (vPvB), and endocrine disrupting chemicals (EDC) (Groh et al. 2019). A general overview of the plastic waste management options and the additives presents in plastics can be found in Hahladakis et al. (2018). The conclusion is that recycled plastics must be sorted in order to avoid the spreading of some unwanted additives in the new products.

The use of plastics in Europe is divided as follows: 39.7% for packaging (food- and non-food-), 19.8% for building and construction, 16.7% for medical equipment, plastic furniture and furniture equipment, technical parts used for mechanical engineering or machine-building, 10.1% for automotive, 6.2% for electrical and electronic equipment, 4.1% for household, leisure and sport, the rest being used in agriculture (3.4%) (PlasticsEurope 2018). For instance, hard plastics and foams now account for about 150 kg in modern cars, 16% of which could be brominated (this study). They are protected against fire with, among others, brominated flame retardants (BFR), some of them being now classified as hazardous or persistent organic pollut-

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ants (POP). An important review on POP substances including BFRs in plastics is (EC 2011).

The compilation of PBDE production data prepared for the POPs Reviewing Committee of the Stockholm Convention estimated the total production of all PBDE from 1970 to 2005 as between 1.3 million and 1.5 million tons (UNEP 2017a). DecaBDE production is estimated at between 1.1 and 1.25 million tons. The three largest producers are Israel Chemical Limited - Industrial Products (ICL), Albemarle Corporation and Chemtura - Great lakes Solutions. They offer products or additives for construction (particularly insulation foam of polystyrene and polyurethane), electrical and electronic equipment (EEE), transport, electrical wires, coal flue gas cleaning, agriculture and oil refining, among others (Albemarle 2018, Chemtura 2018,, ICL 2012). A huge variety of BFR is produced. ICL has a specific product guide "Fire protection for automotive and transportation". The BFR are used in different polymers in the 1-15% range (Arias 2011, Alaee et al. 2003).

To assess the concentration of POP substances in the end-of-life plastics, published or reported data have been gathered and analyzed. The objective is to establish whether pre- or post-shredder sorting of plastics of different electrical and electronic equipment, end-of-life vehicle (ELV), textiles and construction waste is necessary to comply with the EU regulations. Particular attention is paid to the recently banned decaBDE and plastics of ELV.

The EU Waste Framework Directive (EU 2018) has set up a separate collection for plastics, and, by January 1, 2025, for textiles. It aims to promote selective demolition in order to enable the establishment of sorting systems for waste construction and demolition, and by extension, for plastic. Waste containing POP substances above concentration limits cannot be landfilled in the EU 2016. Energy recovery as solid recovered fuel must be done in installations fulfilling strict flue gas emission requirements. This paper offers to give some information on the contaminant concentration in plastics and on which type of sorting could be applied for these.

1.1 Abbreviations

ABS	Acrylonitrile Butadiene Styrene
ASR	Automotive shredder residue
BDE	Bromodiphenylether
BFR	Brominated flame retardant
CLP	Classification and Labelling of Preparations and Substances
c-pentaBDE	Commercial pentabromodiphenylether
DBDPE	Decabromodiphenylethane
DDC-CO	Dechlorane plus
decaBDE	Decabromodiphenylether
EDC	Endocrine disrupting chemicals
EEE	Electrical and electronic equipment
ELV	End-of-life vehicle
EPS	Expanded polystyrene
EU	European Union
GHS	United Nations' Globally Harmonized System
HBCDD	Hexabromocyclododecane
heptaBDE	Heptabromodiphenylether
hexaBDE	Hexabromodiphenylether
Hnnn	Hazard Statement Code of substances, H followed by 3-digits number
HP 1 to HP 15	Hazard Property of waste, 1 to 15
ICL	Israel Chemical Limited
PBB	Polybromobiphenyls
PBDE	Polybromodiphenylether
PBT	Persistent, bioaccumulative and toxic
PCB	Polychlorobiphenyls
pentaBDE	Pentabromodiphenylether
PFHxS	Perfluorohexanoic acid
PFOA	Pentadecafluorooctanoic acid
POP	Persistent Organic Polutant
PS	Polystyrene
PST	Post-shredder treatment
PUR	Polyurethane
REACh	Registration, Evaluation and Authorization of Chemicals
RoHS	Restriction of Hazardous Substances
SCCPs	Short chain chlorinated paraffins
TBBPA	Tetrabromobisphenol A
tetraBDE	Tetrabromodiphenylether
UNEP	United Nations Environmental Program
USGS	United States Geological Service
vPvB	Very persistent and very bioaccumulative
WEEE	Waste of electrical and electronic equipment
XPS	Extruded polystyrene

2. REGULATED SUBSTANCES IN PLASTIC MATERIAL (PRODUCTS AND WASTE)

2.1 Limitation of concentrations in products

In electrical and electronic equipment, the following elements and substances are regulated (Directive Restriction of Hazardous Substances (RoHS), EU 2011): cadmium: 100 mg / kg; chromium (VI), mercury, lead: 1 000 mg / kg; polybrominated diphenyl ethers (PBDEs): 1 000 mg / kg.

In products in general, persistent organic pollutants (POPs) are limited (EU 2016). These substances are defined in the Stockholm Convention as not biodegradable, bioaccumulative and dispersed in the natural environment. Some are prohibited, and actions of emissions reduction must be put in place and monitored (Annex III of EU 2016). The limits in the products are as follows: polychlorinated biphenyls (PCBs): banned from use (Annex III of EU 2016); hexabromocyclododecane (HBCDD): 100 mg / kg in products subject to review by the Commission by 22/03/2019; hexachlorobutadiene (HCBD): 100 mg / kg (EU 2017a) (not sought here); tetra- or penta- or hexa- or hepta-bromodiphenylethers (PBDE): 1 000 mg / kg when the products are recycled (EU 2016); decabromodiphenylether (decaBDE): 1 000 mg / kg (EU 2017a) after 02/03/2019, with a proposition of the European Parliament as unintentional contaminant of the sum of tetra-, penta-, hexa-, hepta- and decaBDE of 500 mg/kg, and decaBDE of 10 mg/kg (EP 2019; decided in 2019 to 1000 mg/kg); short chain chlorinated paraffins (SCCPs): 10 000 mg / kg (not sought here).

In addition, there are three substances and groups of substances that are currently POP "candidates" (UNEP 2017b): dicofol, pentadecafluorooctanoic acid (PFOA –

listed in the Stockholm Convention in 2019), its salts and related compounds, and perfluorohexanoic acid (PFHxS), its salts and related compounds. Dicofol is an insecticide (not used in plastics), while PFOA and PFHxS are water-repellent products used, among other things, in vehicle textiles. They were not sought in this study.

Other substances also are a matter of concern. Decabromodiphenylethane (DBDPE) has been reported in WEEE plastics (Wäger et al. 2012, Wäger et al. 2010). Dechlorane plus (abbreviation DDC-CO, CAS No. 13560-89-9), a flame retardant with 12 chlorine atoms, was added to the REACh list of substances that are extremely worrisome (RPA 2014). Dechlorane plus has two isomers (syn- and anti-). It has been found in plastic of vehicles (coarse and fine fractions), as well as in WEEE (Morin et al. 2017). Some data on these substances are reported in this paper.

2.2 Hazardous waste classification (by hazard properties and POP content)

For the classification as hazardous waste, the 15 EU hazard properties, HP 1 to HP 15, are defined by the presence or the concentration of substances with specific hazard statement codes (EU 2014a, EU 2017b, EC 2003a). The hazard properties of the relevant substances in plastic waste (with their hazard statement codes from the CLP) for hazard classification are: (i) HP 7 "Carcinogenic", if Sb_2O_3 (hazard statement code H351 1B)> 10 000 mg / kg, equivalent to Sb> 8 354 mg / kg, or if hexabromobiphenyl (H350 1B)> 1 000 mg / kg; (ii) HP 10 "Reprotoxic", if hexa- or hepta- or octaBDE (H360 1B)> 3 000 mg / kg, or if HBCDD (H361)> 30 000 mg / kg; (iii) HP 14 "Ecotoxic", if the sum of (tetraBDE, pentaBDE, HBCDD and TBBPA) (H410)> 2 500 mg / kg from July 2018 (EU 2014b, EU 2017b) (> 25 000 mg / kg before July 2018).

Regarding the POP substances, it should be noted that not all of them can classify a waste as hazardous.

A first group classifies waste as hazardous if the following substances (EU 2016, EU 2014b, EU 2017b) are present at a concentration exceeding a limit. About plastic waste, these substances are in practice (in order of increasing concentration): polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD / PCDF)> 15 µg / kg, which can be created unintentionally from RFBs by friction and localized heating of BFRs during waste treatment operations (not sought here); polychlorinated biphenyls (PCBs)> 50 mg / kg; hexabromobiphenyl (HBB)> 50 mg / kg.

A second group is the POP substances that do not make the waste hazardous because they are classified. However, those wastes must be managed specifically (section 2.4): the PBDEs and the HBCDD. They can trigger the classification as hazardous by their hazard statement codes and their concentrations for the hazard properties HP 4, 5, 10 and 14, but the classifying concentrations are higher than the ones for specific waste management by their content (s) in POPs (see below).

2.3 Waste management (by their hazard properties HP)

Waste can be landfilled if it meets maximum leachable content and total substance levels (EC 2003a, EC 2003b). In practice, non-hazardous and non-POP plastics can be landfilled in landfills for non-hazardous waste. For hazardous plastic waste, the loss on ignition (<10%) or the total organic carbon content (TOC <6%) are exceeded, and these wastes are not allowed to be landfilled in landfills for hazardous waste but must be incinerated. The ashes must meet the criteria for the disposal of hazardous waste, which is possibly the case after a stabilization with cement.

2.4 Waste management (by their content (s) in POPs)

The POP substances in waste exceeding the concentration limits of POP regulation for waste (being hazardous or not) must be irreversibly transformed so that the substances no longer have the characteristics of a persistent organic pollutant (EU 2016 Annex IV): polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD / PCDF)> 15 µg / kg; polychlorinated biphenyls (PCBs)> 50 mg / kg; hexabromobiphenyl (HBB)> 50 mg / kg; HBCDD< 1 000 mg / kg; sum of tetra-, penta-, hexa- and heptaBDE> 1 000 mg / kg; and under discussion: sum of tetra-, penta-, hexa-, hepta- and decaBDE> 1 000 mg / kg, and a possible lowering two years later to 500 mg / kg (EP 2019).

These wastes cannot be recycled and must be handled only by the following operations: D9 Physicochemical treatment; D10 Incineration on land and; R1 Main use as fuel or other means of producing energy, excluding wastes containing PCBs.

2.5 Sorting to separate parts with controlled substance(s) above concentration limits

For electrical and electronic equipment, polychlorinated biphenyls of capacitors and brominated flame retardants of plastics (among other substances) must be separated (EU 2016). A technical specification (CENELEC CLC/TS 50625-3-1) recommends the sorting of WEEE plastic with a concentration limit of 2000 mg / kg total bromine. In practice, the sorting is carried out either by density (by flotation or by in-line X-ray transmission), or by measurement of bromine (with a portable or in line X-ray fluorescence device).

For products in general, the POPs Regulation (EU 2016) specifies: 'During this elimination or recovery, any substance listed in Annex IV may be isolated from the waste, provided that it is subsequently eliminated…".

3. REPORTED CONCENTRATION IN PRODUCTS AND WASTE AND DISCUSSION

3.1 Sampling, analysis, reported data

The reported concentration of BFRs in plastic particles/ parts has a skewed distribution (see section 3.10), with many particles/ parts with nil or low concentration, and a small number of particles/ parts with scattered high concentrations. The principle of sampling is that a smaller (sub-)sample of a flow or a heap will have the same composition as a larger flow or heap if "enough" particles/ parts are taken. The sampling of waste is described in EN 14899 with five technical documents FD CEN/TR 15310-1 to -5. Considering the skewed distribution of the concentration of bromine per plastic scrap and the analytical variability of BFR analysis, it can be calculated EN 14899, EN 15002) that a representative sample should contain 100 000 particles (Hennebert 2019). For plastics of WEEE, a technical specification (CENELEC CLC/TS 50625-3-1) recommends the sampling per day of production of the shredder of 10 times 3 litres if the size of the largest particle is < 20 mm, 10 times 5 litres for a particle size of 20-50 mm, and 10 times 10 litres for particle size > 50 mm. The composite sample is coned, mixed and quartered two times (volume divided by 4) and 7.5, 12 or 25 litres are sent to the laboratory. The number of particles in these laboratory samples should be checked (Hennebert 2019). The treatment of those samples is described in (EN 15002). The laboratory will cryo-grind it in two or three steps ([10-5] mm and [1 - <0.1] mm) with an intermediate mixing and quartering steps. An aliquot of 0.1 g will be extracted thanks to a solvent, and some [0.1-0.01] ml of the purified extract will be injected in the analysis apparatus (EN 62321-6).

The extraction and analysis of BFR in plastics should be performed by certified industrial laboratories. A publication and a report on ELV plastics vehicles have performed an incomplete extraction of BFR (Table 1). In one case, the samples were crushed at 4 mm and extracted with an accelerated solvent extractor with toluene at 130°C three times (Morin et al. 2017). In the second case, the sample was crushed between 1 and 5 mm respectively and extracted by cold sonication with hexane for 15 min (SMTT 2012). The extraction yields were not controlled, and the laboratories were not accredited for theses analyses. The measured concentrations in automotive shredder residue are low (Table 1) and those 21 data have not been used in this study. Another case has been reported in car fluff (Norwegian Climate and Pollution Agency 2012).

Published data are heterogeneous. Some are single concentration data, and others are a range of concentration data, with the report of the range values number (n), the minimum (min), the mean (mean), and the maximum (max) of the range. A detailed analysis of these ranges of concentrations cannot be presented here, due to page limitations. The mean concentrations presented here are the mean of all the single concentrations and the min, mean and max concentrations of the ranges. Data reported as "< x mg/kg" (lower than the limit of detection or quantification of the laboratory) have been accounted as "x" mg/kg. Commercial bromodiphenylethers mixtures (c-BDE) were typically produced at three different degrees of bromination, in particular c-pentaBDE, and are reported as such, aside with pentaBDE. Some authors report the sum of BDE, that were classified POP at the time of their publication (tetra-, penta-, hexa- and hepta-BDE), under the name POP-BDE. PBDE is the sum of all the BDEs data for the same sample. Hexabromocyclododecane (HBCDD) and tetrabromobisphenol A (TBBPA) concentrations are significant and are presented.

TABLE 1: Cases of reported BFR concentrations in plastics of vehicles with presumed incomplete extraction during the laboratory analyses.

Mean concentration (mg/kg)	Morin et al. 2017	SMMT 2016
DecaBDE		3.10
PentaBDEs		0.11
HeptaBDEs		0.06
TetraBDEs		0.04
HexaBDEs		0.01
PBDEs	47.24	
HBCDD (Hexabromocyclododecane)		0.565
PBBs (Polybromobiphenyls)		0.05
Sum of PBBs, PBTs (Polybromoterphenyls), dechlorane plus anti and syn, hexabromobenzene, pentabromotoluene and pentabromoethylbenzene	0.36	

Polybrominated biphenyls (PBBs) and polychlorinated biphenyls (PCBs), banned for a long time, have no data for these plastics, and it is known that their concentrations are close to zero in WEEE and automotive plastics (Hennebert and Filella 2017, Hennebert 2018 a, b). Three authors have studied other BFRs. These data are numerous (1 440) and briefly presented in section 3.6. A total of 4 016 RFBs data and 264 Br and Sb data were used (total 4 280 data) (Table 2). The total number of data sorted by category and by bibliographic reference, for all reported substances and all elements, is presented in Table 3. The reference of the Norwegian Environment Agency - BIPRO (2016) includes of 17 citations other than the ones cited in the References, with less abundant data, and source not fully defined). A partial previous version of this work has been presented as an oral communication (Hennebert 2018b).

In this study, since the distribution of concentrations is never gaussian (normal), the minimum, the median, the mean and the maximum concentrations are presented, as well as the percentage of data exceeding a given (regulatory) concentration.

3.2 BFRs in electrical and electronic equipment / WEEE

A total of 962 data (including data of 40 ranges) have been gathered (Table 2). The equipment categories are small and large household (cool/heat, and non cool/heat) equipment, TV, screens, IT equipment, wires, white plastics, and so on, up to a total of 97 different categories. The summarized concentrations are presented in Table 4. No distributions are gaussian (normal): the median concentrations are a smaller fraction of the mean concentrations, and the maximum concentrations are larger multiples of the mean concentrations than in normal distributions (Hennebert 2019). These skewed distributions for the high concentrations are presented with more details in section 3.10. Concentrations > 1 000 mg/kg are highlighted (EC 2003a, EU 2011, EU 2012, EU 2017a).

Large household appliances and cooling and freezing apparatus (two references: Wäger et al. 2012 and Vojta et al. 2017) have a significantly lower concentration of decaBDE than the other equipment (Mann-Whitney test, $p < 0.0001$), but one reported concentration is equal to the concentration limit of 1 000 mg/kg (Table 5).

TABLE 2: Total number of data sorted by category from literature, by BFR and element (Br and Sb), and number of data presented in this study ("range data" is reported as a range of concentration, most of the time with the number of the range values, the minimum, the mean and the maximum of the range).

Parameter	Individual data and range data						Number of ranges					
Category	**EEE/WEEE**	**Vehicles/ ELVV**	**Construction**	**Textiles**	**Packaging**	**Total**	**EEE/WEEE**	**Vehicles/ ELVV**	**Construction**	**Textiles**	**Packaging**	**Total**
BFRs												
PBDE	781	215	716	437		2149	35	26	4	12		77
of which DecaBDE	276	80	81	75		512						
HBCDD	112	21	84	59	57	333	5	2	2	7		16
TBBPA	69	11	6		8	94						
Sub-total	962	247	806	496	65	2576	40	28	6	19		93
Non-regulated BFRS	257	22	771	374	16	1440						
Total BFRs	1219	269	1577	870	81	4016						
Elements												
Br	92	36	9	70	6	213	38	12	3	23	2	78
Sb	32	11	4	4		51						
Total elements	124	47	13	74	6	264	38	12	3	23	2	78
Grand total	1343	316	1590	944	87	4280						

3.3 BFRs in vehicles and end-of-life vehicles

A total of 247 data (including data of 28 ranges) have been gathered (Table 2). Reported concentrations of brominated flame retardants in cars, automotive shredder residue (ASR), and sorted post shredder treatment fractions (PST fractions) are presented in Table S1 (supplementary information). There are 117 data for Car, 46 data for ELV, 51 for ASR and 33 for PST (total 247 data). The results are indicative since these figures are unweighted means of single data, and min, mean and max of range data. The light fluff fractions are documented as "light ASR" or "mixed light plastic" of the ASR category, or the lighter fractions of the PST category. The PST fractions are sorted by flotation, the most brominated fraction being the densest. PST data treatment fractions are scarce. As a result, data from mixed waste (WEEE and ELV) are also presented for this category. A summary is presented in Table 4.

The discussion will focus on the concentration of decaBDE because this substance is the only one that allows direct comparison between categories (Table 6). Significant mean concentrations are observed in some plastic car parts (rounded mean 3 500 mg/kg, above the concentration limit of 1 000 mg/kg), including printed circuit boards, with the highest concentrations in seats, foam and upholstery (27 000 mg/kg). These high values raise the mean concentration, while the median concentration is only 50 mg/kg. The sampling of the parts can be biased: authors may have looked for (previously known) parts with high concentration. In some studies, it is a deliberate choice: a multitude of parts are first measured in the field for total bromine with a hand-held X-ray fluorimeter, and only the most concentrated parts are sent to the laboratory for BFRs analyses. But low concentrations are also reported. This peculiar distribution makes it difficult to understand easily this waste flow. Concentrations in ELV are roughly similar to concentrations reported in car parts.

Much lower concentrations are observed in ASR (rounded mean 400 mg/kg). The sampling of the ASR is probably not biased: sub-samples are taken at periodic interval from the scrap flow, without previous knowledge of concentration. The median of 44 mg/kg is comparable to the median of car parts. It is significantly lower than the mean: the distribution is here also skewed by some high values. Lower concentrations than in car parts (see details in Table S1) are probably because plastics and foam are only 20% of the mass of ASR.

The concentrations in sorted plastics (so-called post-shredder treatment of ASR plastics, here by density separation) are very low (mean 14 mg/kg). The effectiveness of sorting (here mainly by density difference) is proven. Details can be found in Leslie et al. (2016). A recent study (Swerea IVF 2018) confirms it for ELV sorted plastics (a French sample of PS/ABS contained 140 mg/kg decaBDE, and a UK sample of ABS contained 5 mg/kg decaBDE, 15 mg/kg TBBPA, and 14 mg/kg decabromodiphenylethane - DBDPE).

TABLE 3: Total number of data sorted by category from literature, for all reported substances and all elements.

Publication	E	V	C	T	P	Total
Abdallah et al. 2018			1		50	51
Allen et al. 2008	50			30		80
ARN 2015		1				1
Arp et al. 2015	67	25				92
Ballesteros-Gomez et al. 2013	4					4
Chen et al. 2010	17	5				22
COWI 2013	2	2				4
Drage et al. 2018	60	24	24	60		168
EMPA 2010	21					21
Federal office of the Environment (FOEN) 2017	33					33
Gallen et al. 2014	8	3		1		12
Guzzonato et al. 2016	93					93
IVM, IVAM 2013	43	28				71
Japanese Ministry of the Environment (MOE) 2011		6				6
Jinhui et al. 2017	5	2	2	2		11
Kajiwara et al. 2013	1			35		36
Leslie et al. 2016	45	88		7		140
MEPEX 2012		6				6
Morf et al. 2005	1					1
Niinipuu 2013		8				8
Norwegian Climate and Pollution Agency - Mepex 2012		232				232
Norwegian Environment Agency - BIPRO 2016 (17 citations)	47	37	4	11		99
Petreas et al. 2009	12	3				15
Puype et al. 2015	187					187
Rani et al. 2014			24		32	56
RPA 2014	6	4	4	4		18
Sindiku et al. 2015	4					4
Sinkkonen et al. 2004	3	3				6
Stubbings et al. 2014				2		2
Taurino et al. 2010	20					20
Turner et al. 2017 Antimony	4	4	4	4		16
Turner et al. 2017 Bromine	9		9	9		27
Vojta et al. 2017	552	48	1656	816		3072
Wäger et al. 2011	2					2
Wäger et al. 2012	1032					1032
WRc 2012	136					136
WRc addendum 2012	2	18				20
Grand total	**2466**	**547**	**1728**	**981**	**82**	**5804**

The concentrations are lower than what is reported and observed in the corresponding fractions of WEEE. In France, in unsorted shredded plastics of small household appliances, cathode ray tubes and flat screens, the concentrations were 1 950 mg/kg decaBDE in 2014 and 1 395 mg/kg in 2015 (n = 10 - in triplicates) (Hennebert and Filella 2017). For sorted plastics (fraction < 2 000 mg/kg Br), the mean concentrations were 148 mg/kg decaBDE in 2014 and 522 mg/kg decaBDE in 2015 (n = 4).

For elemental concentrations, the most concentrated element is bromine (mean concentration in car parts and ASR = 8 564 mg/kg, n = 27). The concentration used in WEEE for sorting (2 000 mg/kg, (CENELEC CLC/TS 50625-3-1) is trespassed. The second most concentrated element

TABLE 4: Summary of concentration of some brominated flame retardants in plastics of EEE/WEEE, vehicles/ELV, construction, textiles and non-food packaging from literature data, and percentage of data lower and larger than the concentration limit of 1000 mg/kg (**hazardous waste** [bold], concentration allowed in <u>products</u> exceeded [underlined], concentration of technical specification for sorting of WEEE plastics *Br> 2000 mg/kg* exceeded [italics]), in yellow: to be sorted for recycling) (in green: lowest % data per category).

Category	BFR	Concentration data (mg/kg)					% data < 1 000 mg/kg	% data < 500 mg/kg
		n	Min	Median	Mean	Max		
EEE/WEEE	PBDE	781	0	7	*2663*	*154000*	90%	87%
	of which DecaBDE	276	0	50	*<u>5216</u>*	*<u>150000</u>*	84%	80%
	HBCDD	112	0	200	<u>137</u>	<u>1600</u>	99%	99%
	TBBPA	69	20	20	*3155*	*63000*	93%	91%
Vehicles/ELV	PBDE	215	0	6	*1623*	*85000*	92%	88%
	of which DecaBDE	80	0	31	<u>3102</u>	<u>85000</u>	88%	78%
	HBCDD	21	0	10	<u>386</u>	***<u>4400</u>***	90%	90%
	TBBPA	11	20	20	27	87	100%	100%
Construction	PBDE	716	0	0	<u>1713</u>	*300000*	99%	99%
	of which DecaBDE	81	0	0	*<u>8662</u>*	*300000*	95%	94%
	HBCDD	84	0	0	<u>317</u>	***<u>10000</u>***	92%	89%
	TBBPA	6	0	0	0	0	100%	100%
Textiles and upholstery	PBDE	437	0	0	*2080*	*130000*	95%	94%
	of which DecaBDE	75	0	0	*<u>6511</u>*	*<u>120000</u>*	84%	81%
	HBCDD	59	0	0	***<u>3465</u>***	***<u>51000</u>***	86%	86%
	TBBPA	-	-	-	-	-	-	-
Non-food Packaging	PBDE	-	-	-	-	-	-	-
	of which DecaBDE	-	-	-	-	-	-	-
	HBCDD	57	0	11	<u>232</u>	***<u>5897</u>***	91%	88%
	TBBPA	8	0	-	0	1	-	-

TABLE 5: Concentration of decaBDE in large household appliances and other categories of EEE/WEEE from literature data (concentration allowed in <u>products</u> exceeded [underlined]).

DecaBDE (mg/kg)	n	Minimum	Median	Mean	Maximum	Standard deviation
Large household appliances, cooling and freezing appliances	36	0	50	84	1000	162
Other categories	240	0	50	<u>5 985</u>	<u>150 000</u>	22 935

is lead (mean concentration in car parts = 6 499 mg/kg, n = 13) (result not shown, Hennebert 2018a). The concentration of the Restriction of Hazardous Substances (RoHS) (EU 2011) applicable to EEE is trespassed (1 000 mg/kg). According to some authors, it is due to printed circuit boards soldering.

TABLE 6: Concentrations of decaBDE in car parts, automotive shredder residue (ASR) and post shredder treatment fractions – unweighted mean concentrations and median from literature data (concentration allowed in <u>products</u> exceeded [underlined], concentration of technical specification for sorting of WEEE plastics *Br> 2000 mg/kg* exceeded [italics]).

Concentration (mg/kg)	DecaBDE		
Category	Mean	Median	n
Car - parts	*<u>3 469</u>*	50	35
ASR	386	44	21
PST – sorted plastics	(101)	(6)	(11)
Of automotive shredder residue	14	3	6
(Of mixed automotive and WEEE shredder residue)	(205)	(29)	5

3.4 BFRs in construction plastics

In total, 806 data (including data from 6 ranges) were collected (Table 2). The various samples are presented in Table S2 by group of BFR present or not (last column) and by decreasing mean concentration. The distinction between the PBDE - decaBDE groups on the one hand and HBCDD on the other hand is very clear, with more foams and polystyrenes for the latter group. However, HBCDD concentrations are too low to be flame retardant: the maximum is 0.54%, while 0.8 to 4% is recommended (Arias 2001, Alaee et al. 2003). This may be the index of products derived from recycling a mixture of brominated and non-brominated plastics . The RFB-free group includes paint, wood panels, paper-based insulation, etc. A summary is presented in Table 4. The samples from continental Europe (69 out of

85 samples) have low to very low concentrations (results not shown).

3.5 BFRs in textile equipment

A total of 496 data (including data from 19 ranges) were collected (Table 2). Plastics also cover a wide range (0-300 000 mg/kg-Table S3) presented by group of BFR present or not (last column) and by decreasing average concentration. A first PBDE - decaBDE group without HBCDD can be identified, mostly with non - flame retardant concentrations (at least 5% according to Arias 2001, Alaee et al. 2003). A second and smaller group contains HBCDD with or without PBDEs. The group without BFR is made up of articles that do not seem different from the first two groups.

A summary of concentrations for all samples is presented in Table 43. The average concentration of decaBDE is of 6 500 mg / kg (n = 75), which is above the limit for the products. This result is consistent with the high level observed in automotive seats and foams (Hennebert 2018a, this paper). The average HBCDD concentration is hazardous and above the limit for the products. The plastics of textile equipment should therefore be sorted. Here also, the samples from continental Europe (36 out of 55) have low to very low concentrations (results not shown).

3.6 BFRs in non-food packaging

In total, 65 data (without data ranges) were collected (Table 2). These plastics are exclusively expanded polystyrene (EPS) and extruded polystyrene (XPS) with HBCDD as RFB. The concentrations are presented in Table 4. Concentrations are low, but 31 samples have a HBCDD concentration> 10 mg / kg, the highest being polystyrene packaging laboratory equipment, appliances and printers (Abdallah et al. 2018, FOEN 2017). The maximum HBCDD content exceeds the concentration limit for hazardous waste, and the average is greater than what is allowed for the products. These plastics should therefore be sorted.

3.7 BFRs in food-contact packaging

This point is not the focus of this study but is however presented since, according to some authors, there is evidence of recycling of brominated plastics (maybe from construction insulation foams) in food-contact articles. According to two authors (Rani et al. 2014, Abdallah et al. 2018), six data out of 66 of disposable plate and cup, menu box, takeaway food container, PS cold box, vegetable packaging, and similar, all in expanded polystyrene or extruded polystyrene, have a concentration of HBCDD > 10 mg/kg (1516, 50, 29, 15, 14 and 10 mg/kg). This indicates the usefulness of sorting EPS and XPS (particularly from construction plastics) for bromine and BFRs.

3.8 Other BFRs

Three authors (Rani et al. 2014, Abdallah et al. 2018, FOEN 2017) measured 12 unregulated RFBs nowadays (Table 7) on a total of 144 samples. Only the maximum concentrations are presented. The concentrations are low or close to zero. For e-waste, the highest concentrations are reported by (FOEN 2017). Decabromodiphenylethane (DBDPE) has been a substitute for decaBDE for two decades in plastics of EEE (Wäger et al. 2012). For construction plastics, a rubber sample and a recycled plastic material sample have a dechlorane concentration greater than 64 mg/kg and 17 mg/kg, respectively (Vojta et al. 2017).

TABLE 7: Maximum concentrations in non-regulated RFBs.

Maximum concentration (mg/kg)			Category					
Abbreviation	**Name**	**CAS No**	**EEE/WEEE**	**Vehicles/ ELV**	**Construction**	**Textiles**	**Non-food packaging**	**n**
BTBPE	1,2-Bis(2,4,6-tribromophenoxy) ethane	37853-59-1	150	0	1	0	0	144
DBDPE (DBDPER)	Decabromodiphenylethane or 1,2-bis(pentabromodiphenyl) ethane	84852-53-9	340		0		0	15
DBE-DBCH (TBECH)	Tetrabromoethylcyclohexane (sum of alpha- and beta-)	3322-93-8	0	0	0	0		128
DBHCTD (HCDBCO)	Hexachlorocyclopentenyl-dibromocyclooctane	51936-55-1	0	0	0	1		128
DDC-CO	Dechlorane Plus (sum of anti- and syn-)	13560-89-9	33	0	64	1		129
HBB	Hexabromobenzene	87-82-1	0	0	1	1		128
PBEB	Pentabromoethylbenzene	85-22-3	0	0	0	0		128
PBT	Pentabromotoluene	87-83-2	0	0	0	0		128
TBCO	1,2,5,6-Tetrabromocyclooctane (sum of alpha- and beta-)	3194-57-8	0	0	0	0		128
TBP-BAE (BATE)	2-Bromoallyl-2,4,6-tribromophenyl ether	3728-89-5	0	0	0	0		128
TBP-DBPE (DPTE)	2,3-Dibromopropyl-2,4,6-tribromophenyl ether	35109-60-5	0	0	0	0		128
TBX (pTBX)	2,3,5,6-Tetrabromo-p-xylene	23488-38-2	0	0	0	0		128
Total			**340**	**0**	**64**	**1**	**0**	**1440**

3.9 Total Br and Sb in plastics

A total of 265 data (including data of 78 ranges) have been gathered (Table 8). All these plastics are brominated to some extent, the highest being the EEE/WEEE plastics, followed by vehicles/ELV and textiles plastics, and then with lower concentrations construction plastics and non-food packaging plastics. The WEEE technical specification (CENELEC CLC/TS 50625-3-1) recommends the sorting of plastics batches or flows with mean concentration of total Br > 2 000 mg/kg (measured on a representative laboratory sample). If applied to other plastics, the first four categories of Table 8 should be sorted.

These plastics also contain antimony. Two plastic samples are classified as hazardous based on their concentration of antimony (in bold): "construction - plumbing" at 13 000 mg Sb / kg and "dressing - padding" at 9 922 mg Sb / kg.

3.10 Synthesis of regulated BFRs concentrations and distribution of concentration

The 2 576 data from the literature summarized in Table 4 indicate that the five categories of plastics studied always have at least one BFR at medium or maximum concentration classified hazardous, not allowing the recycling of the products, or having a total bromine content> 2 000 mg / kg, and therefore not recyclable without sorting. A large majority of the data is lower than 1 000 mg/kg or 500 mg/kg. Some distributions are illustrated in Figure 1. The interval of concentration in the histograms is 1 000 mg/kg. The first interval of concentration [0-1 000] mg/kg has been set to the concentration limit for unintentional (re)use in products. The distributions of data are skewed by large values, as illustrated by the significant differences between the median and the mean (Table 4). The same distribution applies to individual scraps (Hennebert 2019). If the last percentiles of scraps are sorted out, the mean concentration falls drastically, and these plastics can therefore be recycled.

4. MANAGEMENT OPTIONS

For plastic recycling, the so-called "brominated fraction" should be separated, either by sorting for total bromine by X-ray transmission on line, or by flotation in a bath with a different density, as practiced today for WEEE (Hennebert and Filella 2017, Leslie et al. 2016, Swerea IVF 2018).

The POP fractions above the concentration limits should be destroyed in incinerators or irreversibly transformed (EU 2016). The incinerators must fulfil the emissions requirements of (chloro- and bromo-) dioxins and furans in the flue gas, and of leachable antimony (Sb_2O_3 is used as a synergist of BFRs in plastics) in the ashes and in the air pollution control residue. Corrosion of the installation by bromine is intense (Buekens and Zhou 2014). Bromine is present partly as HBr and partly as elemental bromine Br_2 (g). HBr is easily neutralized by solid sodium bicarbonate or captured by wet scrubbing with sodium hydroxide, but not Br_2. If not destroyed, POP substances will continue their cycle (Norwegian Climate and Pollution Agency 2012, Leslie et al. 2016). POP substances are found in landfill leachate (Morin et al. 2017). Traces of BFRs have been found in food-contact articles (Puype et al. 2015), together with rare earth elements from electronic goods. Large reviews point out the ubiquity of these substances (Norwegian Environment Agency 2015, 2016, 2017). These legacy substances can hamper the achievement of the compulsory objective of recycling in the EU. This point has been well identified by the European Commission (EC 2017). Finland has suggested that the EU recycling targets for ELV (85% of recycling including scraps and 10% energy recovery) be calculated from the material left after removal of hazardous substances and components (specified in Annex VII of the WEEE Directive 2012/19/EU) (Häkkinen 2016).

The "brominated plastics" are today mainly incinerated in incinerators for hazardous waste, in cement kiln, or in non-ferrous smelters refineries, with special management for corrosion by bromine, for the emission of elemental bromine and brominated dioxins / furans in the fumes, and for the presence of leachable antimony in the ashes, slags and air pollution control residues (Norwegian Environment Agency 2017). A non-ferrous smelter, Umicore, claims 70% recovery of Sb, a critical raw material, according to the EU and the USGS. If the POP concentration limit is not exceeded, they could theoretically be reused as flame-retarded plastics. But in practice, they are refused by manufacturers who have developed precise formulated compounds. They are mainly used as solid recovered fuel (emissions in the fumes should be secured) or landfilled in landfills for non-hazardous waste. For instance, in France, 40% of the ELV plastics are burned in different installations and 29% are landfilled (Deloitte and ADEME 2017).

Chemical recycling by cracking of plastics or solvolysis is up till now hampered by the incomplete separation of brominated compounds from the useful un-brominated

TABLE 8: Concentration of total Br and Sb in plastics of EEE/WEEE, vehicles/ELV, construction, textiles and non-food packaging from literature data (**hazardous waste** [bold], concentration of technical specification for sorting of WEEE plastics *Br> 2000 mg/kg* exceeded [italics]).

Plastics	Br					Sb				
	n	Min	Median	Mean	Max	n	Min	Median	Mean	Max
EEE/WEEE	92	0	145	*13374*	*171000*	32	0	1000	3381	**58900**
Vehicles/ELV	36	0	314	*8197*	*106800*	11	34	975	1521	6020
Construction	9	0	45	*2122*	*9410*	4	103	984	3768	**13000**
Textiles and upholstery	70	0	99	*7175*	*128300*	4	90	944	2975	**9922**
Packaging	6	0	10	1153	*5600*	-	-	-	-	-

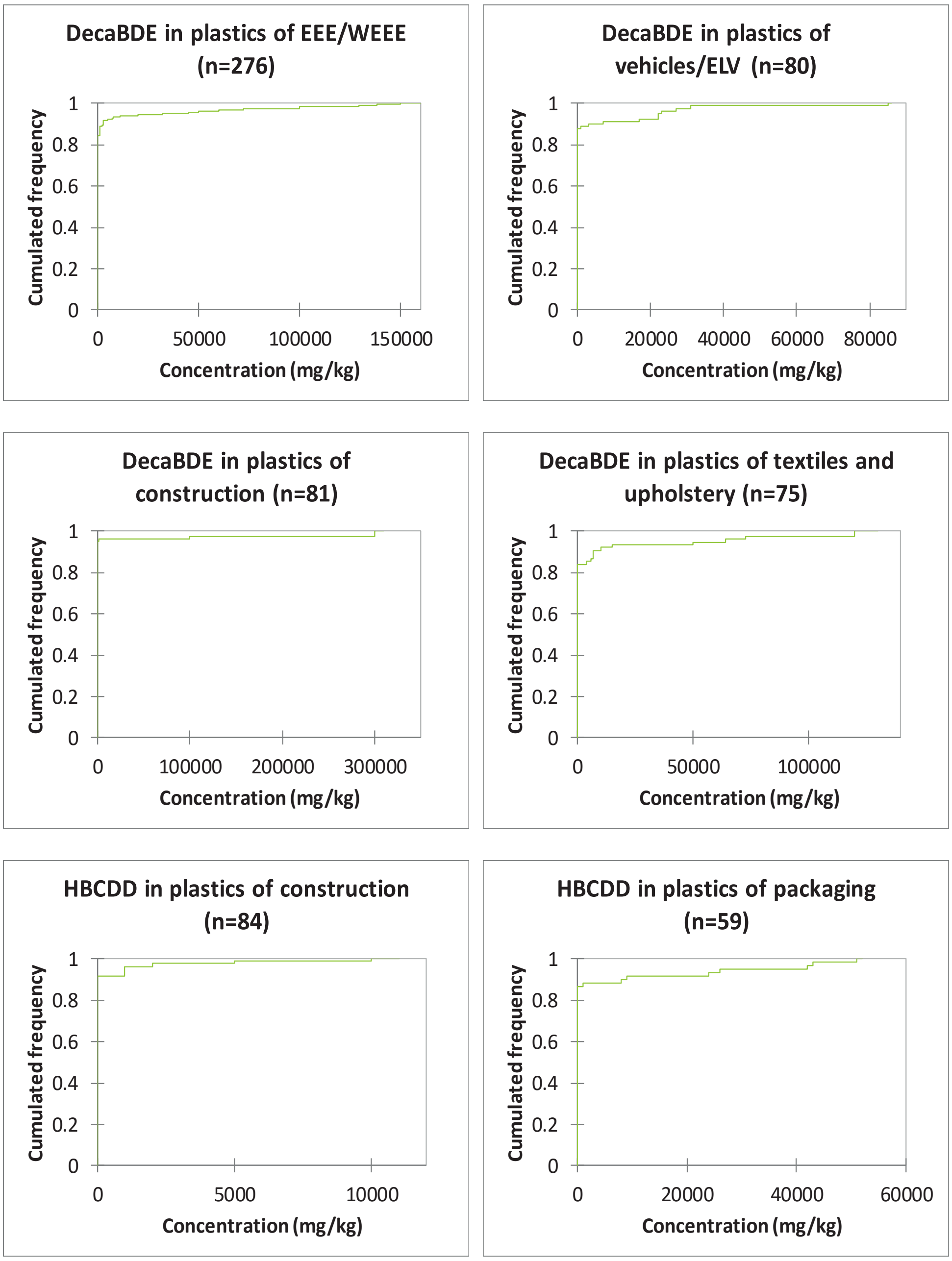

FIGURE 1: Distribution of decaBDE concentrations in plastics of EEE, vehicles, construction and textiles, and of HBCDD in expanded polystyrene foam of non-food packaging from literature data (frequency of data per concentration class of 1000 mg/kg).

feedstock chemicals. The production of syngas or liquid fuel by pyrolysis (heating without oxygen) faces the same obstacle. Exportation is in practice restricted since China decided in January 2018 to stop the importation of unsorted or contaminated plastics. Following others, a pyrolysis technology with recovery of heat, bromine and antimony is claimed at pilot scale in 2018 (Hense et al. 2018). A short synthesis of the advantages and drawbacks of the different management options is presented in Table 9.

5. CONCLUSION

The EU plastics strategy in a circular economy stipulates that plastics containing regulated BFRs must be sorted and managed separately from the non-brominated fraction. Sorting is essential to avoid the uncontrolled dispersion of controlled substances in recycled raw materials.

According to the EU, regulations are in force for each category. These unsorted plastics are hazardous (average or maximum for a BFR), they cannot be reincorporated as such into products, and they exceed 2000 mg Br/kg (operational sorting concentration for WEEE plastics). This study shows that approximately 16%, 12% 8%, 16% and 9% of published concentrations of plastics from EEE/WEEE, vehicles/ELV, building materials, textile equipment and non-food packaging could be greater than 1 000 mg/kg of regulated RFBs. Overall, the results indicate that the sorting

TABLE 9: Management options of brominated and antimoniated plastics.

POP waste? / Technology	Restriction of use (POP, RoHS)?	Hazardous waste?	Recycling as flame-retarded plastics	Chemical destruction	Co-Incineration H waste	Co-Incineration NH waste	Solid Recovered Fuel with flue gas treatment	Landfill H waste	Landfill NH waste	Co-Fuel for cement kiln	Co-Raw material for non-ferrous smelters-refineries (flue gas treatment)	Pyrolysis and post-combustion (flue gas treatment)	Export
Classification													
Yes	-	-		X	X					?	?		
No	Yes	Yes	X			X	X	X?		X	X		
		No	X			X	X		X	X	X		
	No	Yes	X			X	X		X	X	X		
		No	X			X	X		X	X	X		X
Technology													
Recovery	Polymer												
	Heat				X	X	X			X	X		
	Oil											X	
	Gas											X	
	Br										HBr?		
	Sb										70%		
	Metals of non-plastics										X	X	
Drawbacks	C content							X					
	Br2gas				X	X	X			X	X	X	
	Leach. Sb				X	X	X			?	30%?	X	
	Leach. POP							X	X				
	Corrosion				X	X	X			X	X	X	
Technology Maturity level	Routine				X	X	X		X	X	X		X
	R&D											X	
	Laboratory		X	X								X	
	Not practiced		X	X				X				X	

of bromine content for these categories of plastics (except for food packaging) is necessary.

Literature also shows that sorting is effective: data are scarce but sorted ELV plastics have a mean decaBDE concentration of 14 mg/kg. The efficiency of sorting was confirmed in France with WEEE plastics in 2014 and 2015 (Hennebert and Filella 2017). As the plastics of construction and demolition waste must be sorted in the EU and that separate collection of textiles is foreseen at the latest in 2025 (EU 2018), all these plastics (excepted food-packaging) will have to be sorted to weed out regulated substances (PBDE, HBCDD) in a circular economy. The amount to be sorted could reach 40% of the amount of plastics used in the EU, representing 20 million of tons per year (calculated from PlasticsEurope 2018). The amount that could not be recycled is a few percent of that mass. It must be emphasized that the samples of construction plastics and textile and furniture plastics from continental Europe have low to very low concentrations.

To conclude, it should be checked by analytical campaigns whether the different (sorted) plastics fractions have a POP waste status or not, and whether the major actual management practices of energy recovery and landfilling are in line with the EU regulations. Research in technology to recover energy, bromine and antimony is active and promising on a pilot scale.

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SUPPLEMENTARY INFORMATION

TABLE S1: Concentrations of some brominated flame retardants in car parts, end-of-life vehicles, automotive shredder residue (ASR) and post shredder treatment fractions – unweighted mean concentrations from literature data.

Mean Concentration (mg/kg)	PBDE	of which DecaBDE	HBCCD	TBBPA	All
Car (117 data)	3212	5432	431	27	2651
1990-1994 - Printed circuit board	77	200	10	26	53
1995-1999 #1 - Printed circuit board	4	10	10	20	8
1995-1999 #2 - Printed circuit board	10	10	10	87	26
2000-2004 - Printed circuit board	27	50	10	20	22
Airbag	27	50	50	20	30
Car	128	128			128
Car interior	8	14			8
Car seat cover	256	256			256
Car seats	66	66			66
European ELV parts	3469	5751			3469
Interior 1	5677	17000	50	20	3420
Interior material 1	9				9
Interior material 2	137				137
Interior Mazda 1998	52	52			52
Interior Pontiac 1997	18	18			18
Luggage compartment	27	50	50	20	30
Printed circuit boards sample 1	2				2
Printed circuit boards sample 3	2				2
Printed circuit boards sample 4	2				2
PUF from old car seats	1	1			1
PUF from old car seats, high contamination	900				900
PUF from old car seats, low contamination	1				1
PUF from US car seats	24	9			24
PUF Pontiac 1997	8676	522			8676
PUR foam for automotive applications	40000				40000
Radiator, outer	27	50	50	20	30
Rail vehicles	85000	85000			85000
Seat cover	9020	27000	50	20	5426
Seat cover material	51				51
Seat cover Mazda 1998	22700	22700			22700
Seat cover Pontiac 1997	8625	22500			8625
Soundproofing 1	27	50	4400	20	900
Soundproofing 2	2343	7000	50	20	1420
US/Asian ELV plastic parts	21	37			21
ELV (46 data)	925	3253	337		797
ELV foams	111	33	1		74
ELV upholstery	4053	8101	842		2983
Hyundai - foam	0	0	0		0
Škoda - foam	0	0	0		0
ASR (51 data)	179	386			179
ASR	254	429			254
Autoshredder Waste	44	44			44
Input Car shredder residue	6	17			6

Light ASR (foam and textile)	21				21
Mixed light plastic	3				3
Mixed SR	255	413			255
PST (33 data) (ELV and WEEE)	44	101			44
1,1 < density < 1,3	364	810			364
1,1<d<1,3 (black hard)	1	2			1
1,1<d<1,3 (black soft)	2	6			2
1,1<d<1,3 (coloured)	0	1			0
1,1<d<1,3 (white/grey)	5	3			5
d<1,1 (hard plastic)	0	0			0
density (0 - 1)	10	27			10
density < 1,1	2	6			2
Fiber fraction (foam)	43	113			43
Input for PST	10	29			10
Total (247 data)	**1623**	**3102**	**386**	**27**	**1447**

TABLE S2: Concentrations of some brominated flame retardants in plastics of construction - Unweighted average concentrations from bibliographic data.

Mean concentration (mg/kg)	PBDE	of which DecaBDE	HBCDD	TBBPA	Mean PBDE HBCDD TBBPA	n	Interpretation
epoxy adhesive	300000	300000			300000	1	with PBDE - decaBDE
electrical insulation	200000	200000			200000	2	
polyurethane foam (PUR)	130000				130000	4	
construction 2	4799				4799	1	
moisture-resistant membrane / film	1000	1000			1000	1	
pipe insulation	63	626	0		57	11	
recycled plastics	2	18	1		2	11	
phenolic foam insulation	0	4	0		0	11	
foam insulation	0	1	0		0	33	
blue sealing foam	0	0	5400		491	11	with HBCDD
expanded polystyrene (EPS)	0	0	1995	0	816	22	
pale mounting foam	0	0	832		76	11	
air conditioning - aluminum foil	1	6	545		50	11	
polystyrene	0	0	469		43	11	
polystyrene construction	0	0	469		43	11	
air conditioning - inner sheet	0	0	412		37	11	
fiber network	0	0	250		23	11	
polystyrene board	0	0	127		12	11	
extruded polystyrene (XPS)	0	0	28	0	10	14	
air conditioning - cellophane sheet	0	0	16		1	11	
drywall	0	0	9		1	11	
HARDSIL NT insulation	0	0	8		1	11	
air conditioning - fiberglass foam	0	0	8		1	11	
yellow mounting foam	0	0	3		0	11	
green mounting foam	0	0	2		0	11	
polyacrylate material	0	0	1		0	11	
laminate plastic flooring	0	0	1		0	11	
window corner cap	0	0	0		0	11	without BFR

heat exchanger	0	0	0		0	11	
insulation board	0	0	0		0	11	
unknown (construction site)			0		0	1	
insulation aluminum foil	0	0	0		0	11	
paper insulation	0	0	0		0	11	
exterior paint	0	0	0		0	11	
wood fiber insulation	0	0	0		0	33	
rubber	0	0	0		0	11	
plaster	0	0	0		0	11	
asphalt insulation	0	0	0		0	11	
formica	0	0	0		0	11	
oriented strand board (OSB)	0	0	0		0	55	
construction 1	0				0	1	
chipboard	0	0	0		0	55	
green sealing foam	0	0	0		0	11	
water resistant paint	0	0	0		0	11	
window finishing tip	0	0	0		0	22	
cotton insulation	0	0	0		0	11	
mastic	0	0	0		0	22	
paper insulation of recycled beverage cartons	0	0	0		0	11	
decorative polystyrene	0	0	0		0	11	
insulation hemp rope	0	0	0		0	11	
Drain pipe	0	0	0		0	11	
foam	0	0	0		0	11	
plank of wood	0	0	0		0	22	
brown chipboard	0	0	0		0	11	
elastic linoleum	0	0	0		0	22	
linoleum	0	0	0		0	22	
blown cellulose insulation	0	0	0		0	11	
plaster with fire retardant foam	0	0	0		0	11	
blow insulation made of paper	0	0	0		0	11	
building polystyrene panel	0	0	0		0	11	
drinking water pipe	0	0	0		0	11	
Total	**1713**	**8662**	**317**	**0**	**1555**	**806**	

TABLE S3: Concentrations of some brominated flame retardants in textile plastics - Unweighted average concentrations from literature.

Mean concentration (mg/kg)	PBDE	of which DecaBDE	HBCDD	TBBPA	Mean PBDE HBCDD TBBPA	n	Interpretation
commercial decaBDE treated polyester upholstery fabrics used in the manufacture of curtains	120000	120000			120000	2	with PBDE - DecaBDE
PUR foam for upholstered furniture	41040				41040	11	
various textiles	39850	39850			39850	11	
adhesive layer of reflective tapes	30000	30000			30000	11	
polyester	28131	120000	3		25318	11	
PUR foam for mattresses	25000				25000	2	
textile 2	20003				20003	12	
textile 1	11843				11843	11	
foam padding	7023	7023			7023	2	

window blinds	4799	4799			4799	11	
several carpets	907	1810	7		607	44	
several mattresses	115	230	3		78	12	
carpet	85	85			85	12	
velvet (70-80 g / m2)	27	27			27	22	
flat woven (30-80 g / m2)	21	21			21	22	
cotton (30-40 g / m2)	13	13			13	11	
tents	2	2			2	12	
insulation / carpet	1	2			1	11	
polyester	1	5	42500		4251	46	with HBCDD and with or without PBDE - decaBDE
furnishing	9999	19953	15050		11683	1	
curtain	0	0	8333		1087	2	
furniture foam filling	1060	2119	2275		1465	12	
curtains	7	14	15		10	4	
Persian rug	0	0	1		0	11	
treated textile	0				0	11	Sans RFB
Red carpet	0	0	0		0	10	
gray carpet	0	0	0		0	20	
blanket	0	0	0		0	2	
green carpet	0	0	0		0	5	
tablecloth	0	0	0		0	11	
foam	0	0	0		0	22	
textile material	0	0	0		0	1	
blue carpet	0	0	0		0	11	
brown carpet	0	0	0		0	1	
coconut fiber	0	0	0		0	1	
textile cover	0	0	0		0	11	
stuffing material	0	0	0		0	44	
pillow	0	0	0		0	1	
plush	0	0	0		0	1	
insulation of the textile bottle	0	0	0		0	33	
wardrobe	0	0	0		0	2	
bed-cover	0	0	0		0	2	
textile	0	0	0		0	1	
Total	**2080**	**6511**	**3465**		**2244**	**496**	

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OZONE PRETREATMENT FOR THE ANAEROBIC DIGESTION OF ORGANIC SOLID WASTE

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ABSTRACT
The pretreatment of organic solid waste plays a key role in achieving the highest degree of valorization within the anaerobic digestion processes. This work focuses on the use of ozone, discussing its effects, together with particle size and total solid content of waste, on the anaerobic biodegradability of the organic fraction of municipal solid waste (OFMWS). A Design of Experiment (DOE) strategy was used to identify statistically significant factors for further investigation. Experimental results showed that ozonation increased methane production, with the best results being obtained for samples characterized by the highest total solid content. The surplus methane generated by ozonated samples may also result in favourable net energy gain. These outcomes highlight the effectiveness of ozonation when applied in the pretreatment of OFMSW destined to anaerobic digestion and address the need for an energy balance to assess the competitiveness of this technology on an industrial scale.

1. INTRODUCTION

Anaerobic digestion is a mature technology used widely in the treatment of organic residues. The range of substrates includes high-strength wastewater, sewage sludge as well as a variety of organic solid waste (Xu et al., 2018; Li et al., 2019). The main aim is to generate energy from the biogas produced. Consequently, anaerobic digestion has gained a key role in contributing to the transition from a fossil fuel based economy to a bio-economy (De Vrieze et al., 2018). It can convert organic substrates, via biological routes, into a variety of value-added products, such as lactic acid, polylactic acid (PLA), succinic acid, isobutene, acrylic acid, adipic acid, ethylene and polyethylene (Dahiya et al., 2018) and energy carriers, including methane.

Nevertheless, the anaerobic digestion of solid waste still presents a number of challenges, related to the complex route to hydrolysis of organic macro-molecules into soluble compounds for microbial attack. As a result, the study of pretreatments has attracted great attention and the use of physical, chemical as well as biological processes has been assessed extensively for application to OFMSW (Ariunbaatar et al., 2014a; Cesaro and Belgiorno, 2014).

Among the chemical pretreatments options, ozonation has been investigated only at laboratory scale. The larger scale application of this technology does not appear competitive to other pretreatments, due to the need to operate at low ozone doses (Cesaro and Belgiorno, 2013). Successful application has been demonstrated for the treatment of sewage sludge (Sievers et al., 2004).

Ozone is a strong oxidant, which decomposes into radicals and reacts with organic substrates both directly and indirectly, via the hydroxyl ion. Cesaro and Belgiorno (2013) highlighted that ozone doses as low as 0.16 g_{O3}/g_{TS} can improve both the solubilisation and the anaerobic biodegradability of OFMSW. However, increasing ozone doses did not lead to higher OFMSW solubilisation effects in terms of COD: the higher ozone doses resulted in a reduction in biogas compared to untreated samples (Cesaro and Belgiorno, 2013). Ariunbaatar et al. (2014b) showed that the optimal ozone dose to enhance methane generation is highly dependent on the relative presence of sugars, lipids and proteins in the substrate. It was found that direct ozone oxidation can destroy the easily fermentable sugar, thus resulting in biomethane loss; conversely, the indirect reaction of ozone can cause the degradation of complex organic compounds such as lipids and proteins, thus enhancing biomethane generation.

These studies highlight that ozonation can be attractive as a pretreatment for organic substrates containing highly complex and difficult-to-degrade components, like OFMSW. Nevertheless, the biogas yields should take into account not only the composition of waste, but also its

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physical characteristics in terms of particle size and solids content. These can influence the effects of ozonation, promoting differential solubilization of OFMSW components.

This aspect is even more important for process scale up, since at an industrial scale organic waste is usually subjected to mechanical pretreatments, which focuses on size reduction using shredding devices. More recently, conventional shredding processes are being replaced by press-extrusion treatments that split the waste into liquid and solid fractions. The liquid fraction may account up to 85% w/w of the input mass of OFMSW (Novarino and Zanetti, 2012). Due to its high moisture content this can be conveniently diverted to anaerobic digestion; the solid fraction, that still contains organic material, can be sent to either composting or other recovery processes, according to its chemical-physical characteristics (Hjorth et al., 2011).

Another fundamental aspect in determining biogas yields is the solid content in the anaerobic digester. Although the production of methane is closely related to the volatile solid (VS) content of the substrate for anaerobic digestion, the total solids (TS) also affect process yields. Abbassi-Guendouz et al. (2012) evaluated the role of the TS content on anaerobic digestion. They showed that the total methane production slightly decreased as TS concentrations increased from 10% to 25%, with further increase of TS up to 30% resulting in different behaviors. This suggested that a 30% TS content corresponded to a solids threshold, above which methanogenesis was inhibited. Similarly, An et al. (2017) found that the ultimate biogas yield of sludge firstly improved and then decreased with TS increasing up to 10%. The outcomes of both studies highlighted the limited mass transfer occurring under high TS conditions. This means that a proper balance among anaerobic digestion phases is needed to avoid the inhibition of the digestion process.

Aim of this study was to assess the effects of ozone dose, substrate particle size and total solid content on the anaerobic biodegradability of OFMWS samples. A Design of Experiment (DOE) strategy was adopted and the factors investigated were varied in a 2^3 factorial design between "low" and "high" levels, selected according to previous studies.

2. MATERIALS AND METHODS

2.1 Preparation of organic waste samples

For experimental purposes, OFMSW samples were prepared in the laboratory, to ensure their reproducibility in the different test runs. The composition and the chemical-physical characteristics are given in Table 1.

The samples were then ground using a bench scale shredder model M20 (Universal IKA) and sieved to ensure that the particle size ranged between 1 and 3 mm. Some samples were further processed to reduce the particle size below 1 mm, to improve the homogeneity and reach the consistency typical of the press-extruded wet fraction (similar to a jam), as previously described (Novarino and Zanetti, 2012). The TS content of each sample was adjusted to either 10% or 15% by adding distilled water. These values were selected in order to keep operating conditions closer to those of wet digestion, as previously (Cesaro and Belgiorno, 2013).

TABLE 1: Composition by weight and main chemical-physical characteristics of the OFMSW samples.

Composition by weight	
Fraction	**[% w/w]**
Fruit and vegetable	78.6
Pasta and rice	4.9
Bakery products	6.4
Meat and fish	8.2
Dairy products	1.9
Chemical-physical characteristics	
Parameter	Value
TS [%]	23.52 ± 0.27
VS [%TS]	93.13 ± 0.12
sCOD [mg/L]	16250 ± 1573

2.2 Ozone pretreatment

The ozonation of OFMSW samples was undertaken with a UV generator (model Ozone - Procom srl), using air supplied from a compressor. The ozone was introduced at the bottom of a glass reactor containing waste samples at a 0.16 g_{O3}/g_{TS} ozone dose, selected on the basis of our previous work (Cesaro and Belgiorno, 2013).

Exhaust gas was extracted at the top of the reactor and passed through a Drechsel trap, filled with 200 mL of 2% KI solution, in order to neutralize residual ozone. The methodology met with the Semi-Batch Standard Method 2350 US-EPA to determine the ozone demand.

2.3 Analytical set up

The main chemical-physical characteristics of the OFMSW samples were determined. Soluble Chemical Oxygen Demand (sCOD), total solid (TS) and volatile solid (VS) were evaluated according to Standard Methods (AWWA-APHA-WEF, 1998). The Standard procedure to determine the soluble COD (sCOD) was applied on each sample after centrifugation and filtration (<0.45 mm).

The experimental activity was carried out following the DOE strategy. To this end, ozone dose, waste particle size and total solid content were varied in a 2^3 factorial design between a "low" (-) and a "high" (+) level, to evaluate eight pretreatment combinations (Table 2).

The response of the pretreated OFMSW samples was evaluated by their anaerobic biodegradability. This was assessed by batch tests, carried out under mesophilic conditions, using 500 mL flasks with 400 mL working volume. Digested sludge was used as inoculum and added to the substrate in a ratio of 0.5 g_{VS} substrate/g_{VS} inoculum. The seed sludge (TS: 3.01±0.35; VS: 57.81±4.84%TS) was collected from the anaerobic digester of a conventional wastewater treatment plant in Salerno (Italy) and incubated at 35°C for 5 days before use in order to reduce intrinsic gas production. No trace nutrients were added, nor pH of each batch was adjusted. After feeding, headspaces of the bottles were flushed with nitrogen gas and then

TABLE 2: Combination plan for experimental tests.

Experimental run	Ozone dose [gO_3/gTS]	Total solid [%]	Particle size [mm]
1	0.16	**15**	**1 - 3**
2	0.16	10	1 - 3
3	0.16	15	< 1
4	0.16	10	< 1
5	0	15	1 - 3
6	0	10	1 - 3
7	0	15	< 1
8	0	10	< 1

sealed with rubber septa. Daily methane production was determined by a water displacement method (Esposito et al., 2012), and the cumulative production over 21 days was assessed. Results were expressed as the total volume of methane produced during the digestion period and calculated in the normal state (273 K and 1013 hPa), per mass of added substrate, expressed in terms of VS (i.e. L_{CH4}/kg_{VS}).

All the analytical determinations were performed in triplicate and average values have been used in the subsequent evaluation.

2.4 Evaluation of the net energy gain

The competitiveness of ozonation as pretreatment for anaerobic digestion should take into account the process efficiency as well as the energetic viability. In this study the latter was referred to the potential net energy gain (E_{net}), which is the difference between the extra energy produced ($E_{produced}$) and the energy demand ($E_{pretreatment}$) for the pretreatment application (equation 1):

$$E_{net} = E_{produced} - E_{pretreatment} \quad (1)$$

The extra energy produced from the anaerobic digestion of ozonated substrates was calculated using equation 2:

$$E_{produced} = V_{CH4} * E_{CH4} * \eta \quad (2)$$

where:

V_{CH4} is the surplus methane volume from ozonated substrates;

E_{CH4} is the methane energetic potential, assumed at 9.97 kWh/m^3;

η is a conversion factor (0.38 for electrical energy; 0.45 for thermal energy).

The energy demand for ozone pretreatment is closely related to the detailed operation of device. For devices using air, this can vary across the range 2.5-40 kWh/kgO_3, depending on the generation efficiency (Ariunbaatar et al., 2014b). In this study a high-energy efficiency ozone generator was used, and the lower value of 2.5 kWh/kgO_3 was selected to calculate energy consumption.

3. RESULTS AND DISCUSSION

The preparation of the substrate for its use in anaerobic digestion is a fundamental step to optimize methane production. The hydrolysis of complex macro-molecules into soluble compounds depends on the nature of the substrate (Srisowmeya et al., 2020) and it has been largely claimed as the process rate limiting step in the anaerobic digestion of OFMSW (Kondusamy and Kalamdhad, 2014; Kumar Pramanik et al., 2019; Ren et al., 2018; Ye et al., 2018). Therefore, any strategy able to optimize this step results in either enhanced methane production or the speed up of the anaerobic bioconversion process.

Figure 1 shows the cumulative biomethane volumes produced after 21 days during the anaerobic digestion of OFMSW samples pretreated under different conditions.

Experimental results show that ozonation resulted in an increase in methane production ranging between 13 and 95%, which is in good agreement with the findings of previous studies (Cesaro and Belgiorno, 2013; Ariunbaatar et al., 2014). The OFMSW samples were primarily fruit and vegetable peels, which are quite hard to degrade, due to their lignocellulosic components (Pagliaccia et al., 2019).The effects of ozone on this type of waste components was to promote more extensive degradation of the more resistant components, as confirmed by the increase in the ultimate production of methane after 21 days.

Among ozonated substrates, the best performance in terms of methane generation was found for the samples characterized by the lower TS content, despite the waste particle size.

A similar trend was also observed for the experimental studies on un-ozonated samples. The effect of total solid content on the performance of anaerobic digestion has been studied for a wide variety of substrates. As previously highlighted, several authors (Abbassi-Guenduz et al., 2012; An et al., 2017) investigated the role of TS content on solid waste anaerobic digestion: they found a slight decrease in methane production for TS increase up to a substrate-dependant threshold limit value, and attributed this outcome to mass transfer limitation. However, the increase with TS content needs to further account for microbial community change in the digester. Yi et al. (2014) compared the microbial community structure of mesophilic anaerobic digesters treating food waste with TS contents ranging from 5% to 20%. They observed that the different performances in the digesters, in terms of VS reduction, reflected the relative abundance of the main microbial phyla. The same authors pointed out that the bioconversion of organic matter into the precursors for methane generators was the result of the role that each microbial population played in either the hydrolytic step or the acidogenic one. The experimental outcomes reported here are in good agreement with previous reports: other conditions being equal, a lower TS content did not hinder the biological conversion of the substrate, resulting in a higher ultimate methane production potential after 21 days. Depending on the operating conditions in terms of ozone supply and particle size range, the increase in the specific methane production after 21 days was found to be in the range 10-40%.

The particle size influenced methane production as under the same ozonation and TS conditions, samples with the coarser particles (1-3 mm) provided the lower methane production. Izumi et al. (2010) had already pointed out that, when reducing the average food waste particle size from

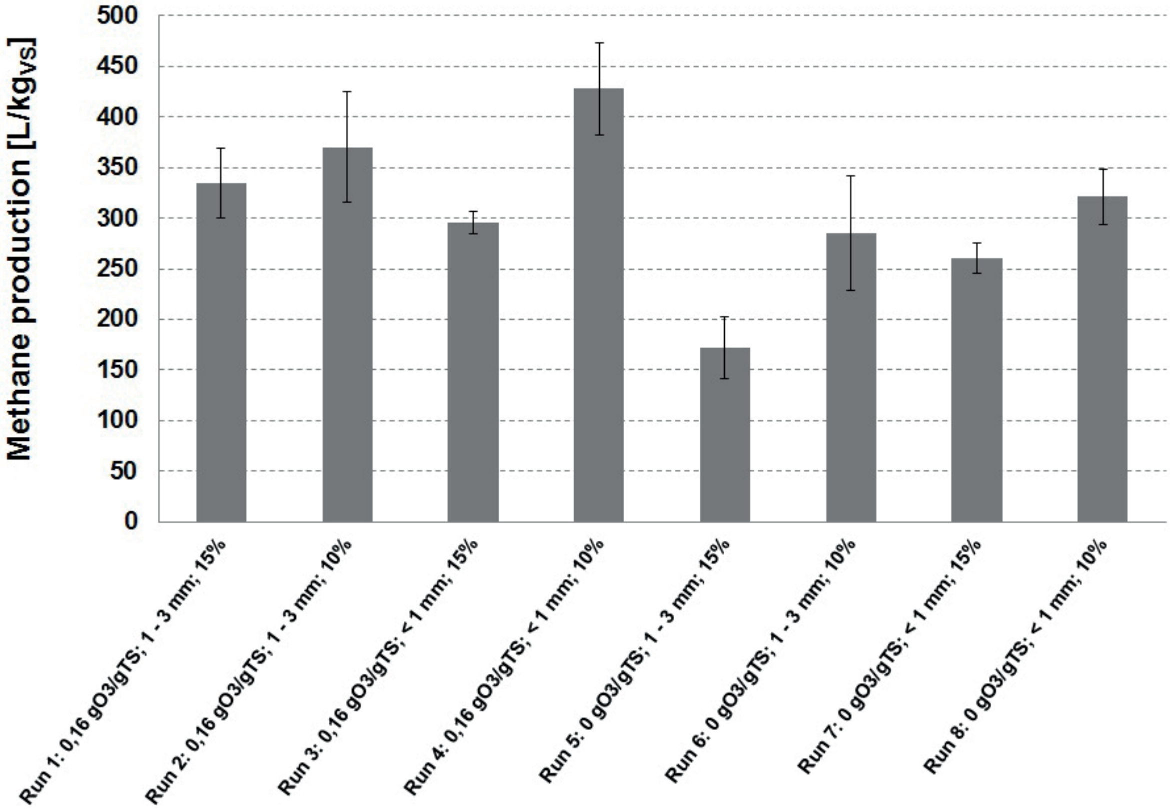

FIGURE 1: Cumulative methane production in 21 days for the investigated operating conditions.

0.888 to 0.715 mm, a 28% methane increase occurred. Nevertheless, a further reduction of waste particle size can determine the acidification of the digestion medium due to the accumulation of volatile fatty acids and a subsequent inhibition of the process that, in turn, can lead to lower methane generation. The authors proved that, although waste size reduction results in the increase of the specific surface area for microbial attack, an excessive comminution can produce the formation of reaction intermediates that are not properly converted into methane and can create inhibitory conditions for the methanogenic bacteria in the digester.

The results of the present study are consistent with previous findings, with an 11-34% increase in methane generation from the press-extruded samples. However, the inverse relationship between waste particle size and volume of methane generated was not fully valid for the ozonated samples with the higher TS content. The comparison of test run 1 (0.16 gO_3/gTS; d = 1-3 mm; TS = 15%) and test run 3 (0.16 gO_3/gTS; d < 1 mm; TS = 15%) highlights that the methane generation from waste samples with the coarser particles was 12% higher than that from press-extruded samples. This is likely to be related to the combined effects of ozonation and press-extrusion on the more concentrated samples, which could result in a significant qualitative change in the substrate making it more suitable for microbial degradation kinetics.

The analysis of variance at a 95% confidence level was also performed on the anaerobic biodegradability trials. The F-test was used to estimate the statistically significant factors (p-value < 0.05), which were found to be both the ozone dose and the total solid content. The combination of the three factors investigated was found to be significant as well, as shown in Table 3.

The particle size could not be considered a statistically significant factor: as the p-value was higher than 0.05, the analysis of variance suggests that the variation of the experimental response associated to the corresponding variation of this factor (particle size) can be attributed to conditions other than the variation of the factor itself.

It is worth pointing out that such an outcome could reflect the choice of the values selected for this study. The assumption that smaller particle dimensions leads to an overall improvement of anaerobic digestion yield is not necessarily correct. Zhang and Banks (2013) studied the impact of different particle size distributions on anaerobic digestion of OFMSW and found that any increase of the specific methane generation occurred when the waste particle size was reduced from 4 to 2 mm. Nevertheless, the same authors observed the increase in methane generation rate during the first 15 days of process. However, this evidence was considered not relevant, since the retention time of the substrate within anaerobic reactors often exceeds 15 days.

Experimental results highlight that ozonation is a viable pretreatment option to improve the generation of methane from OFMSW. However, as ozonation has high-energy consumption, its operation as OFMSW treatment prior to anaerobic digestion is only competitive if the energy produced from the surplus biogas can either balance or ex-

TABLE 3: Results of the analysis of variance (ANOVA) and p-values.

	Sum sq	Mean sq	F value	Pr (>F)
A	62831.69	**62831.69**	**52.10381**	**9.081 E-05**
B	77834.03	77834.03	64.54465	4.236 E-05
C	6263.535	6263.535	5.1941	0.052148
A:B	274.1508	274.1508	0.227342	0.646254
B:C	389.9638	389.9638	0.323381	0.585188
A:C	2881.274	2881.274	2.389325	0.160750
A:B:C	22833.48	22833.48	18.93489	0.002441

where: A is the ozone dose; B is the TS content; C is the particle size

ceed the energy consumption to run the ozonation device.

In this view, the potential net energy gain was estimated for all the operating conditions considered in this study and the results are summarized in Table 4.

If considering the potential net production of electric energy, it was possible to achieve an increase of 26 kWh/t_{OFMSW}. This only occurred for the samples characterized by the coarser particles (1-3 mm) and the higher TS content (15%), as the application of ozone on this kind of substrate almost doubled the production of methane. For the thermal energy, the potential net gain was found to be in the range 5 - 43 kWh/t_{OFMSW}. In this case, the best performance was obtained for the same kind of samples providing the only net electric energy gain.

It is worth pointing out that the evaluation of net energy was based on experimental results obtained at lab scale as well as in batch conditions: these are likely to change when applying ozone pretreatment on larger scale, in a continuously fed reactor. Morever, this study did not evaluate further aspects contributing to the production of methane from anaerobic digestion, related to the operating conditions of the biological process itself, such as the organic loading rate and the hydraulic retention time.

The economic viability for OFMSW pretreatment does not only depend on the energy balance, despite its key role. The higher degree of conversion of the organic matter into methane would also reduce the amount of digestate to be handled at the end of the anaerobic process, providing additional savings. Moreover, the oxidative potential of ozone could further promote the improvement of digestate quality, with particular reference to the presence of persistent organic pollutants, like polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs) and pesticides. These compounds may enter the anaerobic digester together with the OFMSW and concentrate in the digestate due to their poor biodegradability. The application of ozonation in improving the quality and degradability of benzo[a]pyrene-contaminated digestate was investigated by Cesaro et al. (2019). The key finding was the dependence of the integrated ozone/anaerobic digestion process on the biological stabilization extent of the digestate.

All these aspects have to be considered to evaluate the techno-economic competitiveness of ozonation for the valorization of OFMSW destined to anaerobic digestion, to properly promote its scale up.

4. CONCLUSIONS

This study evaluated the potential of ozonation as pretreatment of the organic fraction of municipal solid waste (OFMSW) for anaerobic digestion and influence of particle size and total solid (TS) content.

Experimental results pointed out that an ozone dose of 0.16 g_{O3}/g_{TS} can improve the specific methane generation in the range 13-95%. Although the greatest specific methane volumes were obtained for samples characterized by a 10% TS content, the highest increase was obtained for the samples with 15% TS: in this case, the methane production was 95% higher than the one obtained for the untreated samples with the same characteristics in terms of TS content and particle size distribution. Such increase was also found to result in a positive net energy gain, with respect to either electrical or thermal energy. These outcomes suggest that ozonation can be an attractive OFMSW pretreatment, suitable to pursue the increase in TS content in anaerobic reactors. Conversely, the substrate size distribution was not found to be a statistically significant factor.

Experimental results are promising, but the overall competitiveness of a combined mechanical/ozonation treatment prior to OFMSW anaerobic digestion should take into account the economic affordability of this technology. Further tests at larger scale are thus necessary to optimize the operating conditions of the integrated ozonation/anaerobic digestion process for OFMSW treatment. These tests should also provide the required data to carry out an economic feasibility assessment for its reliable scale up.

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TABLE 4: Results of the potential net energy gain assessment for the investigated pretreatment conditions.

Particle size (d) and TS content	d = 1-3 mm; TS = 15%	d = 1-3 mm; TS = 10%	d < 1 mm; TS = 15%	d < 1 mm; TS = 10%
Methane surplus from ozonation at 0.16 gO_3/gTS [m^3/tOFMSW]	23.0	8.1	4.9	10.1
Electric energy surplus [kWh/tOFMSW]	86	30	18	38
Thermal energy surplus [kWh/tOFMSW]	103	36	22	45
Energy consumption for ozonation [kWh/tOFMSW]	60	40	60	40
Potential electric energy gain [kWh/tOFMSW]	26	-10	-42	-2
Potential thermal energy gain [kWh/tOFMSW]	43	-4	-38	5

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COMPOSTING OF STARCH-BASED BIOPLASTIC BAGS: SMALL SCALE TEST OF DEGRADATION AND SIZE REDUCTION TREND

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ABSTRACT
In Italy, the majority of bioplastic bags used in food waste collection is made of starch-based biopolymer. The compostability of this material in a full-scale plant remains to be demonstrated, largely due to the fact that bioplastic bags are screened and removed together with conventional plastic bags during pre-treatment steps. The present research was performed on a small scale to study the degradation of starch-based bioplastics during composting. Evolution of the physical and chemical parameters of the material was evaluated by means of Fourier Transform Infrared (FTIR), experimental mass loss and granulometric trend. The results obtained suggested that fragmentation (physical size reduction of the material) occurred mainly during the thermophilic phase, while biodegradation (breakdown by microorganisms of an organic chemical into simpler, innocuous compounds) occurred during the curing phase. Based on the monitored parameters (TS, VS, pH, C/N and RI_4), the composting process of the waste matrix ended after 55 days, but the degradation of bioplastics failed to achieve the regulatory standards for assessment of compostability (≤ 10% sized > 2 mm). Experimental data revealed a linear trend for the fragmentation process and a duration of 100 days would be required to meet regulatory requirements.

1. INTRODUCTION

The management of plastics continues to present a series of challenges worldwide (Hahladakis et al., 2018). The magnitude of the problem is highlighted by the finding published by ISPRA (ISPRA, 2019) that plastic waste represents 30% of total waste generated yearly. One of the main waste streams is constituted by plastic bags, a serious environmental concern worldwide due to the enormous quantities produced and low recovery rate. However, production and use of plastic bags is achievable by political means: by banning plastics or taxing the production and use of plastic bags whilst promoting alternatives such as paper or bioplastic bags (e.g.: EU, 2018; Italian Legislative Decree 123/20179) (Byun and Kim, 2013).

Bioplastics, characterised by production from renewable sources (bio-based) and biodegradability (biodegradable), represent an effective and sustainable alternative to traditional plastics (Yates and Barlow, 2013).

Indeed, bio-based plastics are deemed environmentally-friendly materials (Karana, 2012), being produced by means of a cleaner production cycle than petrol-based plastics. Moreover, bioplastics may potentially be suitable for reintroduction into the soil following decomposition (in the case of biodegradable materials)

However, the biodegradability of bioplastics is an important issue as products, either accidentally or on purpose, frequently end their life cycle in the environment (soil or water) (Shruti and Kutralam-Muniasamy, 2019). Indeed, the term biodegradability relates in particular to the environmental context in which bioplastics are disposed, with some commonly used bioplastics being biodegradable through composting, in accordance with EN 13432 (EN 13432, 2000). Importantly, the characteristics required to allow the treatment of bioplastics by means of composting are as follows: biodegradation relates to the breakdown of an organic chemical compound by microorganisms, whilst compostability to the suitability of a packaging material to undergo biodegradation in a composting process. To attest this suitability, following assessment of a fragment of the

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material, no more than 10% of the initial material should fail to pass a 2 mm sieve at the end of the process; thus, a 2 mm dimension is the threshold below which residues are considered assimilable compost in accordance with current regulation.

Biological processes in composting are carried out by a consortium of various microorganisms (bacteria, actinomycetes, fungi, yeasts, etc.) and determine the degradation of organic matter accompanied by release of thermal energy and consequent temperature increase, in many cases exceeding 55°C. Experiments conducted on the degradation of bioplastics through composting have yielded at times divergent results. (Emadian et al., 2017), (Mohee et al., 2008). Balaguer et al, 2016, (Balaguer et al., 2016) observed an effective action in terms of degradation of bioplastics, while others reported only 26.9% of degradation due to the presence of lower temperatures (Mohee et al., 2008).

When subjected to unfavourable composting conditions (mainly low temperatures and insufficient composting time), bioplastics may not disappear completely but merely break down into microscopic pieces. Therefore, the difficulty of using the compost on cultivated fields may impact negatively on market demand for the product.

Currently, the majority of industrial composting plants are not equipped to receive bioplastic bags, as these are selected by pre-treatment and removed from the composting line together with conventional plastic bags. As a consequence, numerous aspects of bioplastic composting remain to be ascertained: how initial size affects the degradation process; whether the time frame applied in food waste composing is sufficient to comply with the standards for bioplastic size reduction; and the fate of microplastics generated once treatment has been completed.

Starch-based bioplastic bags are currently the most widely used carrier bags in Italy. Use of these bags is undoubtedly destined to increase further in the future, particularly in view of the strict limitation of single-use plastics imposed by the European Circular Economy package (Package., 2018) (Hahladakis and Iacovidou, 2018). The biodegradation of starch-based film bioplastics under composting has been studied by previous authors in different conditions, generally at lab scale and with constant temperature and humidity (Ruggero et al., 2019). A degradation ranging from 30 to 45% was found in mesophilic conditions (Mohee et al., 2008) (Accinelli et al., 2012) (Massardier-Nageotte et al., 2006), while if composting is carried out with a temperature not lower than 58°C for a period longer than 45 days, the degradation reaches 90% (Iovino et al., 2008), (Javierre et al., 2015) (Gómez and Michel, 2013).

The present study, performed on a small scale, was aimed at providing an additional insight into the degradation of starch-based bioplastics during the composting process. Important parameters monitored during the process included the concentration, size and mass of bioplastics. The chemical composition and structure of the material were investigated by means of Fourier Transform Infrared (FTIR).

2. MATERIALS AND METHODOLOGIES

2.1 Experimental set-up

The experimental tests were performed in small-scale pyramidal composting heaps of 30 kg of waste, organized in two replicates referred to as Line 1 and Line 2, each comprising three heaps. One blank containing only the organic fraction of waste (OF) and two heaps with a mix of OF and the tested bioplastics manually cut into two different sizes were set up (Figure 1).

Waste composition for heaps was generated considering the need to simulate the waste matrix of food and green waste generally entering composting plants; it was selected on the basis of indications provided by ARPAV (Regional Environmental Protection Agency of Veneto) (Veneto Region, 2015): kitchen waste (53% on wet basis), wood chips (33%), grass (3%), with an inoculum of compost (11%) to facilitate start-up of the process. Heaps were placed in a climatic chamber under controlled temperature and humidity, which was monitored daily. Tests were carried out to evaluate granulometry, concentration, and physicochemical features of bioplastics: sampling was performed on 10th, 25th, 40th and 55th days with an aim to clarifying evolution during the composting process.

2.2 Tested material

Tested material consisted of starch-based bioplastic carrier bags; the composition of this material, commercially available in Italian supermarket, is 30% of starch and 70% of polybutylene adipate terephthalate (PBAT), a biodegradable not biobased polyester. The thickness of the material

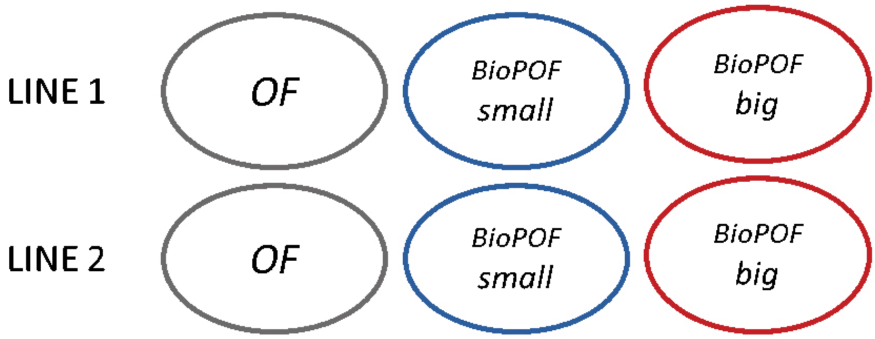

FIGURE 1: Experimental set-up of the composting test.

was 0.5 mm. The initial concentration of the tested material in the waste matrix was of 7 g bioplastics for each kilogram of mixture. Parameters used in identifying the latter were based on: (i) the current quantity of compostable bioplastics produced in Italy (Assobioplastiche, 2018); (ii) the increasing trend of 30% foreseen over the next five years (Bioplastics European, 2019); (iii) the amount of waste to composting (ISPRA, 2017); (iv) variables identified in the use of biobags by Italian families, including number of householders, seasonal variations in collection timing, and type of collection (Peelman et al., 2013).

As shredding tools adopted in composting plants determine a final size ranging from a maximum of 70-80 mm to a minimum of 20-10 mm, the initial size of bioplastic pieces represented a test variable and bags were shredded into two size ranges: from 50 to 70 mm, and from 20 to 30 mm, referred to as BioPOF big and BioPOF small, respectively.

2.3 Composting monitoring parameters

Typical parameters of composting process were monitored throughout the test. First, the heaps were weighted with a balance (gram precision) to evaluate mass trend during the process and to obtain the BioPOF mass for each heap. Total and volatile solids (TS and VS) were measured in accordance with the standard method IRSA-CNR Q 64/84 vol. 2 n. 2. A Benjamin by Vittadini - Suprema 600 oven and a Gefran 1001 muffle were used to analyse TS and VS, respectively.

Temperature was monitored three times a day using an Endress + Hauser - RSG40 probe at the centre and bottom of the heaps to obtain a daily average value for each heap.

Although in the presence of a negligible inorganic fraction, Total Carbon was measured in accordance with the UNI EN 13137 standard using a Shimadzu - TOC-V CSN Total Organic Carbon Analyzer combined with an SSM 5000 Solid Sample Module.

Total Nitrogen (TKN) was measured in accordance with IRSA-CNR Q 64/85 vol. 3 n. 6 mod. standard using the following equipment for the three phases of analysis: Velp Scientifica - DKL Heating Digester, Velp Scientifica - UDK 127 Distillation Unit and Crison – TitroMatic.

The IRSA-CNR Q 64/86 vol. 3 n. 8a standard was used to measure nitric nitrogen for transfer of the solid sample to the liquid phase, and IRSA-CNR 29/2003 vol. 2 n. 4040 A1 standard to measure analytical parameters of the liquid by means of UV analysis using Shimadzu - UV - 1601 UV-Visible Spectrophotometer. The monitoring parameter C/N was subsequently calculated.

Measurement of pH following transfer to the liquid phase was conducted on the liquid sample as established in IRSA-CNR 29/2003 vol. 1 n. 2060 using a Crison – TitroMatic.

To conclude, static and dynamic respirometric indexes RI_4, were measured by means of the SAPROMAT method (Federal Compost Quality Assurance Organization) using a Sapromat model E.

Table 1 reports parameters for the initial waste matrix, in comparison with values required by standard EN 14045 (EN 14045, 2003).

TABLE 1: Chemical characteristics of the initial compostable feedstock.

Parameters	Initial compostable matrix	Reference values (EN 14045)
Moisture (%)	54	> 40-50
VS (%)	91	> 50
C/N	34	20-35
pH	6	> 5
RI_4 (mgO_2/g TS)	50	-

2.4 Evaluation of bioplastics fragmentation and degradation in composting

2.4.1 Granulometry of bioplastics

To evaluate the fragmentation of bioplastics (physical size reduction), sieving analysis as established in EN 13432 was considered as a reference. However, since the focus of the study was to investigate the fragmentation of bioplastics both throughout and on completion of the process, sieving analysis was carried out on the 10th, 25th, 40th and 55th days of the test. For each analysis, 500 g of composted BioPOF big and BioPOF small wastes were sieved through six meshes, in accordance with ASTM international classification: 3/4, 3/8, 5/16, 3.5, 8, and 14, corresponding to 20 mm, 10 mm, 8 mm, 5.60 mm, 2.38 mm, 1.40 mm. The fractions of bioplastics passing through sieve 3.5 were considered micro-pieces.

After sieving, each granulometric fraction was visually screened to recover bioplastic fragments; this procedure for fragments recovery is suggested also in the main standards for bioplastics compostability (e.g. EN 13432). However, to improve the identification of the smallest particles, down to the size of 1 mm, the procedure was enhanced by a magnifying glass, diameter 50 mm and magnification 5x, and carried out for the same sample at least by three operators to reduce the subjectivity of the naked eye.

These fragments were weighted (g BioP) and used to calculate the concentration of bioplastics in the 500 g BioPOF sample.

The granulometric curve, describing the percentage of bioplastics retained in mass for each heap at any sampling time, was drawn.

Finally, bioplastics mass measured in each granulometric fraction was exploited to build a curve showing an average trend of granulometric size reduction during composting, applying Equation 1.

$$(Average\ BioP\ size)_j = \sum_i ((BioP\ \%\ mass)_i (BioP\ size)_i)_j \qquad (1)$$

Where j is the sampling day, from 0 to 55, and i are the granulometric fractions (20 mm, 10 mm, 8 mm, 5.60 mm, 2.38 mm, 1.40 mm); is identical with the mesh size of the sieve and is obtained with Equation 2.

$$(BioP\ \%\ mass)_i = 100 * \frac{(g\ BioP\ )_i}{\sum_i (g\ BioP)_i} \qquad (2)$$

2.4.2 Bioplastics mass

An important information to be obtained from the analysis is related to the decrease of total bioplastic amount (g) in the waste matrix during composting. To extrapolate the total mass of bioplastics (total BioP mass) in the heap at each sampling time, the bioplastic concentration previously obtained in the 500 g sampled from each heap was used. To obtain this unknown value, concentration was multiplied by total mass of the heap (BioPOF mass):

$$\left(\frac{g\ BioP}{g\ BioPOF}\right) * BioPOF\ mass\ (g) = total\ BioP\ mass\ (g) \quad (3)$$

However, two features in particular impacted on the experimental mass calculated as per Equation 1, namely increased moisture content in bioplastics, ranging from 5 to 9%, empirically evaluated by means of total solids analysis, and percentage of mass increase caused by waste adhering to the surface (dirt). The latter variable was defined on the basis of experimental data yielded by calculating the difference in mass before and after the cleaning of bioplastic pieces. The cleaning operation was carried out on samples of five pieces gathered from different size categories, for different days of analysis and from different heaps. These samples were weighted, cleaned using spatulas, hard brushes and tweezers to remove other adhering wastes and once again weighted. The percentage of dirt thus obtained was subtracted from total mass.

2.4.3 Degradation of bioplastics assessed by FTIR analysis

Fourier Transform Infrared (FTIR) spectra were obtained in total reflectance mode (ATR) with Thermo Scientific™ Nicolet™ iS™10 FTIR Spectrometer with 2 cm^{-1} spectral resolution, coupled with OMNIC software. Fragments of bioplastics were recovered during the composting process for the analysis; these had been previously cleaned and tooth-brushed in order to remove all waste residues from their surface. The investigated wavenumber range was 4000-600 cm^{-1}. The spectra were acquired in absorbance. The variation observed in peaks intensity and wavenumbers provides qualitative information on chemical change to the polymeric structure and the degradation process of materials.

FTIR analysis was also applied at the beginning of the research to different brands of bioplastic carrier bags present on the Italian market, to define each spectrum. The results confirmed that, despite the different brands, composition of the tested bags was identical, being all starch-based and made by the same company.

2.4.4 Visual analysis

In accordance with EN 14045 (EN 14045, 2003), visual assessment criteria distribution of particle size of remaining bioplastic particles and signs of microbial colonisation of the material. Ten bioplastic particles were selected to provide an overview of all visible degradation phenomena: consistency and thickness of the material, discolouring, erosion of the material (holes, tunnels, etc.) and ease of detection. Results of each assessment relevant for the test were documented in writing and by photographs.

Furthermore, evolution of the aspect of bioplastics may define the turning point after which pieces of the tested material became assimilable to compost for size, colour, smell and features.

3. RESULTS AND DISCUSSION

3.1 Evolution of composting parameters

As expected, two main phases were identified during the composting process: a thermophilic, or high rate phase, which ended within ten days, and a mesophilic curing phase, lasting forty-five days.

Process temperatures, having a similar trend in the heaps of the two lines, were in the proximity of 60°C over the first ten days (thermophilic phase). After this period, the heaps presented a progressive reduction of temperature to 30 °C, reaching ambient temperature in the final stage of the process.

A significant indicator of process evolution was mass variation of the heaps: mass trend and total solids content are reported in Figure 2. Mass decrease displayed the highest intensity during the thermophilic phase, reaching 60% of initial mass (40% loss) and 30% (70% loss) at the end of the process. Although mass loss may vary depending on the feedstock and evolution of the composting process, this result is comparable with trends reported in literature

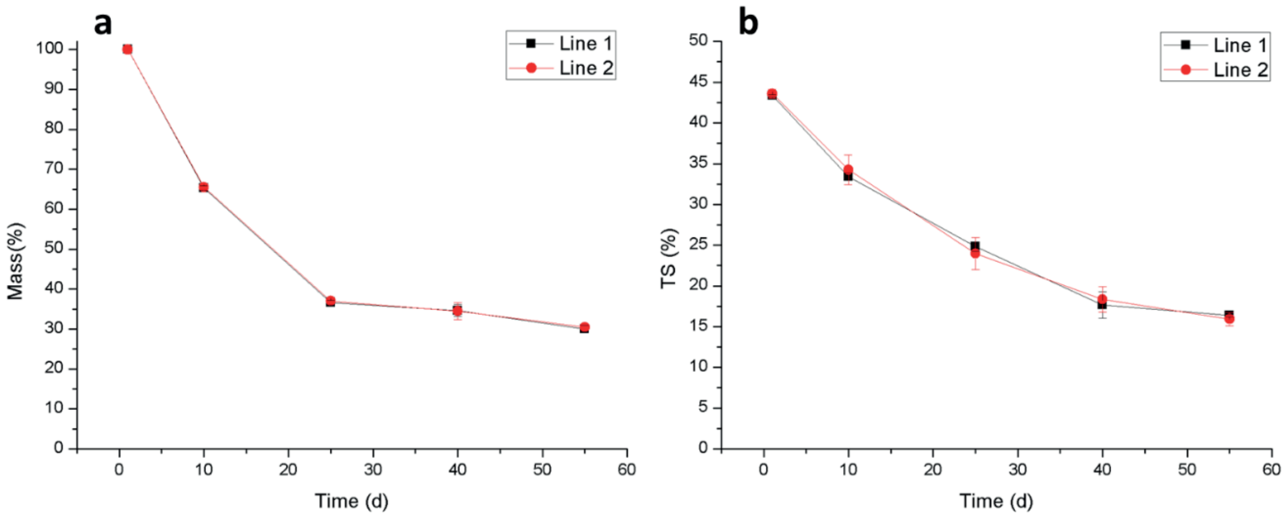

FIGURE 2: Decrease in mass (a) and total solids (b). recorded in the tests.

(Tiquia et al., 2002).

Final values obtained for composting parameters met the requirements of EN 14045 and Italian limits for compost quality (Annex 1 C of Law 748/84 modified by Ministerial Decree dated 27/03/98). Moisture was below 40%, pH was in the range of 7.6-7.7, and C/N in the range between 12 and 13 (Table 2). Respirometric index achieved values compatible with indication of a stable $RI_4 < 5$ mg O_2/ g TS (Barrena Gómez et al., 2006).

Moreover, results obtained for the heaps containing bioplastics (BioPOF) displayed no substantial differences compared to blank heaps (OF); therefore, in the context of composting, bioplastics do not seem to impact on final compost quality. Negligible alteration in the matrix due to bioplastics was observed by previous authors, both in compost (Balaguer et al., 2016) and in soil (Adhikari et al., 2016).

3.2 Granulometry of bioplastics

The fragmentation process was clearly visible in the tested material, mainly during the thermophilic phase of composting. Figure 3 shows the granulometric curves of small and large bioplastics, with an average value between the two lines of each size (BioP small and BioP big).

A substantial reduction of the initial size was manifested during the thermophilic phase: the granulometry of the particles of both BioP small and BioP big moved towards 10 mm. This trend of high fragmentation was expected in view of the typically higher intensity of the first phase of composting (Insam and de Bertoldi, 2007), (Tiquia et al., 2002).

At the end of the process, the granulometric curves of bioplastics revealed that the percentage of pieces passing through 5 mm sieve reached 55% and 20% in BioP small and BioP big respectively. Moreover, the curves outlined the generation of micro-pieces below the 2 mm regulatory threshold.

Figure 4 reports the average trend in size reduction of bioplastics based on data obtained from the granulometric curves. After a strong size reduction found in the end of the thermophilic phase, a linear trend was defined both for BioP small and BioP big during the mesophilic phase. The bullet points on the graphs are calculated in accordance with Equation 1. The graph illustrates the equations for the trend of each line.

The steep slope observed during the thermophilic phase confirmed the high efficiency of this phase in achieving fragmentation (Balaguer et al., 2016). The second, lower slope indicated a slowing down of the fragmentation process during the curing phase. The different size of bioplastics at the beginning of the experiment clearly affected the fragmentation process during the thermophilic phase; however, during the last part of the process the BioP small and BioP big curves definitely tended to converge.

The hypothetic time required to achieve an average final size of 5 and 2 mm was extrapolated using linear equations obtained for the curing phase, corresponding to 80 and 100 days of composting, respectively.

3.3 Bioplastics mass

During the composting process, bioplastic mass decreased to 20-30% of the initial mass, corresponding approximately to the mass achieved by the compostable heaps, as previously discussed. Final results were achieved on inclusion of variables relating to water and surface dirt. In particular, alteration (increase) of mass due to surface dirt was lower at days 10 and 25 than at days 40 and 55th, yielding percentages of 10-20% and 20-30%, respectively. Mass trend is reported in Table 3, illustrating average values obtained from the two lines for BioP small and big. It is evident how mass was still stable after the thermophilic phase, while during the curing phase it underwent a substantial decrease, showing a similar behaviour for both BioP. Mass decrease may be taken as an index of biodegradation, occurring thanks to the activity of microorganisms in the curing phase (Chiumenti et al., 2005) (Insam and de Bertoldi, 2007), (Tiquia et al., 2002).

3.4 Degradation of bioplastics investigated by means of FTIR analysis

Results obtained at FTIR-ATR analysis confirmed that the degradation of bioplastics occurred mainly during the

TABLE 2: C/N ratio and respirometric index RI4 parameters during composting.

	C/N (mgO_2/gTS)					RI_4				
Line 1										
	1st	10th	25th	40th	55th	1st	10th	25th	40th	55th
OF	29	21	13	17	13	52.3	38.7	38.2	26.4	1.3
BioPOF small	25	19	17	22	12	51.9	39.1	41.0	33.3	3.4
BioPOF big	24	19	16	18	12	49.8	36.5	43.7	30.2	5.3
Average	26	19	15	19	13	51.3	38.1	41.0	30.0	3.4
Line 2										
	1st	10th	25th	40th	55th	1st	10th	25th	40th	55th
OF	25	19	18	17	13	49.5	33.9	27.1	13.1	8.6
BioPOF small	23	17	14	14	11	50.0	34.8	35.4	22.1	6.6
BioPOF big	32	18	14	14	12	53.2	27.9	32.1	16.2	8.4
Average	26.8	18.0	15.6	14.8	12	50.9	32.2	31.5	17.1	7.9

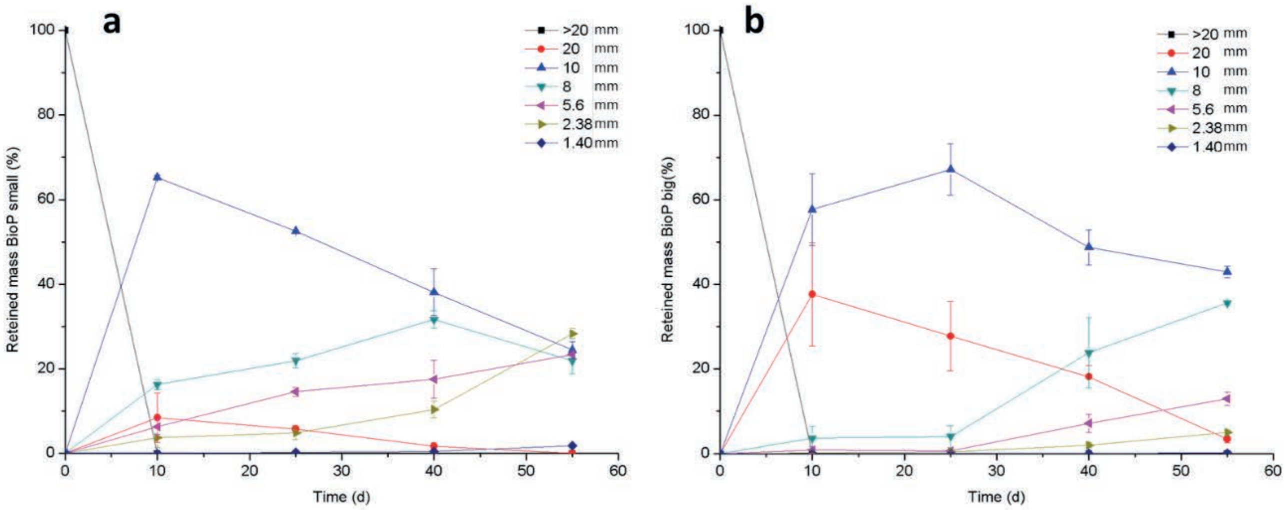

FIGURE 3: Bioplastics granulometric curves during composting process for bioplastics small (BioP small) (a) and big (BioP big) (b).

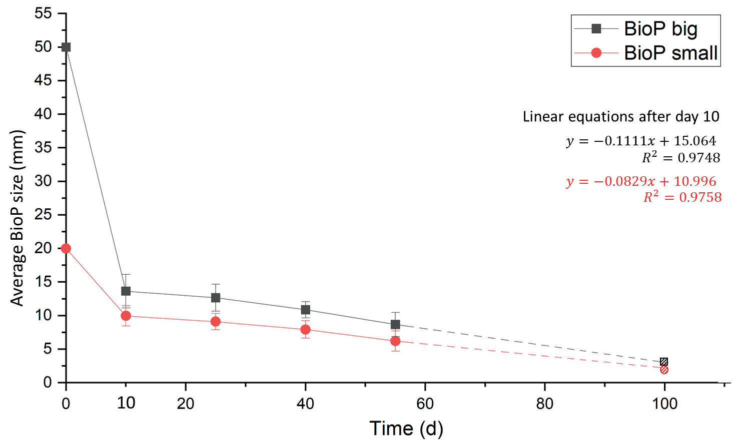

FIGURE 4: Evolution over time of size reduction (average) of bioplastics due to disintegration and biodegradation.

curing phase. In Figure 5, the spectrum of one sample (chosen from hundreds with similar results) for each day of analysis is reported.

After ten days of composting, the spectrum was almost identical to the initial spectrum, and at the end of the thermophilic phase few differences were noticeable; the most substantial changes, however occurred during the curing phase.

Prior to the 25th day, the spectra displayed a peak at 1717 cm^{-1} corresponding to the initial C=O group present in the structure of the tested bioplastic polymers: the peak is strictly relatable to PBAT present in the polymer (Elfehri Borchani et al., 2015). . Indeed, the disappearance of the peak in the spectra of day 40 and 55 can be considered as an index of at least a partial degradation of PBAT in the polymer. However, it is fair to highlight the trend of peak 727 cm^{-1}, which previous authors identified as representative of bonds typical of PBAT (Weng et al., 2013). Even after 55 days of composting, the peak is still present in the spectrum, assessing that this part of the polymer has not yet been degraded. Importantly, the two peaks in the surrounding of 1270 cm^{-1} are the main peaks relatable to starch in the polymer; their substantial decrease is a clear index of starch degradation in composting (Elfehri Borchani et al., 2015) (Ruggero et al., 2020).

Shortly after the 25th day, and particularly on days 40 and 55, an increase in concentration of the peaks in the regions between 1650 and 1600 cm^{-1}, and between 1450 and 1400 cm^{-1} was observed. These new peaks may be attributed to the formation of carboxylate ions (R-COO$^-$) by microorganisms and amidic groups of proteinaceous materials, respectively, in line with the findings of previous studies (Ruggero et al., 2020) (Arrieta et al., 2014). Moreover, an

TABLE 3: Bioplastic (BioP) mass decrease during composting.

	BioP mass (%)	
Time (d)	**BioP small**	**BioP big**
1st	100	100
10th	96.8	95.8
25th	60.0	79.0
40th	45.8	40.0
55th	29.8	18.3

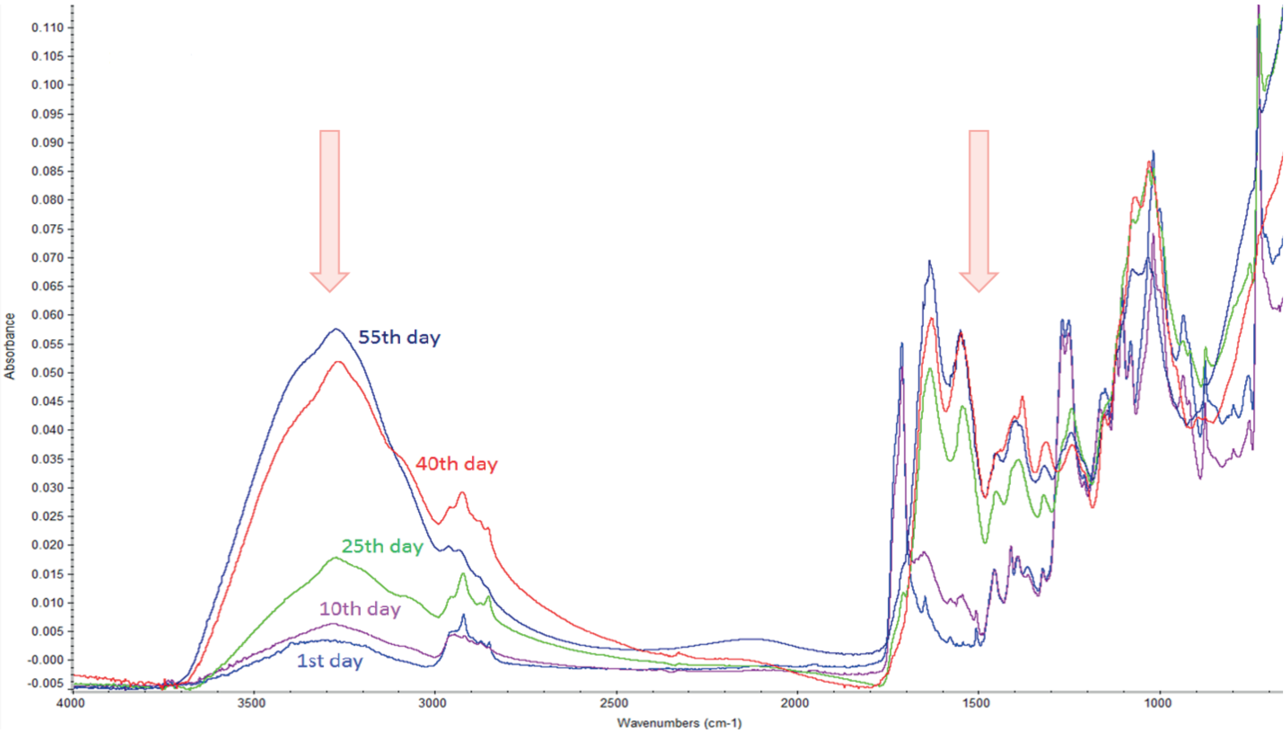

FIGURE 5: Spectra from the same heap at different times of the composting process.

increase of 3270 cm^{-1} peak concentration is imputable to the –OH group, a further sign of hydrolytic degradation of the material.

3.5 Visual analysis

Visual analysis of bioplastics during the process revealed increasing amounts of surface dirt on the pieces produced by contact with organic waste, in addition to limited signs of bacterial activity in terms of lateral erosion. With regard to the smallest bioplastic particles, a marked similarity with compost was detected mainly in the curing phase.

During manual operations carried out in the sieving analysis, it was visually noted that due to softening and increasing humidity, the bioplastics tended to wrap the food waste, thus preventing full degradation of the food waste.

Evolution of the visual aspect of bioplastics was photographically documented during the tests, as shown in Figure 6.

4. CONCLUSIONS

The present study highlighted a series of important observations: (i) based on the parameters monitored, the composting of heaps ended after a period of 55 days; however, at the end of this period, the degradation of bioplastics failed to meet the regulatory requirements for compostability (≤ 10% sized > 2 mm). (ii) changes in the physico-chemical features of bioplastics suggested that biodegradation had mainly occurred during the curing phase, as a result of the activity of microorganisms, while (iii) the granulometric trend of size reduction demonstrates that fragmentation is markedly enhanced during the thermophilic phase. Accordingly, the initial dimensions of the bioplastics were found to impact on the fragmentation process. Moreover, two linear trends were identifiable in the size reduction curve: a steep slope in the thermophilic phase, which evolved into a much slower trend of fragmentation during the curing phase. Extrapolation of experimental data has revealed the need for a 100-day period of composting in order to meet standard requirements for the compostability of bioplastics.

Taking into account the future extended use of bioplastics and marketing of new products, including cutlery, food packaging and coffee capsules, further research should be undertaken. Indeed, further research should extend the experimental day up to more than 100 days and be based on the inclusion of a series of additional features of bioplastic and variables with an aim to providing an in-depth analysis of the conditions (composting time, temperature, initial quantity and size) best suited to promoting degradation in composting.

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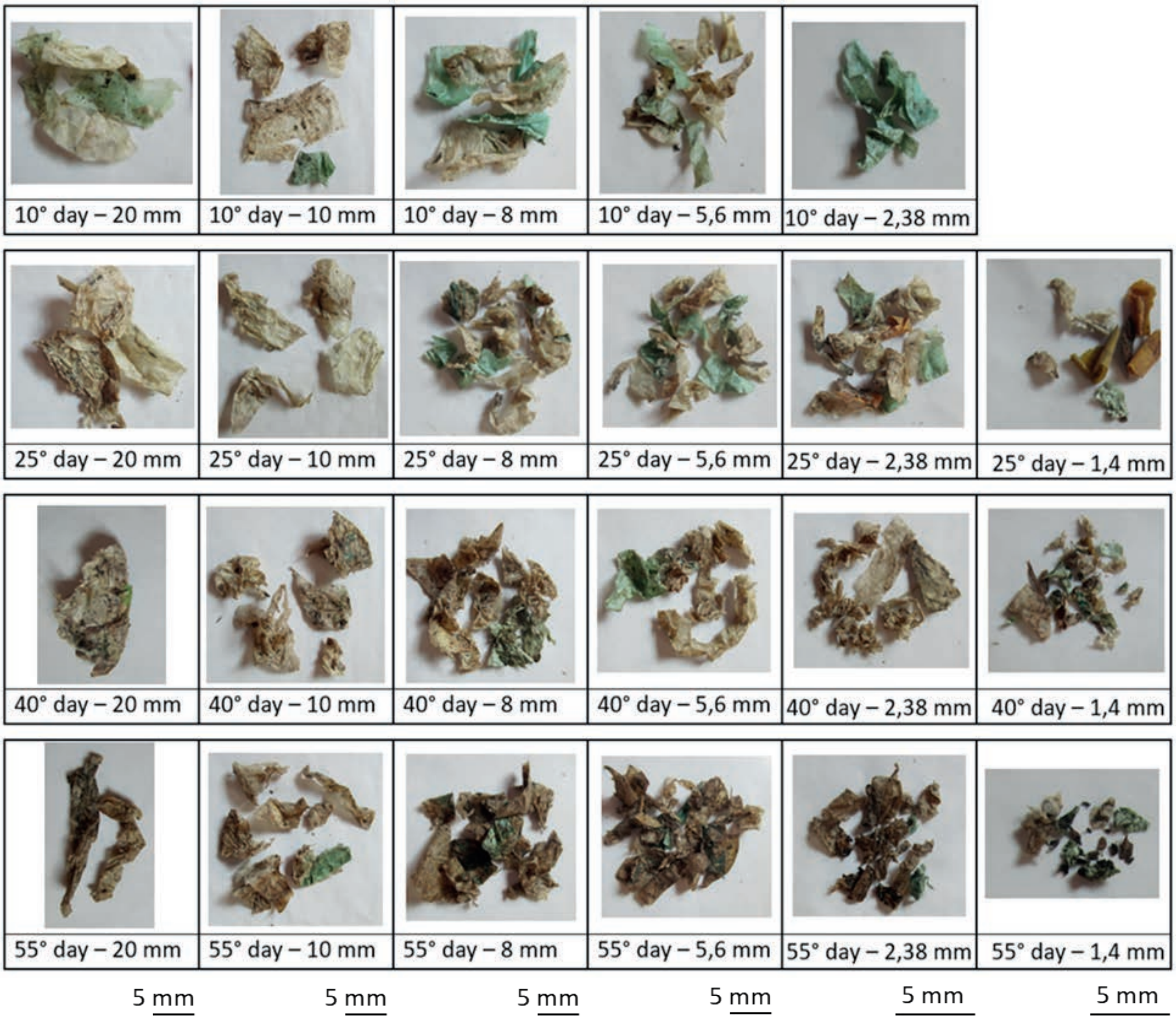

FIGURE 6: Photographs of bioplastic samples showing size reduction and variation over time.

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GASIFICATION OF BIOMASS IN A PLASMA GASIFIER

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ABSTRACT

This paper presents the thermodynamic analysis and experimental results on the plasma gasification of biomass using the example of wood waste. Thermodynamic computations revealed that synthesis gas can be produced from wood waste for utilization in the heat-and-power engineering, metallurgy and chemical industries. The air gasification of wood waste produces a synthesis gas yield of 71.6% (CO-41.9% and H_2-29.7%). Experiments on the plasma gasification of wood waste were conducted in an experimental setup composed of a plasma gasifier with 50 kg/h nominal productivity and a DC plasmatron with 70 kW nominal power. Based on gas analysis, the exit gas of the plasma setup exhibited the following composition, vol.%: CO-42.0, H_2-25.1, and N_2-32.9. The measured temperature in the bottom of the plasma gasifier was 1,560 K. The discrepancy between the experimental and calculated yield of synthesis gas was not more than 7%. Harmful impurities were not observed in the gases or the condensed products generated from the plasma gasification of wood waste.

1. INTRODUCTION

Humans have reached a point in their development at which an awareness of the limitations of natural resources and a need to preserve the environment have merged with the growing problem of recycling waste and maximizing the use of secondary raw materials and energy resources. Solid waste, such as biomass, from renewable energy sources has been shown to exhibit the most important increase in annual production volume. A substantial amount of waste is present worldwide, some of which is recycled, although large amounts of waste are simply dumped, causing problems for people and the environment. For example, the total amount of waste produced in the EU alone has amounted to 2.3 billion tons annually (Katsaros and Nguyen, 2018). Biomass is currently the world's fourth largest energy source, accounting for up to 14% of the world's primary energy demand and biomass power units range from small-scale to multi-megawatt sizes. Biomass is a versatile source of energy in that it can be readily stored and transformed into electricity and heat and also has the potential to be used as a raw material for chemical industry. The development of biomass use contributes to both energy and other non-energy policies (Veringa, 2005). In this paper, we consider wood waste (WW) as a species of biomass constituting the greater part of it.The world produces approximately 1 billion tons of WW annually. The main components of WW (80%) are wood and products from its processing (sawdust, bark, and wood chips). Power stations operating with WW have capacities of 35 million kW, producing 4% of the total primary energy consumption in developed countries; and 26% of the total primary energy consumption in developing countries (Prins Mark, 2005). During traditional WW incineration, the main combustion product is carbon dioxide, which increases the greenhouse effect. The use of WW in the energy sector is the final stage of logging production,and aimed at improving the effectiveness of environmental measures. The most common WW processing technology is gasification (Veringa, 2005; Lan et al., 2018; Mourão et al., 2015). During WW gasification, the main product is synthesis gas, not carbon dioxide, which is primarily generated when WWs are burned. From 1 kg of WW, it is possible to obtain approximately 2.5 m^3 of power gas, and the main combustible components of the gas are carbon monoxide (CO) and hydrogen (H_2). According to the heat input method, WW gasification processes are divided into autothermal and allothermal processes. During the autothermal process, thermal energy that is needed to achieve the required temperature comes from the combustion of a portion of the WW; during the allothermal process,

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heat is supplied from outside; for example, heat can be supplied from a plasma generator. In the process of autothermal gasification, tar is formed and accumulated in the gas purification filters and is difficult to remove, which causes energy losses (Porteous, 2005; Materazzi, 2013). The heat supply method determines the composition of the synthesis gas and the energy consumption for its production. During the air gasification of WW, the specific yield of synthesis gas obtained in the autothermal process is 15.4-52.4% higher (with a decrease in its quality) than the yield of synthesis gas obtained in the allothermal process. In plasma gasification, the proportion of noncombustible components in the producer gas decreases to 23.2-61.0%, and the energy consumption for heating the gasification products to the necessary temperature decreases by 17.4-46.5% (Brattsev et al., 2011). Thus, the advantages of employing plasma methods for WW processing are the complete decomposition of wastes; the decrease in the volume of the exhaust gas produced; the smaller carryover of dispersed particles; the high performance achieved with small-scale equipment; the creation of a desired gaseous atmosphere; the ability to adjust the operation of the process by changing the flow rate of air and power of plasmatrons; and the increase in the production and quality of synthesis gas without changing the consumption of the oxidizing agent. The main disadvantage of WW plasma gasification is the required energy consumption for the generation of plasma. Recently, the plasma gasification of wastes has become widespread (An'shakov et al., 2007; Byun Youngchul et al., 2012; Heberlein and Murphy, 2008; Matveev et al., 2016; Surov et al., 2017; Zhang et al., 2012; Zhovtyansky et al., 2013). Plasma gasification achieves the maximum yield of the synthesis gas ($CO+H_2$) by reducing the concentration of the ballasting gases (CO_2 and N_2). However, in post-Soviet regions, the use of wastes, including WW, essentially does not occur; thus, the problem of its utilization is very relevant.

In this paper, we discuss the results of the thermodynamic analysis of fuel gas, which is free of harmful impurities, produced by the gasification of WW in air plasma. Additionally, the experimental installation system is presented, and the experimental results of the gasification of WW in air plasma are compared with the computation results. The foundation of plasma gasification of WW was established during the creation of the plasma technology for the gasification of solid fuel (Matveev et al., 2008; Messerle and Ustimenko, 2007).

2. MATERIALS AND METHODS

2.1 Materials

In this article, WW was composed of a mixture of sawdust and wood chips. WW constitutes a significant share of the waste generated by the woodworking industry. According to (CHEM, 2016; Graedel, 2003) WW is composed of the following chemical components, wt.%: C - 49.88, O - 43.81, H - 5.98, N - 0.10, K_2O - 0.01, CaO - 0.12, MgO - 0.02, MnO - 0.01, Fe_2O_3 - 0.01, Al_2O_3 - 0.01, SiO_2 - 0.01, SO_3 - 0.01, P_2O_5 - 0.02, and Na_2O - 0.01. The organic part of WW is carbon, oxygen, hydrogen and nitrogen with a total concentration of 99.77%, whereas the mineral part of WW only accounts for 0.23%. The higher heating value (HHV) of the WW was calculated by an equation based on the ultimate analysis (Demirbaş and Demirbaş, 2004): $Q=1{,}000\cdot(33.5\cdot C + 142.3\cdot H - 15.4\cdot O - 24.5\cdot N)$ [kJ/kg]. The HHV was 18,450 kJ/kg.

2.2 Computations

The Terra (Gorokhovski et al., 2005) software package was used to perform thermodynamic calculations of WW plasma air gasification. This software was intended for numerical calculations of high-temperature processes and possesses its own extensive database of thermodynamic properties of 3,000 individual substances over a temperature range of 300 to 6,000 K. The database includes the thermodynamic properties of the organic and mineral components of WW and contains the thermochemical properties of radicals, ionized components and electronic gas, which are accounted for in thermodynamic calculations. In contrast to traditional thermochemical methods of equilibrium computations, which use the equations of chemical reactions, Gibbs energy, equilibrium constants, and the Guldberg and Waage law of mass action, the Terra software is based on the principle of maximum entropy for isolated thermodynamic systems in equilibrium and does not use the equations of chemical reactions, operating with many chemicals within the database.

2.3 Experimental installation system

Experimental studies of WW gasification were performed in the experimental installation system (Figure 1), which consists of a plasma chemical reactor (Figure 2) with WW productivity up to 50 kg/h and a long life DC plasmatron (plasma generator) with 70 kW nominal power (Golish et al., 2009). Figure 2 shows an image of the reactor with a lifted lid and plasma flame. To increase the service life of the plasmatron, a method was developed for the plasma pyrolysis of hydrocarbon gases with the subsequent deposition of the condensed products generated from pyrolysis on water-cooled copper electrodes of the plasmatron. According to the method, a propane-butane mixture is fed into the zone of arc discharge between the cathode and anode (in thermodynamic analysis, due to the very low flow rate, the propane-butane mixture was not taken into account). As a result, carbon vapour is formed in the cavity of the cathode and on the inner surface of the anode. Electron microscopy and Raman spectroscopy studies of the electrode coating showed that it consisted of a composite nanostructured carbon material including largely single-walled and multi-walled carbon nanotubes in addition to other carbon forms with a certain amount of copper atoms intercalated in the carbon matrix. An experimental study of the long life plasmatron showed that at a plasmatron power of 72.6 kW (I=220 A, U=330 V), a plasma forming air flow rate of 250 l/min and a propane-butane flow rate of 1.8 l/min, the temperature at the plasmatron nozzle exit section was 5,500 K (Figure 2). Based on the resource tests of the plasmatron for 1,000 hours, the erosion of the copper electrodes was not fixed, as the true electrode functions were performed by the regenerable nanocarbon coatings of the copper electrodes (Il'in, et al., 2010).

The composition of the experimental setup (Figure 1), except reactor 4 and plasmatron 1, includes the plasmatron power supply, gas and water supplies into the reactor and plasmatron and exhaust gas cleaning 7. The experimental installation system is equipped with a sampling system for the analysis of the gas and condensed products generated from WW gasification (Messerle et al., 2018). A plasma reactor was designed for plasma gasification of WW. The reactor is a cube lined by refractory bricks with thicknesses of 0.065 m. The size of the inner side of the cube is 0.45 m, which gives the reaction volume of the reactor of 0.091 m^3. The pipe for supplying WW briquettes 2 can be used to measure the temperature inside the reactor using an infrared pyrometer. A Pyrometer Ircon Ultrimax Plus UX10P is used to measure temperatures from 600 to 3,000°C (873-3,273 K). The measurement error depends on the temperature range, and it is ±0.5% of the measured value for the temperature range up to 1,500°C (1,773 K), ±1% for the temperature range from 1,500-2,000°C (1,773-2,273 K), and it increases to ±2% for the temperature range over 2,000°C (2,273 K). The temperature resolution is not lower than 1°C. The device is equipped with a Serial port to enable connection to the computer, and the temperature can be controlled by the RS-232C protocol during the experiment, which generates results in an on-line regime. The sampling interval of the device is 0.5 seconds.

The process of WW plasma gasification is as follows. Plasmatron 1 is started, and the inner liner surface of the bottom of reactor 4 is heated to a temperature of 1,215 K (approximately 15 minutes); then, the WW briquettes are introduced into gasification zone 3 through pipe 2 (Figure 1). The weight of each briquette is 0.33 kg. It takes two minutes to supply 5 briquettes. The WW is gasified in the air plasma flame, providing an average mass temperature in the reactor volume of up to 1,600 K. The gaseous products are removed from the reactor and transferred into cyclone combustion chamber 6, and the condensed products accumulate in the bottom of the reactor. The combination of the heat release zone from the plasma flame with WW gasifier 3 and slagging contributes to the intensification of WW processing. The cooled gaseous products enter gas purification unit 7, after which with the help of sampling system gas is supplied to the analyser. Ventilation system 9, including the pressure control valve 8, provides a low pressure in the reactor up to 10 mm of the water column.

3. RESULTS AND DISCUSSION

3.1 Thermodynamic calculations

The Terra (Gorokhovski et al., 2005) software package was used to perform the thermodynamic calculations of WW plasma air gasification. The calculations were carried out in the temperature range of 298-3,000 K and at a pressure of 0.1 MPa. The aim of these calculations was to determine the integral parameters of the gasification process, such as the equilibrium composition of the gas phase of the gasification products, the degree of carbon gasification and the specific power consumption of the process. The initial technological mixture with a mass ratio of WW to air of 1 was used for air gasification. This mass ratio is based on a series of calculations of plasma-air gasification of WW with varying excess air coefficients. The mass ratio selection criteria were the achievement of 100% carbon gasification, the maximum concentration of combustible components ($CO + H_2 + CH_4$) in the waste gasification products and the suppression of nitrogen oxide formation. We should also note that the stoichiometric ratio is the exact ratio between air and WW at which com-

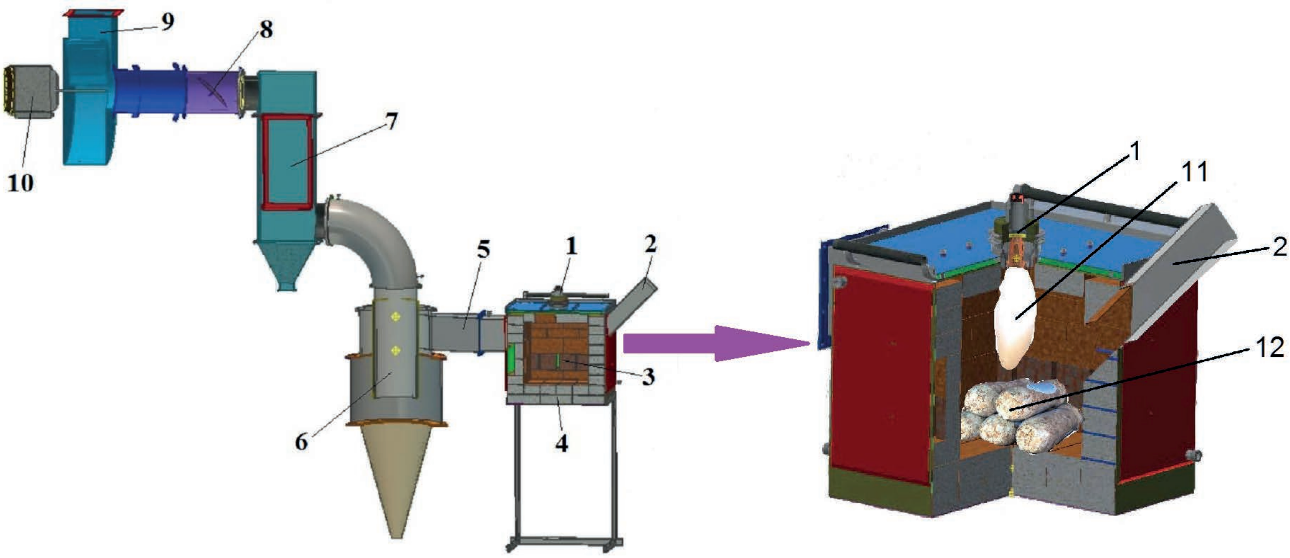

FIGURE 1: Layout of the experimental unit for plasma gasification of WW and scheme of the plasma reactor: 1 – arc plasmatron; 2 – pipe for supplying the WW briquettes; 3 – WW gasification zone; 4 – reactor; 5 – chamber for combustible gas removal from the reactor; 6 – cyclone combustion chamber; 7 – gas purification unit with a bag filter; 8 – control valve; 9 – ventilator of the exhaust system; 10 – engine of the exhaust system; 11 – plasma flame; 12 – WW briquettes.

plete combustion occurs, and the products only contain carbon dioxide and steam. The stoichiometric ratio for WW combustion is 6.

Figure 3 shows the variation in the concentration of gaseous components depending on the temperature of the WW air gasification. With increasing temperature, the synthesis gas yield increases to a maximum at T = 1,600 K. This result occurs due to an increase in CO and H_2 concentrations and a corresponding decrease in CH_4 and CO_2 concentrations because of their thermal dissociation. The maximum concentration of combustible components in the synthesis gas reached 71.6% (CO - 41.9% and H_2 - 29.7%). It should be noted that at 1,200 K, the total synthesis gas concentration is 71.3%, which differs little in value from its maximum value. The concentration of oxidants (CO_2+H_2O) at these temperatures does not exceed 0.7%. With increasing temperature, the concentration of synthesis gas is slightly reduced due to the appearance of atomic hydrogen (H) in the gas phase, which reaches a concentration of 7.8% (T = 3,000 K). The concentration of ballasting nitrogen (N_2) remains almost constant in the temperature range of 1,200-3,000 K and is 27.8-26.7%. The concentration of methane (CH_4) decreases sharply, and its concentration converges to zero at a temperature of 1,200 K.

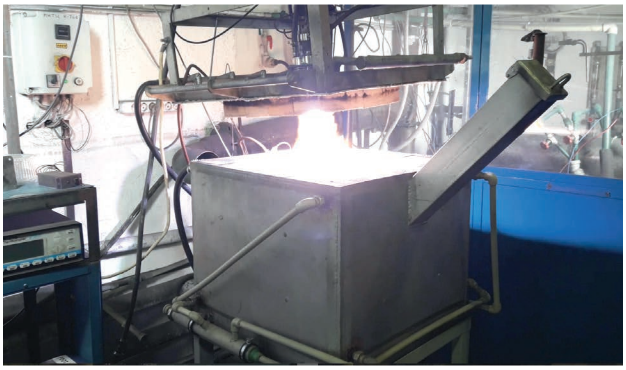

FIGURE 2: Photo of the plasmatron in operation with the reactor lid lifted.

The degree of carbon gasification X_C (Figure 4) is determined from the carbon content of the solid residue. Specifically, X_C is calculated according to the following expression: , where C_{ini} is the initial amount of carbon in the WW, and C_{fin} is the final amount of carbon in the solid residue. As shown in Figure 4, the degree of carbon gasification amounts to 100% at a temperature of 1,200 K. Carbon is completely transformed into the gaseous phase, forming CO at a temperature higher than 1,200 K. This process provides a hundred percent carbon gasification.

The specific power consumption Q_{SP} (Figure 5) was defined as a difference between the total enthalpy of the final I_{FIN} (current temperature of the process) and initial I_{INI} (T=298 K) state of the working substance (mixture of WW and gasifying agent) reduced to 1 kg of WW. Q_{SP} is calculated according to the following expression: , where m_{WS} is the mass of the working substance, and m_{WW} is the mass of WW. The specific power consumption for the process of WW gasification increases with temperature throughout its range. For the temperature T = 1,600 K, at which the yield of synthesis gas reaches its maximum (Figure 3), the specific power consumption for air gasification of the WW constitutes 2.49 kW h/kg. Such moderate energy consumption for air gasification of WW is associated with compensation in the endothermic effect due to the heat of the oxidation reaction of carbon in air.

The carbon gasification efficiency characterizes the WW gasification process efficiency and is an important indicator of the energy efficiency of the gasification process (Matveev et al. 2008). The criterion of the energy efficiency of the solid fuel gasification process is defined as the relative thermal power of the produced combustible gas. The physical heat of the produced combustible gas was not considered when the relative thermal power was determined. The efficiency of the WW gasification process can be calculated by the following formula: , where m_G is the mass of combustible gas produced, Q_G is the heat of combustion of the combustible gas, [kJ/kg], and Q_{WW} is the heat of combustion of WW [kJ/kg]. The WW gasification efficiency (Figure 6) increases sharply with

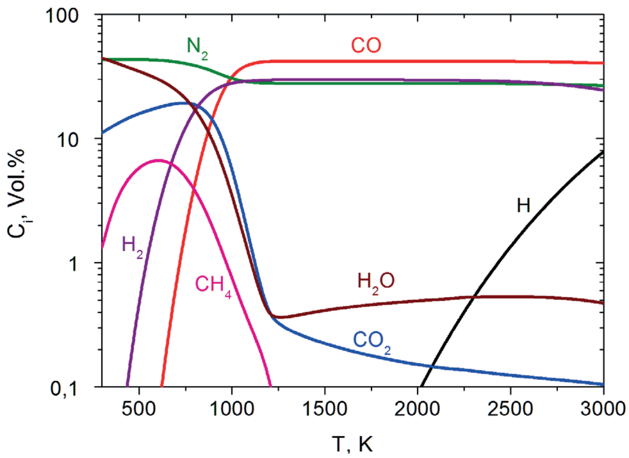

FIGURE 3: The variation in the concentration of gaseous components depending on the WW air gasification temperature.

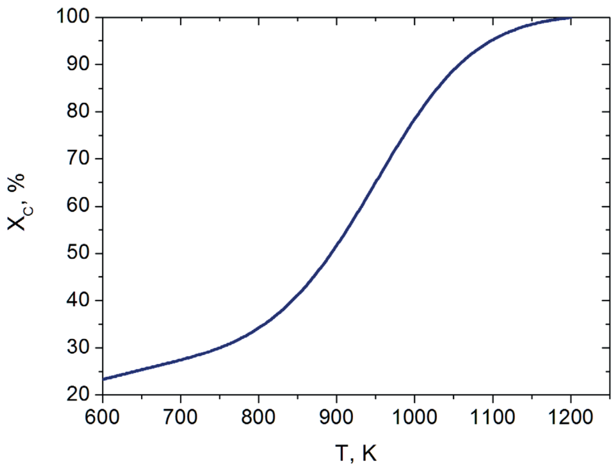

FIGURE 4: The degree of carbon gasification depending on the WW gasification temperature.

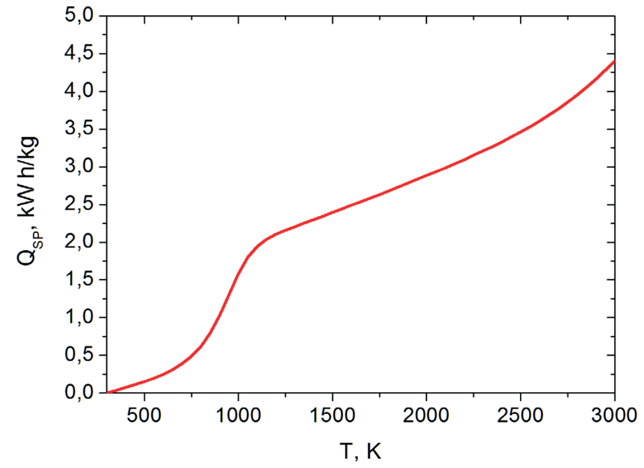

FIGURE 5: Specific power consumption for the WW plasma gasification process depending on temperature.

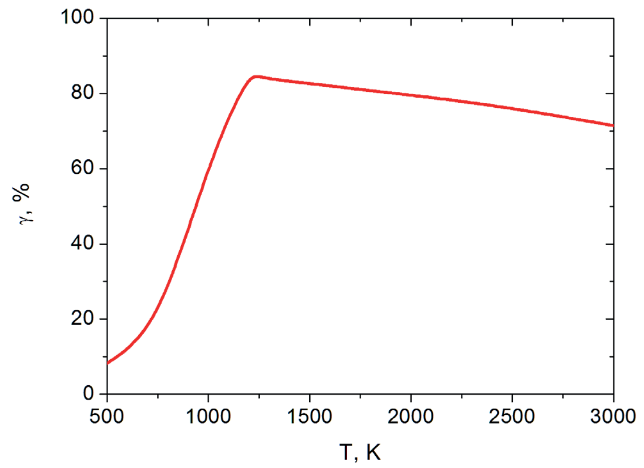

FIGURE 6: The dependence of the WW gasification efficiency on temperature.

temperature to a maximum of 85.6% at a temperature of 1,200 K. With a further increase in temperature, the WW gasification efficiency gradually decreases, which is explained by an increase in the specific power consumption of WW gasification at the previously achieved 100% of carbon gasification. For the temperature T = 1,600 K, the WW gasification efficiency for the air gasification of WW is rather high at 82%.

The parameters that were determined and the regularities of the WW plasma gasification process that were identified were used to develop an experimental installation system.

3.2 Experiment

As a result of the WW plasma gasification, synthesis gas was produced. Figure 7 shows the flame of the gas exiting pipe 2 at a short time disabling of the exhaust system. The resulting fuel gas is intensively ignited in air. The measured temperature of this flame was approximately 1,600 K. During the experiment, the fuel gas was withdrawn using the exhaust system. The measured temperature in the bottom of the reactor was 1,560 K. Under the influence of the air plasma jet, the weight average temperature in the reactor reached 1,600 K, an organic portion of the WW was gasified, and an inorganic portion (ash) of the WW was accumulated in the slag formation zone of the reactor and in bag filter 7 (Figure 1). The obtained synthesis gas was incinerated in cyclone combustion chamber 6. The combustion products were continuously removed from the installation through the cooling and purification systems. The ash was removed from the reactor after the plasmatron was shut down and the reactor was cooled. The plasmatron was turned off 25 minutes after the download of the first WW briquette. Thirty briquettes with a total mass of 9.9 kg were gasified during this period. The consumption of the WW briquettes was 23.8 kg/h. The air flow rate through the plasmatron was 23.6 kg/h. The parameters of the synthesis gas (composition and temperature of gasification products) were maintained during the periodic loading of the waste into the reactor by the uniform supply of briquettes every 50 seconds by sampling the gas after establishing a stationary regime of gasification for 20 seconds. We should also note that since the measured temperatures were comparatively low in the experiment and varied within 1,560-1,600 K, the role of radicals, ionized components, and electronic gas could not be significant. This phenomenon is confirmed by the calculated concentrations of these components at these temperatures (for example, at T=1,600 K, vol.%: electronic gas - $0.5\cdot10^{-15}$, H^- - $0.2\cdot10^{-19}$, OH - $0.1\cdot10^{-5}$, and CH - $0.4\cdot10^{-13}$).

The experimental study results for the plasma reactor operating conditions during WW plasma gasification were determined; in addition, an exhaust gas analysis was performed, the samples of the condensed products were collected from the slag formation zone of the reactor, and the residual carbon content in the slag was measured. Gas analyses were performed on a gas chromatograph SRI 8610C, which showed the following gas composition at the exit of the gas purification unit, vol.%: CO - 42.0, H_2 - 25.1, and N_2 - 32.9. The specific heat of the combustion of the synthesis gas produced by air gasification amounted to 9,400 kJ/kg. The total concentration of the synthesis gas was 67.1%, which agreed well with the thermodynamic calculations. The calculated yield of the synthesis gas at 1,600 K was 71.6% (CO - 41.9% and H_2 - 29.7%). Thus, the discrepancy between the experimental and calculated target product yields (synthesis gas) did not exceed 6%. The thermodynamically predicted concentration of N_2 was 27.8%. This difference could be because of the unguided dissolution of the experimentally produced synthesis gas by ambient air.

After gasification of 9.9 kg of WW 0.013 kg of ash were collected from the bottom of the reactor. This quantity of ash is approximately 0.12% from the initial quantity of WW. Residual fly ash (0.1% ash) was transferred away with the exhaust gas. The volumetric orifice flow rate of the exhaust gas was 48.3 kg/h. The discrepancy between the experimental and calculated flow rates is 2%.

In the experiments and in the calculations, neither tar nor harmful impurities were found in the products of the WW plasma gasification. The carbon content of the slag in

FIGURE 7: Photo of the combustible gas control flame from the pipe for supplying the WW briquettes.

the sample was 1.13 wt.%, which corresponded to 96.6% WW carbon gasification. The carbon content was determined using the absorption-gravimetric method. The discrepancy between the experimental and calculated values of carbon gasification did not exceed 3.5%.

According to the results of experiments, the specific power consumption of the WW gasification in the plasma reactor reached 3.05 kWh/kg of the working substance. The calculated specific power consumption of the air-plasma gasification of the WW was 2.49 kWh/kg (Figure 5). The discrepancy between the calculated and experimental values of the specific power consumption for the process was 18%. This discrepancy is because in thermodynamic calculations, the lowest possible energy consumption in an isolated thermodynamic system is determined without taking into account the exchange of heat and work with the environment. In practice, the plasma reactor itself and the plasmatron are characterized by considerable heat losses to the environment with cooling water.

4. CONCLUSIONS

Thermodynamic calculations showed that the maximum synthesis gas yield from the plasma gasification of wood waste in an air medium is achieved at a temperature of 1,600 K. A total concentration of 67.1% was obtained for the WW synthesis gas with the air plasma gasification of WW. The specific heat of combustion of the synthesis gas produced by air gasification reached 9,400 kJ/kg. In the experiments and in the thermodynamic calculations, there were no harmful impurities found in either the gas or the condensed products of WW plasma gasification. A comparison of the experimental and calculated results of the plasma gasification of WW showed good agreement.

In contrast to traditional gasification methods, 100% WW gasification is achieved in the plasma-air gasification of WW, the WW gasification efficiency reaches a significant value of 85.6% at a relatively low process temperature (1,200 K), and gasification products do not contain harmful substances.

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UPGRADING BIOGAS TO BIOMETHANE BY USE OF NANO-STRUCTURED CERAMIC MEMBRANES

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ABSTRACT

In order to meet the demands of growing economies while considering environmental implications, the use of clean and renewable sources of energy has increasingly become of interest. Biogas utilisation is a means by which these rising needs can be met. This involves the use of waste materials; which are deposited on a daily basis by agriculture, sewage, household, to produce energy that may be used for heating, electricity, transportation and other daily needs. This paper would look into the use of nano-structured ceramic membranes for the upgrading of biogas to a high value fuel that can be used for a variety of purposes. The use of membranes offers great advantages including low running costs, high efficiency and the elimination of the need for phase change of the gas. Experiments were carried out using membranes of different pore sizes (15nm, 200nm and 6000nm) to ascertain which would be the most suitable for use in terms of permeability and yield of product gas. The 15nm membrane showed the greatest exit flow of methane compared to carbon dioxide and a mechanism approaching an ideal knudsen regime. Taking into account the effect of molecular weight and viscosity, these results show that the smallest membrane pore size of 15nm had a greater impact on the flow mechanism and thus improvement can be made by modification of the membrane to achieve a mechanism of surface diffusion of the particles.

1. INTRODUCTION

Biogas is evolved from the anaerobic digestion of biogradable materials such as human or animal waste, food scraps, cotton, wool, wood and other organic sources. The biogas obtained can be used directly as a fuel but in order to fully harness its potential, it can be cleaned and upgraded to a point where its heating value is very high, and impurities are removed such that it can be injected into the national gas grid.

The treatment or upgrading of biogas is essential because: (i) the presence of CO_2 in the gas reduces the power output from the engine, takes up space when biogas is compressed for storage and causes freezing problems when the compressed gas undergoes expansion at valves and metering points (ii) traces of H_2S can produce H_2SO_4 which corrode pipes, fittings, etc. (iii) moisture reduces the heating value of the biogas and causes corrosion. Nonetheless, several safety aspects need to be considered during the treatment and utilization of biogas. It is very crucial to be aware of the associated risks and to minimize them. The most common risks include flammability, poisoning (due to the presence of H_2S), suffocation and the risks associated with high pressures and temperatures. However, the advantages include the fact that biogas is lighter than air and any gas leakage would rise upward. Moreover, upgraded biogas has a greater temperature of ignition than both petrol and diesel so the possibility of a fire or explosion is reduced (Svenskt Gastekniskt Center, 2012).

The advantages of utilizing biogas are numerous; biogas upgrading technology can turn the cost of waste management into a revenue opportunity. Turning waste into a renewable source of energy by this upgrading process will reduce dependence on importation of fossil fuels, reduce greenhouse gas emissions, improve environmental quality, increase local jobs and provide revenue by export thereby boosting the economies. The benefits cannot be overstretched as even the digestate from anaerobic digestion offers an opportunity to recycle nutrients in the food supply, reducing the need for both petrochemical and mined fertilizers. Research has proven that upgraded biogas shows lower carbon intensities compared to other vehicle fuel (Penev et al., 2016; Scarlat, Dallemand, & Fahl, 2018).

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Currently, American liquified natural gas (i.e. methane) is being exported to counties like China, India and Japan who still rely on coal for power generation but wish to substitute their coal for clean gas (Weinstein, 2019). Thus, this technology can be implemented on a large scale for the export of upgraded biogas to other countries without knowledge, infrastructure and expertise, this would provide revenue for the economy as well as an abundance of jobs. This highlights one of the advantages of biogas compared to other forms of renewable energy such as solar and wind that cannot be stored and transported.

This paper would introduce a novel method for the upgrading of biogas to a clean and useful fuel replacing fossil fuels. The use of nano-structured membrane technology would be implemented, where biogas components pass through the membrane as the feed gas to observe the separation characteristics at various operating conditions for three different membrane pore sizes.

The main goal is to analyse the flow characteristics of biogas through a membrane in order to measure the permeability and selectivity of the gas components through various pore sizes and determine the flow/transport mechanisms of biogas components.

2. LITERATURE REVIEW

2.1 Current Technologies

The current state-of-the-art technologies for biogas upgrading include absorption, adsorption, membrane separation and cryogenic separation. Absorption may be by physical or chemical scrubbing. In the case of water scrubbing, biogas is injected into the bottom of the absorption column where carbon dioxide is absorbed by water that runs from the top at high pressure. The absorption column is filled with random packing to increase the gas-liquid-contact. The counter current flow of gas and liquid is necessary to achieve a high process efficiency. During the process, carbon dioxide and some methane are physically absorbed in water and the selectivity of the process depends on the much higher solubility of carbon dioxide compared to methane in water. Finally, the carbon dioxide is then released from the water again in the desorption column, by using air at atmospheric pressure as stripper medium. The process of chemical scrubbing is similar to the water-scrubbing process, however the solubility of carbon dioxide in an organic solvent is increased by about five times compared to water. This means that the volume of solvent that is requires in the system is significantly reduced.

The pressure swing adsorption method may also be used to remove the unwanted constituents of biogas by changes in pressure within a system that cause each gas to move or be adsorbed at different rates. The adsorbents may be of the equilibrium type, which adsorbs more of carbon dioxide than methane. It may also be of the kinetic type, adsorbing carbon dioxide at a faster rate than methane due to con- trolled diffusion rates. Some examples of adsorbent materials which are commonly used include activated carbons, natural and synthetic zeolites, titanosilicates, silica gels and carbon molecular sieves.

Membrane technology involves the use of a barrier that restricts the flow of some components of biogas which allows the resultant gas to be stripped of the restricted gas. Cryogenic technology is still relatively new and works at very high pressures and low temperature to permit the separation of gases. Research has shown that membrane technology and absorption are the most economical and efficient methods that possess high potential for scaling up to achieve an efficient and profitable means of upgrading because of their high gas recovery and minimal loss of gas. Currently, absorption by chemical scrubbing is widely used to upgrade biogas, but this results in a significant amount of waste products that need to be properly disposed of thereby increasing costs. In this research, the use of membrane technology is explored as an effective and efficient means of upgrading biogas. The energy consumption is relatively lower than the conventional upgrading processes as they do not consume energy in the latent heat of evaporation and the possibility of methane slip or losses is minimal. Other advantages are that they are compatible with temperature sensitive materials and are not chemically altered, separation does not involve phase change, other reactions can be processed simultaneously due to the versatility of membranes, there is a high efficiency of separation, they are simple to operate and membranes have high selectivity and permeation rate (Scott & Hughes, 2012).

2.2 Membrane Gas Transport Mechanisms

The mechanism of gas transport in membranes are derived from Graham's law and Fick's law (Shehu, Okon, Orakwe, & Gobina, 2018). Graham's law states that the rate of diffusion of a gas is inversely proportional to the square root of its molecular weight. In mathematical form (Domenico De Meis, 2017):

$$\frac{Rate\ a}{Rate\ b} = \left({}^{M_b}/_{M_a}\right)^{1/2} \qquad (1)$$

Where,

Rate a and Rate b denote the rate of diffusion of the first gas and second gas respectively,

M_a and M_b are the molar masses of gases a and b in g mol^{-1} respectively.

Fick's law relates the molar flux to the concentration gradient through the membrane thickness. This can be written in mathematical form as (Domenico De Meis, 2017):

$$F_i = \frac{P_e}{L}(P_1 - P_2)A \qquad \left(\frac{mol}{s}\right) \qquad (2)$$

Where,

P_e is the permeability, mol m^{-1} s^{-1} Pa^{-1}

L is the length, m

P_1, P_2 is the pressure at point 1 and 2 respectively, Pa

A is the permeation area, m^2

Gas transport in membranes can take place through several mechanisms including Hagen-Poiseuille flow, Knudsen diffusion, surface diffusion, capillary condensation and molecular sieving.

2.2.1 Hagen-Poiseuille

This mechanism comes to play when the pore diameter is large compared to the mean free path of the gas molecules (λ). Here, the gas permeance is inversely proportional to the gas viscosity (Domenico De Meis, 2017; Oyama, 2011).

$$P_e = \frac{\varepsilon\eta r^2}{8\mu RT} P_{av} \quad (\text{mol m}^{-1}\ \text{s}^{-1}\ \text{Pa}^{-1}) \tag{3}$$

Where,
Ɛ is the porosity, dimensionless
μ is the viscosity, Pa s
η is the shape factor assumed equal to the reciprocal tortuosity, dimensionless
R is the universal gas constant, J K^{-1} mol^{-1}
r is the pore radius, m
T is the temperature, K
P_{av} is the mean pressure, Pa

2.2.2 Knudsen Diffusion

This may occur when the pore size is larger than that of the gas molecules but smaller than its mean free path (λ). There is elastic collision between the gas molecules and the pore wall and therefore no interaction between them. The permeance is given as (Domenico De Meis, 2017; Oyama, 2011):

$$P_e = \frac{2\varepsilon\eta r v}{3RT} \quad (\text{mol m}^{-1}\ \text{s}^{-1}\ \text{Pa}^{-1}) \tag{4}$$

Where,
Ɛ is the porosity, dimensionless
η is the shape factor assumed equal to the reciprocal tortuosity, dimensionless
r is the pore radius, m
v is the molecular velocity, ms^{-1}
R is the universal gas constant, J K^{-1} mol^{-1}
T is the temperature, K

2.2.3 Surface Diffusion

This occurs at low temperatures where contact between the gas molecules and inner surface is so strong compared to their kinetic energy such that the molecules cannot escape. The permeance is given as (Domenico De Meis, 2017; Oyama, 2011):

$$P_{SD} = P_O \exp\left(\frac{-\Delta Ha - \Delta Esd}{RT}\right) \tag{5}$$

Where,
P_0 is the pressure, Pa
(-ΔHa – ΔEsd) is the energy barrier for diffusing molecules to permeate through the membrane, J m^{-1} s^{-1}
R is the universal gas constant, J K^{-1} mol^{-1}
T is the temperature, K

2.2.4 Capillary Condensation

Capillary condensation usually occurs at higher gas pressures with temperatures lower than the critical temperature. Therefore, condensed gas molecules are transported across the pores of the membrane (Uchytil, Petrickovic, Thomas, & Seidel-Morgenstern, 2003; Uhlhorn, Keizer, & Burggraaf, 1992):

$$\frac{\rho RT}{M} \ln \frac{P_t}{P_o} = -\frac{2\sigma cos\theta}{r} \tag{6}$$

Where,
r is the density, kg/m^3
M is the gas molecular weight, kg mol^{-1}
q is the contact angle
P_t is the total pressure, Pa
s is the interfacial tension, N/m
r is the radius, m
R is the universal gas constant, J K^{-1} mol^{-1}
T is the temperature, K
P_o is the vapor pressure, Pa

2.2.5 Molecular Sieving

This mechanism separates the molecules by their size using membrane pores of similar size of the molecules. The typical pore sizes for molecular sieving are less than 2nm (Nagy, 2019; Rackley, 2017).

$$J_s(T) \sim \rho q_{sat} D_S^0(0)(1-\theta)^{-1} \exp(-E_{D,S}/RT) d\theta/dx \tag{7}$$

Where,
J_s(T) is the flux at temperature T, mol/s
r is the density, kg/m^3
q_{sat} is the saturated molar volume, m^3/mol
$D_s^o(0)$ is the limiting surface diffusivity, m^2/s
q is the fraction of available adsorption sites that are occupied, dimensionless
$E_{D,S}$ is the surface diffusion activation energy, kJ/mol
R is the universal gas constant, J K^{-1} mol^{-1}
T is the temperature, K

3. METHODOLOGY

This research involved the use of a shell-and-tube system. Three different membrane modules of different pore sizes were studied with the membrane fitted into center of the tube sealed with graphite seals. Methane and carbon dioxide gases analyzed under different temperatures and pressures.

3.1 Experimental Set-up

The experimental set-up used in this work is shown in Figure 1 and Figure 2 shows the different views of the membrane support. The experimental set-up contains a gas cylinder (4) with regulator (3) which contains the feed gas, this can be sent to the membrane. It contains a heat regulator (5), pressure gauge (1), temperature indicator (7), volumetric meter (6), the membrane module that has been sealed to prevent leakage of gas and covered in heating tape with insulation (2) with an exit line through which the outlet gas flows to the fume cupboard. This chamber was set up to determine the flux of each gas through the membrane under different operating conditions.

3.2 Experimental Procedure

A leak test was conducted prior to each experiment. At the inlet, the methane gas to be analysed was fed in at a predetermined pressure and readings were recorded while operating at thermal stability of 100°C respectively. The stability of the flow meter confirmed that a steady constant driving force was being maintained.

At the outlet, there was also a flow meter to measure the flow of the outgoing gas. The flux was then obtained given that both inlet and outlet flow rates were measured.

FIGURE 1: Experimental set-up showing all equipment including; pressure gauge(1), membrane module covered with heating tape(2), gas regulator(3), gas cylinder(4), heat regulator(5), volumetric meter(6) and temperature indicator(7).

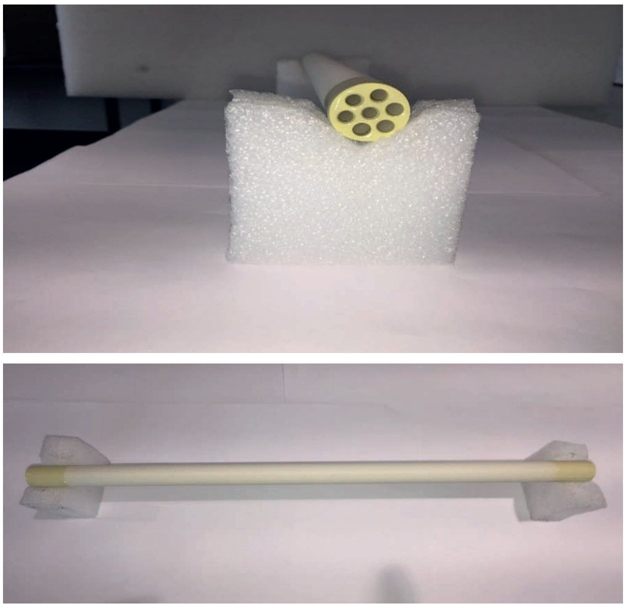

FIGURE 2: Top view (above) and side view (below) of a membrane.

The experiment was carried out at 0.2, 0.6, 1.0, 1.4, 1.8, 2.2, 2.6 and 3.0 bar.

The membrane module was flushed prior to changing the type of gas flowing through e.g. to enable us to measure the flow characteristics of carbon dioxide gas by repeating the procedure. By comparing the flow characteristics of the different gases, the perm-selectivity of the membrane was measured.

Additionally, to note how the membrane module would separate the two gases, a gas mixture containing a known composition of both gases and maintained at a set pressure would be analysed by passing the mixture through the chamber to measure the total permeation across the module and measuring how fast the outlet pressure increases. By checking the outlet gas composition, we would also measure how much of the exit gas from the total permeation is methane versus carbon dioxide which would enable us to figure out how well the membrane will separate the gases in an industrial application.

4. RESULTS AND DISCUSSION

The following set of graphs show the relationship between pressure drop and the ratio of CH_4 and CO_2 gas flowrate at set temperatures. The behaviour of the gases which is based on their molecular weight is typical of the Knudsen regime where the pore size is larger than that of the gas molecules but smaller than its mean free path (λ). There is elastic collision between the gas molecules and the pore wall and therefore no interaction between them thus permeability using these membranes are found to be in acceptable range. On the other hand, for selectivity using pure gases, the separation factor is defined by the ratio of permeability coefficient of gas A to gas B whilst the theoretical (Knudsen) separation factor is defined by:

$$\propto_{A/B} = \sqrt{\frac{MW_B}{MW_A}}$$

Where MW_B AND MW_A are the molecular weights of B and A respectively

The Figures 4 and 5 show that the 200nm and 6000nm membranes, which have very large pore sizes, do not approach ideality (i.e. the orange line depicting ratio of the square root of the molecular weight of gases) compared to the 15nm membrane. Instead, the larger pore-sized membranes show that the gas selectivities are farther away from the ideal knudsen selectivity and show a viscous flow which does not fall into the desired selectivity range. The blue plot shows the degree of selectivity of the membrane to methane gas which increased as the membrane pore decreased as the highest selectivity reached for the 6000, 200 and 15nm membranes were 1.55, 1.57 and 1.61 respectively. This means that in order to achieve higher membrane selectivity, a reduction in the pore size of the membrane is necessary. Selectivity above the knudsen range (i.e. the orange line) is desirable for effective separation of the biogas mixture. From Figure 3, it can be seen that the 15nm membrane was able to achieve the highest degree of selectivity at 1 bar compared to the selectivity values derived from other larger pore sized membranes. The results also indicate that the flux of methane through the membrane is greater than that of carbon dioxide using these membranes and portrays a good potential for upgrading of biogas which is a mixture of both. In all cases studied, the gas flux is dependent on the gas molecular weight as the heavier, CO_2 gas molecules showed lower permeability through the membrane compared to the lighter, CH_4 gas. This distinctive feature makes this technology viable for gas separation.

5. CONCLUSIONS

This research proposes membrane technology as an efficient means to transform biogas and shows good permeability characteristics for the gases studied. The results from experiments show that the molecular weight of the gases determines their permeation rate. In the case

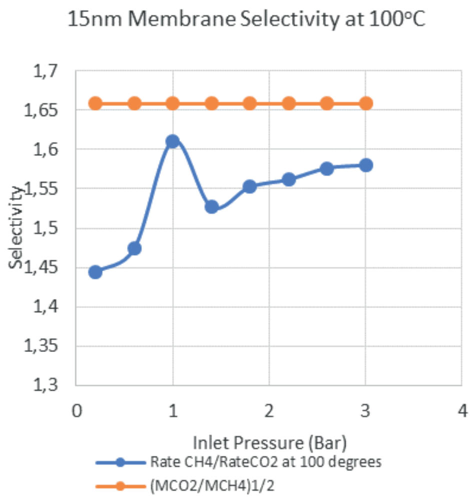

FIGURE 3: Effect of pressure drop on gas ratios at 100 degrees through a 15nm membrane.

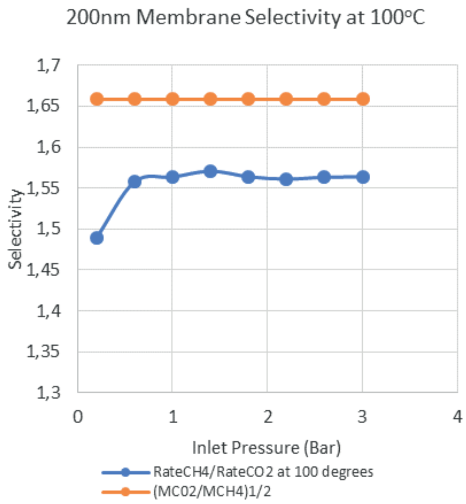

FIGURE 4: Effect of pressure drop on gas ratios at 100 degrees through a 200nm membrane.

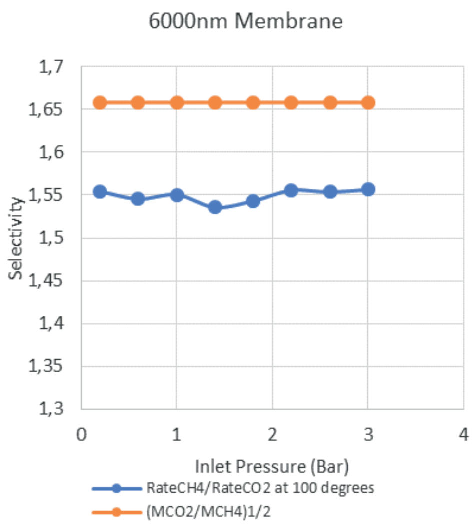

FIGURE 5: Effect of pressure drop on gas ratios at 100 degrees through a 6000nm membrane.

where the pore size is large, as in the 200nm and 6000nm membrane, there is viscous flow through the pores which is undesirable because the selectivity of the membrane is reduced. However, where membranes of smaller pore size is used, pressure drop across the membrane caused the gases to diffuse at different rates through the pores, showing increased selectivity to methane gas thus proving that gas separation can take place.

It can also be deduced from these results that the 15nm membrane shows the greatest separation efficiency as the flux of CO_2 is restricted compared to CH_4; and the ratio of methane flowrate to carbon dioxide approaches the ideal knudsen regime. An inverse relationship was observed between permeance and molecular weight of the individual gases which also confirms Knudsen flow mechanism.

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PROBLEMS IN TRADITIONAL LANDFILLING AND PROPOSALS FOR SOLUTIONS BASED ON SUSTAINABILITY

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ABSTRACT

In recent years, the Circular Economy has become the key lynchpin underlying the waste management system. However, the emphasis placed on recycling has led, on one hand, to an underestimation of the critical issues that are currently emerging so dramatically (i.e. limited recyclability of materials, instability of markets for secondary raw materials, and accumulation of contaminants present in the recycled materials), whilst on the other to neglect the inescapable role of landfill in waste management. In many cases, landfills are seen as a simple and economical means of disposing of waste, and from a political, legislative and technical viewpoint they are frequently denied the attention devoted to other engineering works, lacking adequate financial investment to cover the costs required to ensure a sustainable landfill system. Landfill should be designed and constructed in line with the principle of environmental sustainability, by adopting technical measures aimed at guaranteeing waste stability and immobilisation of contaminants over a period of less than one generation and ensuring a Final Storage Quality in equilibrium with the environment. This article summarises the concept of sustainable landfilling, identifies the technical strategies that characterise this system, describes the critical issues frequently encountered after decades of operation and proposes a series of solutions aimed to control long-term behavior.

1. INTRODUCTION

In recent years, the Circular Economy has become the key lynchpin underlying the waste management system. This has led to an increased focus on the role of recycling, viewed as a definitive solution for waste management, and to landfill being deemed an obsolete and potentially redundant system.

Recycling however is by no means a perfect system (Cossu et al., 2020b):

- not all materials can be recycled and recyclable products cannot be recycled endlessly;
- contaminants contained in the products tend to accumulate in the recycled materials and residues;
- even when materials are recycled waste will be produced;
- the system relies on the ongoing availability of the market for secondary raw materials and the recycled products obtained, whilst failing to account for instability factors (generalised economic crises, fall in price of specific primary materials, political or social crises, border closures, natural calamities, health emergencies, etc.)

Indeed, no type of waste management system can disregard a balanced use and integration of different methods of treatment and disposal, each suited to a specific field of application.

International data, routinely communicated, relating to the quantities and percentages of wastes, which, downstream of collection, are forwarded to the main treatment and management options (recycling, thermal treatment and landfilling), typically fail to include the disposal flows of treated waste. This oversight masks the effective use of both landfills and incineration. Cossu et al. (2020b) estimated the actual status quo on an international level by considering final waste disposal, which is achieved in term of end-products for recycling, in term of gasification for thermal treatment and in term of permanent deposit for landfilling. Based on this study the actual amount of waste (combustion and recycling residues) going to landfill is much higher (more than 20%) than officially declared.

In particular, landfill constitutes a necessary and irreplaceable system with which to close the material loop in a Circular Economy, providing, in line with the Back to Earth concept, a sink for all those substances and materials that

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would otherwise remain dispersed in the biosphere, thus adding to a diffuse environmental pollution (Cossu, 2016).

The general tendency to strive to "conceal" the unavoidable evidence of a landfill whilst failing to pay sufficient attention to the associated political and legislative requirements has raised a series of critical issues:

- In many cases, landfills are seen as a simple and economical means of disposing of waste, denoted by a frequent lack of technical attention devoted to other engineering works, and a paucity of financial investment required to cover the costs of ensuring a sustainable landfill system.
- The design and operation of a landfill is generally perceived as a simple task, although, in reality, a high degree of awareness, knowledge and experience are mandatory in order to avoid mistakes, which might produce serious consequences in terms of environmental impact and remediation costs (Cossu and Stegmann, 2019).
- The potential impact of landfills remains virtually unchanged due to not having been designed as sustainable systems.
- General populations are frequently fiercely opposed to the development of new landfills.
- As a consequence of anti-landfill or incinerator campaigns, large quantities of wastes are often shipped abroad. A significant example of this is represented by Slovenia in which, although classified as one of the countries featuring a lower use of landfills and high rates of recycling, the waste management system is based largely on the exportation of waste, with the Slovenian statistical office reporting the exportation of 1 million tons of waste out of a total of 6 million tons generated (Republic of Slovenia, Statistical office, 2017).

2. SUSTAINABLE LANDFILLING

Following the Rio de Janeiro Conference (1992) and setting up of the Kyoto Protocol (1997), the concept of environmental sustainability has produced a marked change internationally in the strategies applied to protect the environment. However, to date no legislation relating to landfills has contained any mention of the concept of environmental sustainability.

The Back to Earth concept should be achieved by securing, within the time frame of one-generation, a final storage quality of the landfill in equilibrium with the environment. Accordingly, the landfill should be designed and developed in line with the principle of environmental sustainability, adopting all measures required in order to guarantee waste stability and immobilisation of contaminants in a time frame of less than one generation and ensuring the reaching of an equilibrium with the environment (Heimovaara et al., 2014).

2.1 Barriers

Potential waste contaminants are essentially present in either a mobilizable or non-mobilizable form. The mobilizable fractions contained in landfilled wastes and exposed to the atmosphere are transformed and pass from one phase to another in line with their characteristics of degradability and leachability. Following these transformations, the mobilised substances accumulate in the emissions generated (leachate and gas), potential environmental pollutants. Controlling of emissions and risk of contamination in both the short and the long term, may be achieved through application of the multibarrier concept (Cossu, 2018), using a series of systems (barriers) that impinge on the following:

- Quantity of deposited wastes
- Quality of deposited wastes
- Emission control

Barriers suitable for use, graphically depicted in Figure 1, are described in Table 1, identifying the feature affected and the objectives pursued.

Pre-treatments, and in-situ treatments in particular, are essential measures with which to achieve the aims of environmental sustainability, and the choice of measures used and efficacy of the same should be specifically linked to the type and characteristics of the wastes, i.e. biological degradability, calorific potential and leachability of contaminants.

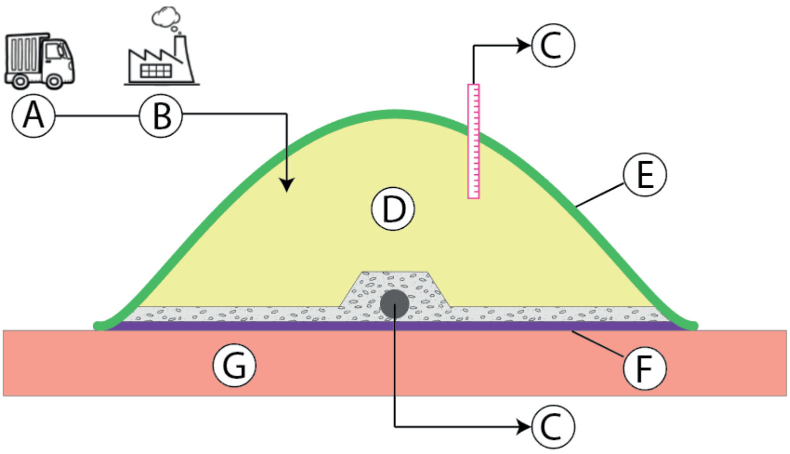

FIGURE 1: Schematic illustration of the multibarrier concept. A, Waste minimization; B, Pre-treatment; C, Biogas and leachate collection and treatment; D, In situ treatment; E, Top cover; F, Lining; G, Siting and morphology. (modified from Cossu et al., 2020c).

TABLE 1: Barriers, features affected with a view to sustainability and objectives.

	Barrier	Features affected	Aim
A	Waste avoidance and minimisation	Waste quantity	To minimize waste landfilling
B	Pre-treatment	Waste quality	To minimize the emission potential of wastes prior to landfilling
C	Biogas and leachate collection	Emission control	To maximize biogas and leachate collection and treatment by removing contaminants from the landfill
D	In situ treatment	Waste quality	To minimize the emission potential of wastes subsequent to deposition in the landfill
E	Top cover	Waste quality, Emission control	To regulate water ingress on the basis of specific process requirements. To implement cultivation layer and/or gas emission mitigation systems
F	Lining	Emission control	To minimize the uncontrolled dispersion of contaminants
G	Environmental barrier (Siting and morphology)	Emission and mechanical stability control	To exploit the self-depuration capacity of the environment to mitigate potential uncontrolled emissions. To ensure mechanical stability of the landfill

2.2 Pre-treatment

The objective of any form of treatment to be applied to wastes forwarded to final sustainable disposal should be that of removing, transforming or immobilising potential contaminants into a stable form. This aim may be achieved through the combination of a series of processes and unit operations, as described in Table 2, some of which intended to prepare the material for stabilisation (Physical treatment). The choice of intervention and expected efficacy will be linked to type and characteristics of the waste (putrescibility, calorific value, leachability of contaminants, etc.).

In the case of municipal solid waste (MSW), biological pre-treatment as well as thermal pre-treatment are practised; both further to reduce the organic content to low values (biological waste stabilisation), allow a significant volume reduction, particularly with thermal treatment.

As a general rule, the biological pre-treatment of MSW is always combined with mechanical processes to afford the so-called Mechanical Biological Treatment (MBT), the aim of which is to stabilise undifferentiated wastes prior to landfilling (Stegmann, 2018). During mechanical pre-treatment the high calorific value fraction of MSW is separated mainly by sieving. Following additional mechanical treatment, the remaining fraction will be biologically stabilised prior to landfilling.

The main effects produced by MBT on wastes to be landfilled are as follows (Leikam and Stegmann, 1999; Stegmann and Heyer, 2001, 2002):

- Weight reduction (40-70%).
- Reduction and homogenisation of granulometry.
- Increase in specific surface area with consequent potential improvement of biological stabilisation process within the landfill.
- Enhancing compaction in the landfill.
- Reduced settling rates.
- Increased instability due to reduction in granulometry and removal of elements resistant to traction (i.e. fibres and plastics) by means of mechanical treatment, to achieve maximum gradients of 1:3 on slopes.
- Reduction of hydraulic conductivity with potential increase of neutral pressure and consequent risk of mechanical instability.
- More than 90% reduction in the emission potential of the wastes with decreased COD and total nitrogen in leachate and lower stability indexes (R_{I4}, GP_{21}).

Amongst the different forms of thermal treatment (combustion, pyrolysis, gasification, etc.), combustion (or incineration) is the most widely process applied for municipal solid wastes. The residues of thermal treatment account for 10% of the initial volume and 25-30% of the initial weight, including bottom ashes, grate siftings, boiler and economizer ashes, fly ashes, and Air Pollution Control (APC) residues. The various types of residue are characterised by a diversity of physical, chemical and mineralogical properties with a markedly differentiated composition. In particular, residues such as fly ashes and residues from flue-gas treatment feature a higher concentration of metals (Sabbas et al., 2003) and toxic trace organics (e.g. halogenated hydrocarbons).

Combustion residues may be landfilled or recycled and used as road construction material meeting specific conditions (e.g. beneath an impermeable asphalt layer) or in cement production. Whichever form of final disposal is applied, the potential environmental impacts of incineration residues are generally associated with emission of dust, gas and leachate, with the latter representing the major source of concern. Pre-treatment comprising the inertisation or leaching of residues will facilitate the reduction of contaminant load or modify leachability.

If thermally or mechanical-biologically pre-treated waste is landfilled the emission potential will be significantly reduced, although emission control will still be required (Heyer, et. al, 2013).

In the opinion of the authors, a status similar to that of an MBT landfill may be achieved by implementing the separate collection of kitchen and yard waste and landfilling residual waste in a bio-reactor landfill with subsequent in-situ aeration. Of course, this process needs significantly longer time to reach the emission values expected from MBT landfills.

The physicochemical treatment of waste washing (or elution) is aimed at separating wastes to facilitate the re-

TABLE 2: Classification and description of unit operations applied in waste pre-treatment or in-situ treatment according to process type.

	Process	Unit operations	Aim
Pre-treatments	Physical	• Shredding • Size separation (Sieving)	• Pre-treatment, recovery of RDF
	Biological	• Anaerobic digestion • Aerobic stabilisation • Composting	• Biological stabilisation • Resource recovery (methane, hydrogen, compost)
	Thermal	• Combustion • Pyrolysis* • Gasification* • Thermal inertisation (vitrification, melting, sintering)	• Reduction of waste volume • Destroying of organic contaminants • Energy recovery • Recycling of ashes
	Physical-Chemical	• Washing (with or without chemical agents) of inorganic waste	• Removal of contaminants • Reduction of emission potential
	Chemical - Physical	• Solidification by both inorganic (e.g. Hydraulic binder) and organic (e.g. Thermoplastic materials) reagents.	• Reducing leachability • Increasing mechanical stabilisation • Resource recovery (recycling of stabilised material)
In situ treatment (Bioreactor landfills)	Biological	• Controlled leachate recirculation • In-situ aeration • Natural/passive aeration	• Biological stabilisation

** Mainly applied in Japan or occasionally in other countries for industrial waste*

covery of recyclable materials and/or at reducing emission potential of the waste with a view to sustainable disposal (GeoSyntec, 2003). During the washing process, any leachable compounds are transferred from the solid to the liquid phase. In the case of inorganic substances the efficacy of the process depends on a series of factors, including solubility, pH, redox potential, and presence of chlorides and dissolved organic matter.

Another physicochemical treatment method to significantly reduce the leachability of suitable solid wastes with a prevalently inorganic matrix is called inertisation or solidification. The advantage of some of these forms of pre-treatment is represented by the possibility of achieving a higher efficiency compared to that afforded by washing, resulting in some instances in the possibility of recycling the inertised product for use a secondary raw material (Andreola et al., 2017). The main drawback is associated with the need to use additives (binders, reagents, etc.) and/or high amounts of energy resulting in a costly treatment. Often the long- term effects of the solidified waste are not well known. In case of elution high amounts of water are used, sometimes with the addition of chemicals (control pH, etc.), and the eluted sludge like compounds have to be further treated or landfilled.

2.3 In situ treatment (Landfill Bioreactors)

Due to evident economical and technical issues, waste pre-treatment prior to landfilling may fail to result in a sufficient stabilisation of wastes – either in terms of biological stability or in terms of stability against leaching – to guarantee an adequate final storage quality. This may still be achieved by means of in-situ treatments.

These measures may be implemented either during the operational phase of the landfill or during the post-management phase and should be selected, dimensioned and described in detail during the design stage.

Potential in-situ treatments may comprise one or more of the following options (Table 2):

- Controlled leachate recirculation
- Natural aeration
- Forced aeration

Controlled water infiltration (in general treated or untreated leachate recirculation) may promote the biodegradation processes due to an increase in moisture content of the waste and water flux. This approach makes only sense when the water content of the waste is insufficient to promote biological processes. High addition of water or leachate may result in water ponding, which may reduce the stability of refuse mounds. Moreover, the option of providing for the sole leaching of landfilled waste by means of addition of high water volumes for the purpose of releasing a landfill from aftercare after a relatively short period (comparable to in-situ aeration) is, in the opinion of the authors, neither practical nor feasible. The findings of the international research project EVAPASSOLD demonstrated that to reduce the emission potential of a landfill to environmentally acceptable concentrations, water volumes corresponding to approx. 3-5-fold the deposited waste volume (liquid/solid factor 3-5) will need to migrate through the waste, which may not prove to be a feasible option (Allgaier and Stegmann and Heyer, 2002).

Landfills adopting the different unit operations either individually or in combination are typically recognized as landfill bioreactors, which are categorized into the following types:

- Anaerobic landfill, featuring – where appropriate - a controlled recirculation of leachate.
- Aerated landfill, comprising the injection of air at low pressure through vertical or horizontal wells into the landfill body and simultaneous extraction of the corresponding amount of gas, which will require treatment prior to release into the environment. In view of the authors in-situ aeration shall only be applied to landfills with a low content of organic degradable matter (Ritzkowski, et al., 2006, Ritzkowski, et. al, 2013, Hupe and Stegmann, 2013).
- Semi-aerobic landfill, which thanks to the specific

construction of a system of chimneys and pipelines, is characterised by a natural convective circulation of air obtained through the difference in temperature between the waste mass and the external environment (Matsufuji et al., 2018).

- Hybrid landfill, with a sequence/alternation of aerobic and anaerobic conditions (Cossu and Grossule, 2018). This concept has been mainly applied in lab studies and relevance in full scale application is lacking.

Achievement of the aims of environmental sustainability is guaranteed by the multiple benefits afforded by in-situ treatments, including reduced time frame for interventions and lower post-management costs, shorter duration of environmental responsibility for the landfill management, accelerated reduction of emission potential (due to an increased degradation and leaching) and mechanical stabilisation of the waste mass. On the other hand, the main disadvantages may include higher costs, higher complexity of construction, management and operation.

Specific advantages and disadvantages of each landfill bioreactor category are summarized in Table 3.

In the opinion of the authors the only method, by which an enhanced reduction of the residual emission potential of a closed landfill can be achieved, is the in-situ-aeration. In many cases the existing landfill gas extraction system with some additions can be used. The results gained from actual full- scale aerobic in situ stabilization landfill projects show an enhanced biological stabilization with accelerated reduction of settlements (Ritzkowski et. al., 2006). This process may be supported by a controlled water infiltration (Stegmann, 2017a).

2.4 Final storage quality and closure of the aftercare

A landfill may be deemed sustainable if - following the integration of a series of barriers - the emission potential of the landfilled waste and the quality of emissions reached is such as to not produce any non acceptable effect on the environment. This status should be reached over the time frame of one generation.

This quality is typically defined as final storage quality (FSQ), according to which, on terminating management of a sustainable landfill, the remaining uncontrolled contaminant load will not necessarily correspond to zero. It is mandatory however that residual pollutants should be compatible with the surrounding environment and guarantee an equilibrium that does not perturb the quality of the environment, whilst exploiting its ability to self-depurate.

The design of a sustainable landfill therefore should be based on the ability to achieve the FSQ, subsequent to which the main aftercare phase is terminated, which may release the landfill operators from any form of environmental liability. Anyway, in the opinion of the authors, a certain monitoring and maintenance of a landfill will be always necessary for a non-predictable time.

The question as to when a landfill can be released from aftercare is difficult to answer. Some studies have defined FSQ by proposing limit values for landfill emissions (leachate quality and load, gas production, etc.). In Germany, several studies put forward FSQ values for use by the German Federal Environment Ministry (Stegmann, et al., 2006, Stegmann, et al., 2003, Stegmann, et al., 2011), whilst in Italy FSQ values were proposed by a national technical committee (CTD) in 2006 and implemented in the official regulation for sustainable landfilling of the Lombardia Region (Deliberation of the Regional Council 2461/14; Cossu et al., 2020d).

The combination of pre-treatments and in-situ treatment should be envisaged with an aim to pursuing the FSQ objective, exploiting their complementarity to reduce the emission potential of wastes (S_t), as graphically represented in Figure 2.

It is evident that financial provisions to be ensured over a defined time frame (in Italy, 30 years) are not sufficient to guarantee Final Storage Quality (FSQ) of the landfill. If no technical interventions are implemented to achieve the FSQ, on termination of the financial provisions, the place of what should simply be a closed landfill will be taken by a contaminated site with unsustainably high costs of recovery.

Aftercare costs are frequently overlooked or not given due consideration in budget planning. For a long period,

TABLE 3: Categorisation of different landfill bioreactors, identifying the characteristic unit operation and specific advantages and disadvantages (Cossu,and Grossule, 2018; Grossule et al., 2018; Grossule and Lavagnolo, 2019; Ritzkowski and Stegmann, 2003, 2007).

Bioreactor type	Unit operations	Advantages	Disadvantages
Anaerobic	Leachate recirculation *	• Improvement of biodegradation processes by controlling water content and flux • Improvement of leachate quality • Biogas generation enhancement, concentrated to a shorter period of time • Better removal of soluble compounds**, including ammonia under anaerobic conditions and organic acids from the acidic and acetogenic phase, reducing possible inhibition of fermentation phase	• Costs of recirculation • Potential of leachate ponding
Aerobic	Forced aeration	• Acceleration of biodegradation kinetics • Better waste settling • Reduction of uncontrolled methane dispersion • Nitrogen removal	Energy demand
Semi-aerobic	Natural aeration	• Acceleration of biodegradation kinetics • Nitrogen removal • Low cost system	Careful management and operation for optimal system performance

** Leachate recirculation, not applicable at high water content of waste mass, **It has to be distinguished between raw or treated leachate recirculation.*

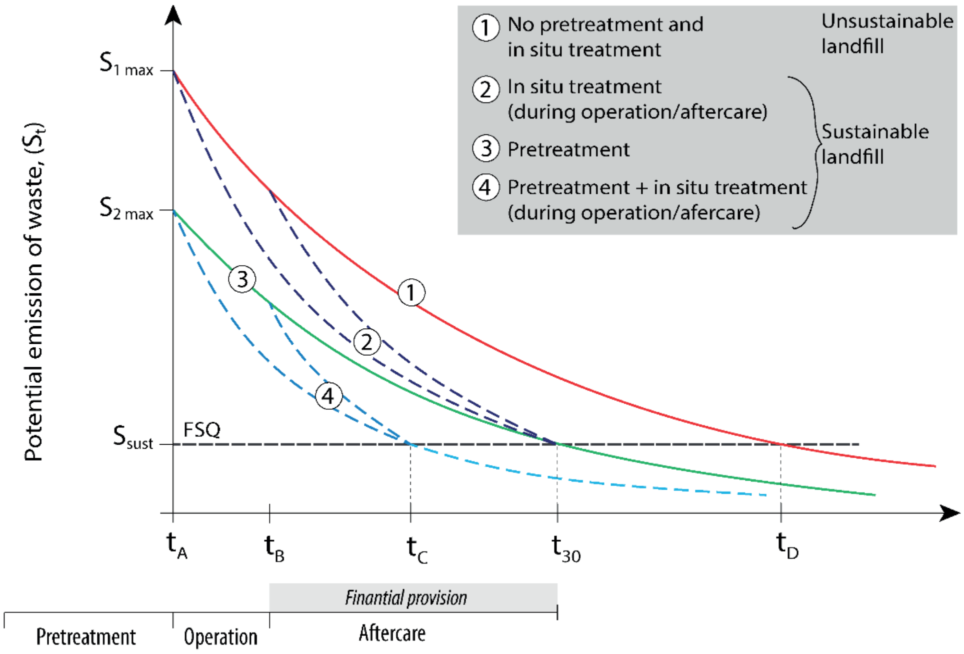

FIGURE 2: Time trend of the emission potential for release of contaminants from a landfill. FSQ= Final Storage Quality, according to which the emission potential reaches a value of S_{sust} in equilibrium with the environment; t_A, peak in emission potential during landfill operations; t_B, Closure of landfill operations and commencement of aftercare; t_C, Anticipated achievement of FSQ; t_{30}, Sustainability target and termination of financial provisions; t_D, FSQ beyond the 30-year threshold (unsustainable landfill). (Modified from Cossu et al., 2020c).

the unrealistic opinion was held that 30 years after closure landfills no longer require special care and - if operated by the private sector - may be returned to the communities. Experience however has clearly demonstrated that non-sustainable landfill aftercare will be required for numerous decades and, in the case of large landfills, possibly even a century or more. This is also the case for landfills with a low emission potential (Heyer, 2018).

Potential combinations of pre-treatments and in-situ treatments suitable for reaching the status of a sustainable MSW landfill in due time are graphically displayed in Figure 3.

3. TYPICAL TECHNICAL AND REGULATIVE PROBLEMS ASSOCIATED WITH TRADITIONAL NON-SUSTAINABLE LANDFILLS AND SOLUTIONS

Landfill technologies implemented worldwide vary considerably. In low-income countries often open dumps are

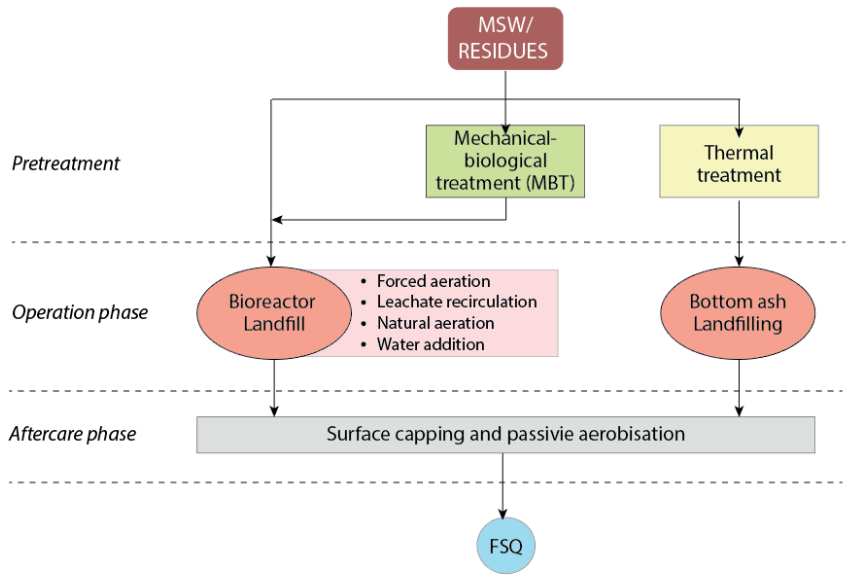

FIGURE 3: Scheme illustrating the possible combinations of pre-treatment and in-situ treatment options for municipal solid waste (MSW) with a view to ensuring landfill sustainability and achievement of FSQ.

in use in which all kinds of wastes are disposed. This form of disposal implies a total lack of emission control, absence of waste compaction, sites selected with no regard to reducing environmental impact and very steep slopes. Waste dumping results in a range of problems, including: mechanical instability, fires, littering, odours, uncontrolled leachate, gas and toxic fumes emissions. On the other hand, the transfer of modern landfill systems, developed in industrialized countries, to tropical countries should be carefully managed to avoid failures, whilst bearing in mind the specific local situation (different waste composition, high temperatures and strong rainfall periods) (Lavagnolo and Grossule, 2018).

Conversely, even in industrialised countries, although landfills have undergone marked technical advances, a series of problematic issues raise. These are linked largely to reliance on physical containment measures alone (lining and leachate and gas extraction systems), despite the finite duration of this form of containment. Increasingly, drainage systems become clogged, drainpipes collapse, gas extraction wells lose functionality, and plastic membranes fail to maintain their initial quality. Moreover, the problem of long-term landfill emissions remains inadequately addressed (Heyer, et.al. 2005). Landfills will continue to require regular maintenance and repairs (depreciation period for buildings approx.30 years) but the duration of ongoing aftercare remains unclear and commonly, insufficient funds are earmarked for aftercare costs.

The lack of an appropriate landfill design project corresponding to the principles of sustainability based on use of the above-mentioned technical strategies, has resulted in a series of critical aspects highlighting both regulatory and design inadequacies. Typical technical issues in modern non-sustainable landfills are listed in Table 4 and classified according to the barriers used (previously illustrated in Figure 1, Table 1). In particular, the following paragraphs describe in detail these technical issues and provide potential technical solutions.

3.1 Waste minimisation

The minimisation of waste forwarded to landfill is one of the key European strategies aimed at "preventing det-

TABLE 4: Typical technical problems in modern non-sustainable landfills.

Barriers	Critical element	Consequences	Environmental impact
A	Waste minimisation as the only strategy to address impacts from landfills	Absence of a legislative and technical upgrade of traditional landfill towards the sustainability concept	Environmental unsustainability
B	Pre-treatment is not designed to foster achievement of FSQ	Long-term environmental impact	Environmental sustainability may be reached after very long times; many decades to century
C	Legal ban of acceptance of high calorific value wastes	Recycled high calorific value wastes have to be thermal treated	"carbon sink" only for humic like substances as a result of biological treatment
C	Inadequate design of drainage system including materials	Drainage dysfunction and inefficiencies/high water table	Increased risk of pollution and mechanical instability
C	Use of geotextiles to protect drainage bed	Premature clogging of drainage system / Leachate build-up	Increased risk of pollution, odours
C	Limited long-term functionality of drainage system including collection pipes	Drainage dysfunction and pipe rapture /high water table	Increased risk of pollution, odours, loss of mechanical stability
C	Gas wells and pipes	Uncontrolled gas emissions	Environmental unsustainability
D	Neglect of scientific development relating to in situ waste stabilisation (in-situ aeration, leachate recirculation)	Insufficient biological stabilisation and leaching of contaminants	Environmental unsustainability
D	Scarce attention to quality of daily cover	Reduced permeability of waste mass, ponding, gas circulation hampered	Odours, mechanical instability, dust
D	Build-up of high temperatures	Inhibition of methane generation, deformation of plastic liners and pipes	Increased risk of pollution, risk of fires
D	Lack of definition of Final storage quality (FSQ)	Uncertainty when releasing a landfill from aftercare	Uncertainty regarding aftercare period
E	Landfill top sealing directly after end of operation	Insufficient water supply, reduced stabilisation, conservation of emission potential of waste	Environmental unsustainability
E	Scarce concern over potential use of methane-oxidizing cover	Escaping of the dispersed residual biogas	Environmental unsustainability
F	Duration of geomembranes	Rupture, leachate infiltration	Increased risk of pollution
F	Duration of efficiency of clay layers	Leachate infiltration	Increased risk of pollution
G	Scarce consideration of the morphological features of the landfill	Incongruous insertion into the landscape, unstable drainage	Inconvenience for the population
G	Scarce consideration for landfill siting taking into account natural capacity for attenuation	Landfill development below the natural surface levels, even in sensitive areas	Increased risk of pollution
Other	Height limit for the landfill above ground	Steep slopes, unsightly appearance, extension of deposit below natural surface levels	Impact on the landscape, risk of instability, precarious runoff
Other	Viewing of the landfill as an economic system	No investment aimed at strengthening barriers	Environmental unsustainability

rimental impacts on human health and the environment" ascribed to landfills (2018/850/EU). However, the Circular Economy is quite far to reach the "Zero Waste" target, and landfill continues to play an unavoidable role in receiving residual wastes within the Circular Economy. Although the quantities of waste forwarded to landfill have decreased significantly, the change in quality and long-term impacts produced have been somewhat overlooked from a regulatory and design viewpoint. In particular, although the amounts of putrescible organic substances have been reduced, little is known with regard to a potential increase in inorganic substances (including heavy metals) and persistent organic substances that tend to accumulate during waste recycling.

With regard to paper for example, a study conducted by Pivnenko et al. (2015) identified 51 hazardous substances (mineral oils, phthalates, phenols, parabens and other groups of chemical compounds) that tend to accumulate during the recycling process. Many of these compounds have been detected at higher concentrations in recycled paper compared to paper produced from the raw material.

3.2 Pre-treatment

In the majority of cases, both from a regulatory and design viewpoint, a strategic approach to waste pre-treatment prior to landfilling is lacking. Indeed, although European regulations establish acceptance criteria, these fail to guarantee sustainability and compliance with achievement of a specific final storage quality required for termination of the aftercare period.

The lack of a comprehensive strategic vision relating to waste pre-treatment is highlighted by the classification of landfill based on type of wastes deposited (inert, non-hazardous, hazardous). In the presence of an increased hazardousness, the regulations merely prescribe an increased thickness of physical barriers, failing to implement any significant measures to reduce the potential contaminant load by means of appropriate pre-treatment or in-situ treatment with a view to achieving environmental sustainability.

Lastly, the prohibition of landfilling for wastes with a high stable TOC or calorific value will ineluctably rule out the potential role of landfill as a carbon sink, which would contribute towards reducing the impact on generation of greenhouse gases (Cossu, 2012). The principles established by the EU whereby the landfilling of wastes potentially suited to recycling or other forms of recovery is prohibited, with the exception of wastes for which landfilling is indicated as representing the most effective environmental solution (2018/850/CE). As a compromise the authors support the idea of landfills as an intermediate storage area for high calorific materials as plastic, in case there are no options for material or energy recovery.

3.3 Biogas and leachate collection

A series of critical aspects have emerged with regard to leachate drainage.

- Inadequacy of the granular drainage material used (in terms of granulometry, homogeneity, cleanliness, resistance, etc.) may result in clogging of the drainage system, potentially compounded in the case of leachates having a high biodegradable organic load, due to the formation of bacterial film combined with precipitation of iron and carbonates. In view of the unfeasibility of intervening directly to maintain drainage, it is fundamental that the drainage bed covers the entire bottom of a landfill and is made up of large, homogenously-sized inert material (mainly gravel) in order to provide a large volume and to avoid/reduce clogging. In addition it increases the movement of leachate on the bottom liner (slope towards the drainpipes >3%) towards the drain pipes and facilitates the transfer of potential clogging material through the drainage pipes (the sole inspection point throughout the entire drainage system). Therefore the slots in the drainpipes should be around 1 cm wide. In order to reduce organic leachate concentrations, the authors recommend waste pre-treatment and installing a layer of up to ± 2 m of composted MSW on top of the gravel layer of the leachate collection system. This composted waste layer will act as an anaerobic filter for the migrating leachate from the upper waste layers (Stegmann, 1995, Lavagnolo, et. al. 2018). In this way, the readily degradable organics are partly anaerobically degraded to biogas. Moreover, the composted MSW layer will also contribute positively towards reducing incrustations in the leachate collection system, as these are strongly associated with the presence of high organic acid concentrations in leachate (Brune et al., 1991). This concept has been successfully implemented in a series of landfills in Germany (Stegmann, 1995). Finally, the use of geotextiles to protect the drainage bed against clogging, frequently prescribed in Italy and other countries by the competent authorities prior to issuing of authorisation, may actually worsen the problem rather than solving it.
- The long-term functionality of the drainage system is naturally subject to degeneration: clogging of the pipes due to incrustations is widely observed, despite the regular inspection and cleaning (by high pressure flushing) of the drainpipes; deformation or collapse of drainpipes may occur even when using prescribed quality material. However, an inadequate design, inappropriate material quality and a lack of maintenance (high pressure flushing 1-2 times a year) may further limit the lifespan of the drainage system. Moreover, unless the main drain headers are located close enough (< 50m), "fish bone" type drain systems should be avoided, because they are difficult or impossible to clean. In addition maintenance of drainpipe is more complex in landfills constructed in a pit. Ceramic pipes, which were used in Germany in the 1980s, failed to resist and were found to be unstable and partly collapse, hampering or preventing the removal of incrustations and loosing its functionality. HDPE pipes may become brittle/deformed after decades of emplacement, and this may occur also to HDPE liners (Löwe, 2016). As a result, also in order to meet the legal requirements, drainpipes will need to be repaired or replaced, a very costly task, the result of which may be far from desirable. The same

methods used in the repair of sewage pipes have also been applied in the remediation of landfill drainpipes. One procedure envisages a relining process obtained by placing new pipes inside collapsed ones (a frequently impossible task). Other methods involve digging down to the bottom of the landfill and substituting pipes. The burst-lining technology may also be applied, in which a new pipe is pressed from one manhole to another. In some cases 3-5m diameter manholes have to be constructed in 30-40 high landfills, always respecting the strict work safety procedures. All these procedures are very expensive and even if pipe functionality is restored, the drainage system surrounding the pipe may remain clogged from incrustations.
- Construction of large diameter concrete vertical leachate collection shafts throughout the waste mass, which in views of the authors should be avoided, may be subject to strong deformation, rupture, and impossibility to carry out inspections.

The critical aspects listed above unfailingly contribute to the formation of high water tables (which may, dependent on landfill height under Mid-European climate conditions, reach levels of between around 10 to 15 m). Water storage may also be layered water in landfills. In particular, in line with an increase in landfill height (>15-20m), pressure exerted on the waste and accordingly density decreases in the lower part of the landfill. In addition in the lower part of a landfill gas competes with water in the waste pores, which results in a further decrease of the permeability of the waste mass (may be 1-2 orders of magnitude or even more), resulting in a number of serious issues (Figure 4):

- Due to the weight of the overlying waste and water-saturated pores of the waste material, water overpressure may build up. This situation results in lower friction values of the landfilled waste and consequent reduced mechanical stability. Therefore pore water pressures should be monitored and – if necessary – appropriate measures taken (e.g. dams at the foot of landfills, reduced slopes, water level reduction). This is particularly important for landfills in areas with high precipitation rates and/or in cases when landfilled wastes are characterised by relatively high moisture content. Due to a lower pore volume and relatively high sorption capacity, MBT landfills may need to be especially carefully monitored.
- A rise of the water table may increase the rate of leachate migration through a mineral clay layer placed at the bottom of the landfill, which does not provide complete impermeability.
- A regular distribution of the negative pressure values for gas extraction can be hampered and positive pressure values might originate causing uncontrolled gas and odours emissions;
- Limited circulation of gas (biogas under anaerobic conditions, air under aerobic conditions).
- Uncontrolled discharge of leachate.
- To extract the leachate and reduce elevated water tables, as well as in cases in which the drainage system fails to function adequately, vertical wells should be drilled to the bottom of the landfill and the leachate pumped out (Figure 5).

3.4 In-situ treatment (Landfill Bioreactors)

The role of this type of barrier is often completely neglected both from a regulatory and design viewpoint:

- The possibility of regulating the presence of water to be used in biodegradation is frequently overlooked, with leachate recirculation being prohibited by the authorities.
- There is a lack of cultural openness to the use of in-situ waste treatment systems (in-situ aeration, semi-aerobic landfill, etc.), largely due to the perception that the effectiveness of this technology has not been well demonstrated, although this technology is practiced in several countries since many years and has proven its applicability and success (Ritzkowsli and Stegmann, 2018).

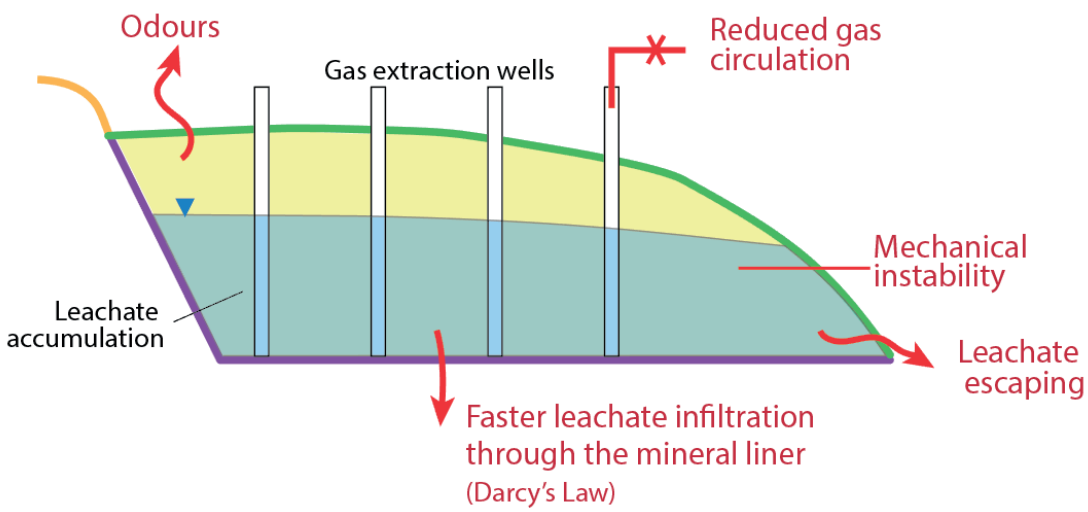

FIGURE 4: Graphical illustration of the main issues associated with the build-up of high water table in landfill, (modified from Cossu et al., 2020c).

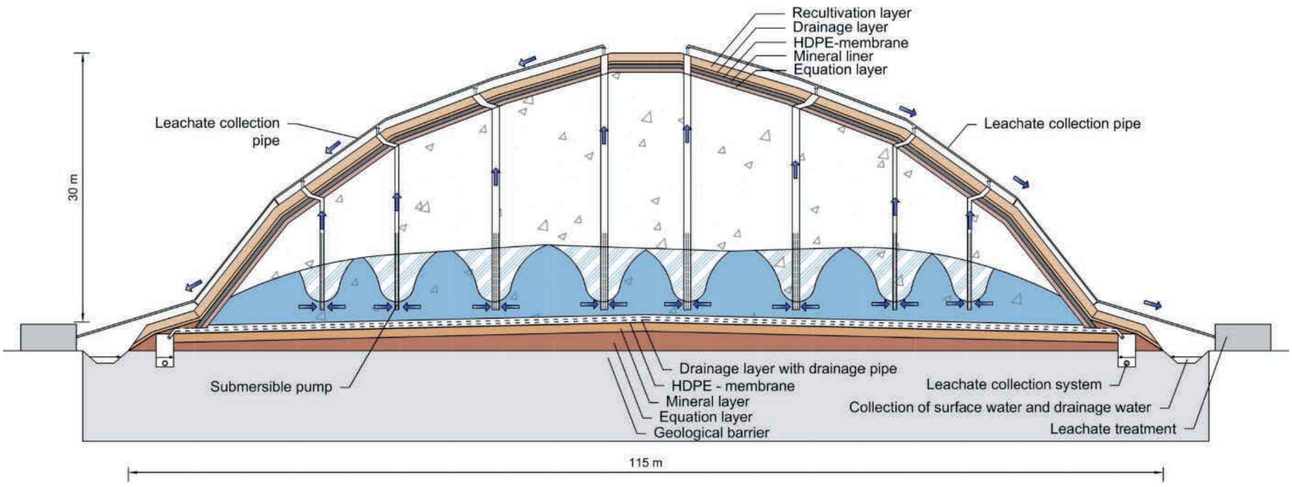

FIGURE 5: Lowering water table by means of vertical wells with submersible pumps.

- Scarce attention paid to the quality of materials used for daily cover, which in some countries is also applied for aesthetical reasons. In particular, the use of soil with low permeability should be avoided. In Germany on most landfills no daily cover is used due to the immediate high compaction of the incoming waste by heavy weight compactors.
- Relatively high temperatures (in some cases > 65°C) observed in several high/deep landfills is another phenomenon that needs to be better addressed and investigated. In the opinion of the authors these occur typically due to the production of microbiological heat and low heat transfer in the waste (low water flux and significant insulation capacity of the landfilled waste). The high temperature, shift of NH4+ to NH3 (a highly toxic gas, toxicity limit around 150 mg/L free NH3) and the accumulation of organic acids due to low water flux may result in the inhibition of methanogenic bacteria and production of highly concentrated leachate. In addition chemical processes may also be responsible. High temperatures may also affect stability of the plastic liners and pipes.
- No final storage quality (FSQ) which is compatible with the environment and on which to base ad hoc projects and initiatives is defined for the landfill.

3.5 Top cover

The flexible role carried out by a final cover, based on both the type of landfill and quality of landfilled waste, with an aim to achieving environmental sustainability, is completely neglected. European legislation enforced throughout a series of countries establishes the need for landfills or landfill sections to be immediately capped once they have been filled to their final height. As a result, the emission potential of the landfill will be preserved (resulting in the so called dry tomb landfill, Stegmann, 1993). However, in view of the fact that surface liners may not remain effective indefinitely, emissions from the landfilled waste may occur once landfill operations are no longer monitored. This concept may only be applied to landfills where non-biological degradable waste alone is landfilled (e.g. different kinds of inert waste and hazardous and industrial wastes). In the case of wastes containing degradable compounds (MSW), water content and, accordingly, water infiltration through the top cover should be controlled with the aim of promoting degradation of organic substances.

Another aspect that is frequently overlooked is related to the potential effects that surface covering of the landfill may produce on the oxidation of residual biogas emissions (small quantities and low methane concentrations). Such top layers of fractions should consist of composted material e.g. deriving from separate collection and treated by means of active biological stabilisation. Indeed, by including zones of bio-oxidising material (compost, wood shavings, etc.) in the cover structure, methane can be oxidised and contaminants such as Chlorofluorocarbons (CFC) present in the gas, degraded (Kjeldsen and Scheutz, 2018).

3.6 Lining

The main role in controlling landfill emissions has long been attributed to the lining system, although this does not guarantee a perfect and eternal insulation of the landfill from the surrounding environment.

If geomembranes are applied, lesions may potentially be manifested even during installation due to faulty welding. In addition to damage and breakages may occur during the management phase due to inadequate mechanical protection. Likewise, when using mineral materials, the creation of a layer that complies fully with the established legal requirements is not easy to achieve. In addition to an initial high quality material, rules of good geotechnical practice should be adhered to (compaction, dimensions of layers, type of compaction, humidity, controlled presence of granular material, etc.) and appropriate monitoring of both the bottom liner and frequently neglected slopes. In the latter case moreover, care should be taken to avoid degradation caused by atmospheric agents (erosion, drying, cracking, etc.). The latter aspects are frequently overlooked, thus resulting in potential dysfunctions and problems that are only detected subsequently. On the other hand, clay liners do not provide total permeability, but rather theoretically (1m hydraulic head, homogeneous layer, water as leaching liquid) a 1m layer of clay with a permeability k=10-9m/s (typically prescribed by legislation), will expedite (Darcy's Law) the passage of liquid over a time frame of little more than thirty years. The actual timings involved have however been shortened even further in the presence of leachate rather than water, in addition to the other critical factors highlighted (faulty installation, environmental degradation, etc.) (Cancelli et al., 1994).

The limited duration of the physical barriers should therefore be taken into account during the design stage in order to allow appropriate measures to be implemented. Should the ability of these barriers to contain emissions not be perpetual then it should be ensured by means of pretreatment and/or in-situ aeration that non acceptable emissions do not last longer than the lifetime of the physical barriers.

3.7 Environmental barrier

The environmental barrier (siting, morphology of the landfill, attenuation capacity of the unsaturated ground and water tables) is frequently overlooked during the design stage, although it is frequently revisited following the occurrence of cases of pollution.

Although lining systems are mandatory (Stief, 1989), appropriate landfill siting is also essential and may reduce the environmental impact of a landfill significantly. Areas having low permeability subsoil, where surface water is unable to enter the landfill and the ground water table is either very low and/or the quality of the water is too poor to use as drinking water (e.g. high natural salt content) are preferable for siting of a landfill.

Landfill morphology may negatively affect the efficiency of the drainage system; for this reason, morphologies envisaging deposition only above ground level are to be preferred. Landfills should be constructed as mounds in order to control leachate in the long-term, particularly following closure: leachate drainage systems can thus be better maintained and leachate runoff from the landfill will occur by gravity; conversely if landfills are constructed in pits, leachate will need to be persistently pumped from the bottom of the landfill. Moreover, waste deposition above ground level provides the advantage of a greater distance between the bottom of the landfill and the groundwater tables, also facilitating intervention when needed (maintenance, "landfill mining", etc.). Landfills should not be located in a valley, as in this case existing watercourses are often piped and waste is landfilled on top of these pipes. Pipes may subsequently become subject to leakage and the watercourse will be polluted by leachate (Cate, F.M., 1993). When landfills are built in mountainous areas on a mountain slope, a well-designed water control system (drainage system) should be installed between the mountain and the landfill, and unpolluted water transported separately out of the landfill site.

Aspects relating to the architectural design and economic planning of a landfill will impinge considerably on its environmental sustainability. Often a functional planning and designing of a landfill based on intended after use and insertion into the landscape is missing. Due to the wish to conceal view of the landfill, and the availability of gravel or mining pits may result in the construction of landfills below ground level and an ugly and scarcely functional aesthetics. Landfill should not be viewed as the final use of a specific area, but rather as an architectural structure intended to provide a series of functions geared to life of the community.

4. NEW LANDFILL CONCEPT

In order to better control long-term functionality of the installations, to ensure long term effective emission control and to avoid/reduce the above described landfill problems, landfills should be constructed and operated in a modified way. As a way to deal with many of these challenges the installation of horizontal layers with slopes to the outside of a landfill mound is proposed (Stegmann, 2017b). These layers should consist of inert coarse material at different levels of the landfill and should be constructed during landfill operation (Figure 6). The layers should be placed at vertical distances of approx. every 3m in the lower part of a landfill (up to a height of about 12-15 m), and subsequently – when the permeability is somewhat higher - every 5m. If high water content waste is landfilled the coarse layers may be installed every 3m throughout the entire landfill height (e.g. situation in countries with a very high content of putrescible waste). These layers should be approx. 30-50 cm thick and consist of coarse carbonate "free" material - preferably with a largely homogenous grain size (about ± 30 mm)-; the material should be free of fines in order to avoid clogging and readily allow water and gas movement. Drainpipes perforated at their upper part may be installed to support leachate run-off and for monitoring. Once these layers have been installed, the amount of daily cover may be reduced and the horizontal layers used for multiple purposes:

- to avoid/significantly reduce water build-up in the landfill
- to allow better compaction and operation of high moisture containing waste
- to increase mechanical stability (higher friction angle)
- to allow gas extraction at an early stage during operation, avoiding significant climate gas emissions (about 20-30 % of the total gas production potential), and by these means increase energy production.
- to be used - once gas utilization is no longer possible/feasible – for in-situ aeration and later for passive aeration to maintain the landfill under high aerobic conditions
- to reduce the build-up of high temperatures and inhibition of methane production due to the reduction of pore water pressures and water storage
- to reduce clogging of the base drainage layer due to more homogeneous temperatures and allow precipitation in the upper drainage layers

Positive experiences have been gained using horizontal sand layers from a landfill filled with dredged material from Hamburg harbour. In addition horizontal drain layers were

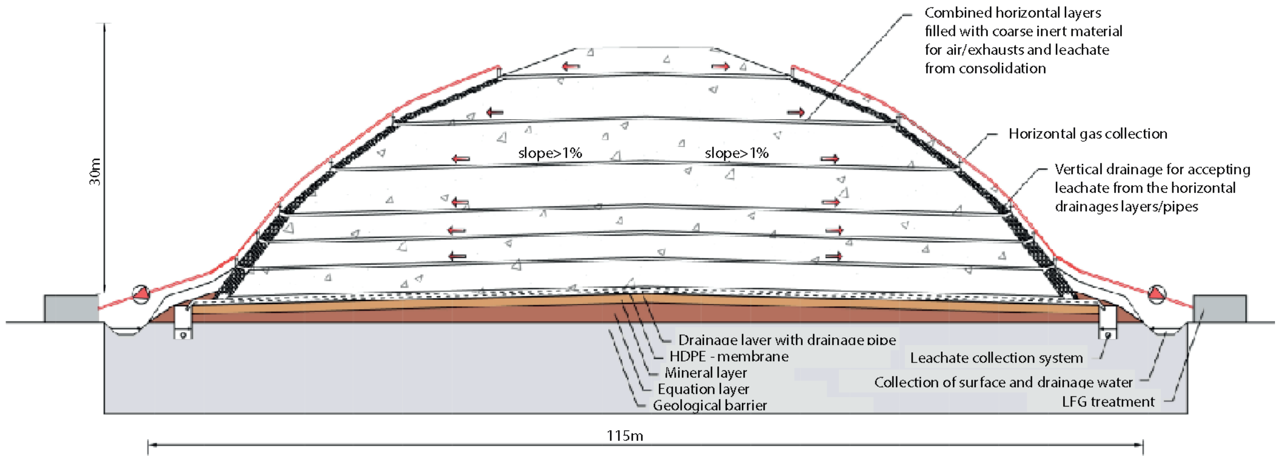

FIGURE 6: Modified landfill concept with multi-purpose horizontal layers of coarse material (Stegmann, 2017).

installed in a MBT waste landfill in Lower Saxony, Germany.

By developing an appropriate design for the base drainage system, water should be able to freely discharge from the bottom drain gravel layers of the landfill even in the presence of clogged or damaged drainage collection pipes. Frequent drainpipe inspection and cleaning should be ensured.

5. CONCLUSIONS

Landfill should be designed and constructed in line with the principle of environmental sustainability, in order to achieve over a period of less than one generation, a landfill with a low emission potential. Based on the multibarrier concept, technical measures aimed at controlling emissions and risk of contamination, in both the short and the long term, may include: minimization of contaminant mass introduced into the landfill, appropriate waste pre-treatment and in-situ treatments to minimize the emission potential of wastes, appropriate biogas and leachate collection systems, flexible use of the top cover, lining and appropriate landfill siting and morphology. However, landfill is typically deemed an obsolete and potentially redundant system and from a political, legislative and technical viewpoint it is frequently denied the attention devoted to other engineering works, lacking adequate financial investment to cover the costs required to ensure a sustainable landfill system. The lack of an appropriate landfill design project corresponding to the principles of sustainability based on use of the above-mentioned technical strategies, has resulted in a series of critical aspects in traditional non-sustainable landfills, which required specific technical solutions with a focus on ensuring landfill sustainability:

- Appropriate design of in-situ aeration, moistening and pre-treatment of waste, having consideration for the specific quality of the residues from the Circular Economy, which might accumulate contaminants during the recycling process.
- Use of proper granular drainage material to extend its long-term functionality and use of horizontal drainage layers at different levels of the landfill to avoid the building up of high water table and assure the freely discharge of water even in the presence of clogged or damaged drainage collection pipes.
- Use of morphologies envisaging deposition only above ground level.
- Exploitation of the flexible role carried out by a final cover, based on both the type of landfill and quality of landfilled waste.
- Use of bio-oxidising material in the top cover of the landfill to oxidize residual biogas emissions.
- Consider the limited duration of the physical barriers already during the design stage in order to take appropriate measures to ensure that emissions polluting the environment do not last longer than the lifetime of the physical barriers.
- Appropriate selection of the siting and morphology of the landfill, considering the attenuation capacity of the unsaturated ground and water tables.
- Consider landfills for appropriate after-use

A modified concept for landfilling of raw and pre-treated waste is presented in order to avoid a lot of problems that landfills with increasing age will face.

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INFLUENCE OF MODELLING CHOICES ON THE RESULTS OF LANDFILL ODOUR DISPERSION

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ABSTRACT

Landfills are an important source of odour pollution, potentially causing nuisance to adjacent populations. The most commonly used odour impact assessment for this type of plants usually involves a combination of dynamic olfactometry with dispersion modelling. Despite the advantages associated with the use of dispersion models, there are still some important issues related to their uncertainty. The dispersion model requires the Odour Emission Rate (OER) as input, expressed as units of odour emitted per unit time. Source term characterization and the estimation of the OER are typically the most important steps in the model's implementation, accounting for the highest contribution to the overall uncertainty. Another important element of uncertainty when modelling emissions from landfill surfaces is the geometrical implementation of the emission source in the dispersion model. This entails the definition of the initial dimensions of the emission, which is critical in the case of large area sources. This paper discusses issues related to uncertainty in the use of dispersion models for the evaluation of landfill odour impacts, particularly focusing on the estimation of the OER and the emission's initial vertical dimension. This study shows that modelling choices may lead to a variance in the resulting modelled odour concentrations at receptors differing by up to a factor 3. This variability should not cause distrust in the method, but rather indicates the importance of having odour dispersion modelling studies carried out by experts with deep knowledge of the physical-chemical mechanisms underlying atmospheric emissions.

1. INTRODUCTION

In recent decades, public awareness of air quality issues has increased considerably. This led to the inclusion of odours, which can negatively affect human well-being without necessarily having an adverse effect on health (Sucker et al., 2001; Zhao et al., 2015), among the atmospheric pollutants subject to control and regulation in many countries (Brancher et al., 2017). Indeed, due to the fact that residential areas are often very close to industrial activities, odours are currently a major cause of complaints to local authorities (Henshaw et al., 2006; Marchand et al., 2013). In many cases odour complaints become the limiting factor for the operation of existing plants or for the realization of new ones. This is particularly true for plants involved in the treatment and disposal of waste, which are a common source of odour emissions and of consequent concerns for adjacent populations (Ying et al., 2012; Marchand et al., 2013; Sironi et al., 2006). Landfills can be particularly problematic in relation to odour nuisance (Che et al., 2013; Sakawi et al., 2017), and thus require specific protocols for odour control and measurement (Chemel et al., 2012; Lucernoni et al., 2016; Sarkar et al., 2003; Tansel et al., 2019).

As already mentioned, odours are currently subject to control and regulation in many countries. Dispersion models are the preferred approach to odour impact assessment in most of the regulations concerning odour around the world (Brancher et al., 2017). This is due to their ability to simulate odour dispersion from the emission sources into the atmosphere, and to calculate ambient odour concentration values in the simulation space-time domain. In some cases, odour regulations specify a minimum distance between the closest inhabited area and the location of possible odour-producing industrial or agricultural facilities (Brancher et al., 2019; Capelli et al., 2013). Minimum distances are typically calculated by applying dispersion models or using simplified mathematical formulas with specific coefficients derived from dispersion modelling (Schauberger et al., 2012a, 2012b). In other cases, regulations set acceptability standards in terms of the frequency with which a given odour concentration may be exceeded (Brancher et al., 2017; Capelli et al., 2013).

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Despite the great advantages of using dispersion models for odour impact assessment, there still are some critical aspects which require further study. One of the major open issues related to dispersion modelling is validation (Hayes 2006). This is particularly complex due to the difficulty of measuring odours in the environment (Capelli et al., 2013, 2018). Another important aspect is the uncertainty associated with dispersion models when applied to odour impact assessment studies (Brancher et al., 2019; Oettl et al., 2018).

The choice of a particular dispersion model will affect the resulting calculated impacts (Piringer et al., 2016), but also within each model there are different choices and parameters that may produce significant discrepancies in the model outputs. In general, dispersion models require three types of input data: emission data, topographic, and meteorological data of the site. The model combines these to produce an estimate of how pollutant emissions are diluted and diffused into the atmosphere. Each of the input datasets represent a possible source of uncertainty, as well as any other model-specific settings. In the case of odour dispersion modelling, the emission data input for the model is represented by the Odour Emission Rate (OER), expressed as units of odour emitted per unit time (Capelli et al., 2013). For characterizing the emissions source, the OER shall be put in relation with the geometrical and physical parameters of the source, which are also required by the dispersion model. For some types of sources, source term characterization and the estimation of the OER can be extremely complex. For instance, such cases include sources that have variable emissions over time, where it can be difficult to associate a specific OER to every hour of the simulation domain, or diffuse sources, for which the emitted air flow is diifult to estimate. Source term characterization is therefore typically considered to be the most critical step, and thus the major contributor to the overall uncertainty, in the implementation of an odour dispersion model (Capelli et al., 2014).

Characterization and implementation of a dispersion model is particularly problematic for landfill surfaces as a source. The determination of odour emissions from landfills is a complex and still hotly debated task (Lucernoni et al., 2017). Even under the assumption that the odour emission is associated mainly with the emission of landfill gas (LFG) escaping the collection system (Chemel et al., 2012; Saral et al., 2009), there are currently no universally accepted methodologies for evaluating the OER associated with this type of emission source (Lucernoni et al., 2017; Capelli and Sironi, 2018).

Another important element of uncertainty when modelling emissions from landfill surfaces is the geometrical implementation of the emission source in the model. In particular, the definition of the initial dimensions of the emission is critical in cases of large area sources.

This paper will discuss uncertainties in the application of dispersion modelling by referring to a specific case study regarding the assessment of the odour impact from a landfill with a surface of 55'000 m2. For the study, it was decided to apply the CALPUFF dispersion model, which is a commonly used model for regulatory purposes in Italy (Capelli et al., 2018; Ranzato et al., 2012).

More in detail, this paper will focus on two main sources of uncertainty. First, it will compare the different OER values obtained by using two different sampling methods, both of which are applicable for odour sampling on landfill surfaces (Capelli et al., 2018; Lucernoni et al., 2017). Second, it will investigate the effects of the so-called "initial vertical sigma" σZ,0, a parameter specifically required by the CALPUFF model when modelling area sources, related to the vertical dimension of the emission.

The present work, besides summarising some basic principles for the selection of the sampling method and the definition of the source geometry, will demonstrate the extent to which different choices regarding the model setting may affect its results, i.e. the simulated odour impact of the studied landfill.

2. MATERIALS AND METHODS

2.1 Case study description

The selected case study regards the odour impact assessment of a landfill with an emitting surface of 55'000 m2. Since the principal aim of the study is to consider the influence of different choices in some critical parameters on the outputs, it was decided to limit the evaluations to the emissions from the closed landfill surface. This avoids introducing other variables and thus other possible sources of uncertainty. In this specific case, due to the large surface of the closed portion (55'000 m2) compared to the typical daily tipping area (about 1'000 m2), the closed portion of the landfill can reasonably be assumed to be the main source of odour emissions (Lucernoni et al., 2016).

The dispersion of the odour emissions from the landfill surface was evaluated using the CALPUFF model (Scire et al., 2000). CALPUFF is a multilayer, multispecies, non-steady-state, puff dispersion model. It is currently the most commonly used dispersion model in Italy for odour impact assessment evaluations and regulatory purposes (Capelli et al., 2018; Ranzato et al., 2012).

The meteorological data used for the study is one year of 3D hourly data for 2015, processed using the WRF (Weather Research and Forecasting) model with a 1 km resolution relevant to the studied area. The meteorological domain and the simulation domain were set as the same, comprising an area of 4000 m x 4000 m, with a resolution of 100 m, giving a total of 1600 horizontal cells. 10 cells were considered on the vertical plane, giving a total of 16000 cells for the study.

The emission data was derived from an olfactometric sampling carried out on the landfill surface, as described in the next paragraph.

2.2 Odour sampling methods

Up to now, no universally accepted methodology for sampling and assessing odour emissions from landfill surfaces has been established (Lucernoni et al., 2017; Capelli et al., 2018).

One common approach to assess both odour and landfill gas emissions from landfill surfaces involves the use of sampling hoods, such as static hoods, flux chambers, or wind tunnels (Di Trapani et al., 2013; Lucernoni et al., 2016, 2017; Rachor et al., 2013; Schroth et al., 2012). Wind tunnels are the "official" method foreseen by local Italian guidelines on odour impact assessment when conducting odour sampling on passive area sources (Capelli et al., 2018). The main difference between wind tunnels (WTs) and flux chambers (FCs) is the air flow rate at which the two systems are operated:

- In WTs the air flow is directional, whereas in FCs the inlet flux is mixed inside the hood;
- The typical air flow for WTs is about one order of magnitude higher than the air flow used in FCs.

Previous recent studies on the matter have proven that FCs provide a better representation of odour emissions from landfill surfaces (Lucernoni et al., 2017).

Since the aim of this study is to evaluate the effect of the most significant sources of uncertainty when assessing odour emissions from landfills, the choice of the sampling method was included in the aspects to be evaluated. Both a WT and a FC were used in this study, and the specific odour emission rate (SOER) values evaluated with the two methods were compared.

The WT used for this study has a base area of 0.125 m^2, and is operated with an air flow of 40 L/min. The FC has the same base area as the WT (i.e. 0.125 m^2) and is operated with an air flow of 4 L/min.

For both sampling methods, the SOER can be evaluated as follows:

$$SOER = (c_{od} Q_{air})/A_{base} \qquad (1)$$

Where SOER is the Specific Odour Emission Rate ($ou_E/m^2/s$), cod the measured odor concentration (ou_E/m^3), Q_{air} the airflow rate inside the hood (m^3/s) and A_{base} the base area of the sampling hood (m^2). The SOER value is the parameter that is used as input for implementing area sources in dispersion models. Odour concentration was measured according to the EN 13725:2003.

For the collection of the odour samples, the landfill surface was divided into 9 sub-areas (Figure 1, left). One sample was collected at the centre of each sub-area.

2.3 Definition of the initial vertical dispersion coefficient $\sigma_{z,0}$

The CALPUFF model requires the definition of some specific dimensional parameters when characterizing an area source. Besides the source area and height, it is also necessary to define the so-called "initial vertical sigma", $\sigma_{z,0}$, which is a measure of the initial vertical dimension of the emission.

Considerations on how to set this parameter can be found for Gaussian models (US EPA, 2004, 2011). The most common rule for the evaluation of $\sigma_{z,0}$ is to set it equal to the vertical dimension of the source (i.e., the source height) divided by 2.15, as suggested in Table 3-2 of the US EPA User's guide for the regulatory model AERMOD (US EPA, 2011). The 2.15 coefficient is derived from the Gaussian distribution of the pollutant concentration inside the plume. However, despite this simple rule, considerations for the correct setting of $\sigma_{z,0}$ are not so simple.

As stated earlier, $\sigma_{z,0}$ represents the initial vertical dimension of the plume (for a Gaussian model; in CALPUFF, $\sigma_{z,0}$ represents the initial vertical dimension of the puff at the emission, but the considerations are similar). Therefore, as a general rule, $\sigma_{z,0}$ is directly related to the source height. However, the source height is not the only parameter that may affect the initial vertical dimension of the plume. Especially for certain particular sources, as is also mentioned in the AERMOD User's Guide (US EPA, 2011). The guide specifies that in cases where the emission may be turbulently mixed near the source and therefore occupy some initial depth, the $\sigma_{z,0}$ should be set so as to account for this initial vertical dimension of the emission. Unfortunately, it is not specified how to do that. Different methods

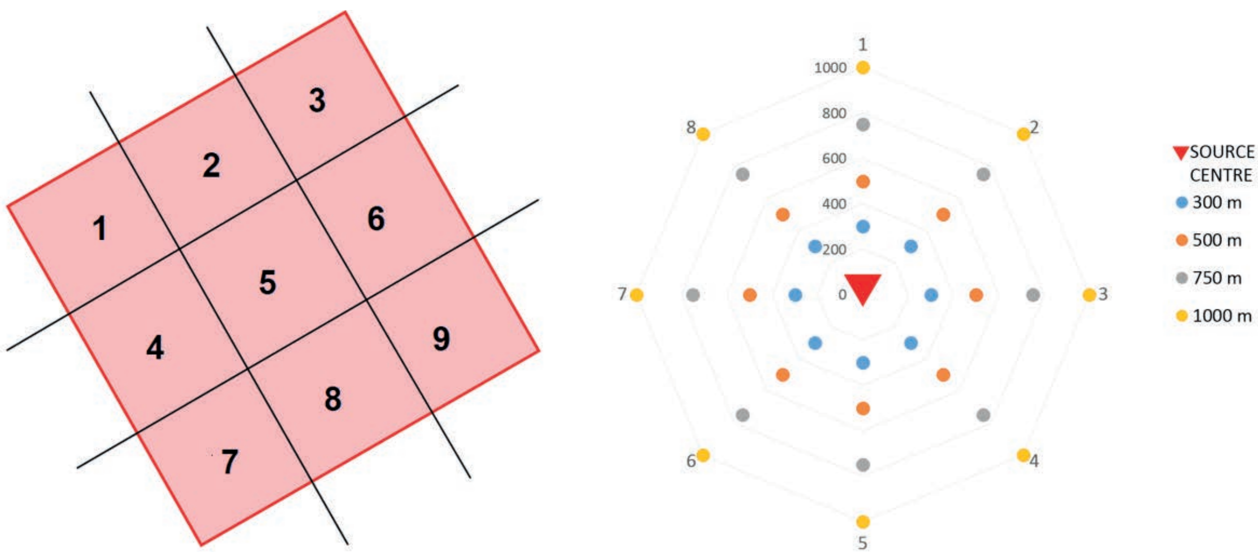

FIGURE 1: Division of the landfill into 9 sub-areas for olfactometric sampling (left) and location of the 32 discrete receptors selected for comparison of the model outputs obtained with different values of $\sigma_{z,0}$ (right).

for the calculation of $\sigma_{Z,0}$ can be found in the US EPA's Haul Road Workgroup final report to the Air Quality Modeling Group, though these are specifically related to the emissions from road transport. However, this document at least clearly shows that $\sigma_{Z,0}$ is not necessarily solely a function of the source height.

When it comes to area sources, $\sigma_{Z,0}$ is the initial vertical dimension of the area source plume. Passive area sources, such as wastewater treatment tanks, are typically relatively small (compared to a landfill) and the emission occurs passively due to natural convection from the liquid surface to the atmosphere. In these cases, there is no reason to assume that turbulence plays an important role and cause the plume to have an increased initial dimension due to turbulent effects.

However, the case of landfills is different. First, landfills cannot be treated similarly to passive area sources because of the different mechanisms that regulate the emission from the surface to the atmosphere. There is a small – but not negligible – flux of gas that crosses the landfill body and is emitted into the atmosphere through the landfill surface. For this reason, landfills should be considered as "semi-passive" area sources. Second, landfills are typically very large area sources (55'000 m^2 in the case study considered), characterized by an uneven surface - even considering the same parcel, the landfill surface can have different heights due to the presence of reliefs. Moreover, the landfill surface is typically scattered with "obstacles", such as LFG extraction wells. All these elements contribute to the presence of some degree of turbulence over the landfill surface. This means that the initial plume emitted by the landfill will have a height related to this turbulence. Finally, due to the large dimensions of the landfill, it can be reasonably supposed that the initial plume height (PH) will also be related to the horizontal dimensions of the landfill. Higher plumes can be assumed to be emitted from very large landfills, where the turbulent contribution is higher, and the pollutants are carried along the landfill surface for its entire length to form the initial emission puff.

Because of the lack of precise rules for the evaluation of $\sigma_{Z,0}$ in such complex cases, and because of the complexity of the geometrical features of the landfill, it is not possible at this stage to establish one unique and unequivocal way for setting the value of $\sigma_{Z,0}$ in the model.

For these reasons, it was deemed useful to evaluate how the choice of different values of $\sigma_{Z,0}$, selected within a reasonable range, could affect the model results.

For this study the model was run using the following values of $\sigma_{Z,0}$: 1, 2, 5, 10, 20, and 30 m. Besides comparing the different odour impact maps resulting from the model runs, further evaluations were made by comparing the odour concentrations calculated by the model on a set of selected receptors. A receptor nest was created by placing 8 receptors at distances of 300, 500, 750 and 1000 m from the source centre, respectively, giving a total of 32 receptors, according to the scheme shown in Figure 1 (right).

3. RESULTS AND DISCUSSION

3.1 Odour sampling on the landfill surface

As already mentioned, olfactometric sampling was performed both with a Flux Chamber (FC) and a Wind Tunnel (WT). In order to compare the results obtained with the two methods, Table 1 reports the results of the olfactometric measurements carried out on the landfill site in terms of odour concentration (in ou_E/m^3) and SOER, which was evaluated as described in section 2.2.

The lower odour concentration values were found in the samples collected using the wind tunnel. The higher operational airflow causes a higher dilution of the sample collected at the hood outlet. However, for the SOER, the opposite trend is observed: the SOER derived from the FC measurements is about three times lower than the SOER resulting from the wind tunnel measurements. This can be explained by the fact that the SOER is obtained by the product of the odour concentration and the airflow.

This in turn means that, depending on the sampling method adopted, the resulting emission data to be used as input for dispersion modelling – and consequently the resulting ambient concentrations – may vary by a factor of 3. This clearly represents a significant source of uncertainty: the breach of acceptability criteria, or the determination of suitable separation distances, may be totally overturned by such an uncertainty factor.

As previously mentioned, there are recent studies re-

TABLE 1: Results of the olfactometric measurements of the odour samples collected over the landfill surface by means of a flux chamber (FC) and of a wind tunnel (WT).

Sampling point	FC	FC	WT	WT
	Measured c_{od} [ou_E/m^3]	SOER [$ou_E/m^2/s$]	Measured c_{od} [ou_E/m^3]	SOER [$ou_E/m^2/s$]
1	750	0.40	140	0.75
2	410	0.22	81	0.43
3	270	0.14	60	0.32
4	66	0.04	52	0.28
5	1330	0.71	240	1.28
6	840	0.45	310	1.65
7	310	0.17	88	0.47
8	860	0.46	280	1.49
9	380	0.20	210	1.12
Geometric average	440	0.23	134	0.71

garding the estimation of odour emissions from landfill surfaces proving that WTs tend to overestimate emissions (Lucernoni et al., 2016, 2017). This overestimation is assumed to be due to the fact that odour concentration values measured using WTs are in some cases so low that they are likely to be attributable more to the odour of the landfill soil coverage than to the emission of LFG through the landfill surface (Lucernoni et al., 2016). Indeed, among the odour concentrations of the samples collected using the WT in this case, some values are so low that they are close to the lower detection limit for dynamic olfactometry (Lucernoni et al., 2016). This is less relevant for FC measurements, where the odour concentrations are considerably higher, and thus should prevent the interference of possible background odours.

3.2 Evaluation of the effect of different values of $\sigma_{z,0}$

As described in section 2.3, the model was run by setting different values for $\sigma_{z,0}$ in order to evaluate the influence of this parameter on the model outputs, i.e. on the simulated odour impact of the landfill on the surrounding territory.

The odour impact resulting from dispersion modelling was evaluated in terms of 98th percentile of the hourly peak odour concentration values simulated by the model on the receptor grid, in conformity with the prescriptions of the local regulations about odour pollution that are currently in force in Italy.

Figure 2 shows the maps of the 98th percentile hourly peak odour concentration values simulated by the model runs, obtained by setting different values for $\sigma_{z,0}$, i.e. 1, 5, 10 and 30 m, respectively. In order to allow for comparison, the same scale was used for the different maps.

It can be observed that the extent of the odour impact shrinks when $\sigma_{z,0}$ increases: a higher initial dimension of the emission causes a better dispersion of the odour, and thus a lower resulting impact. This behaviour is particularly evident for the highest value of $\sigma_{z,0}$ tested (i.e. 30 m), whereas such differences are less pronounced in the maps resulting from the simulations with $\sigma_{z,0}$ values of 1, 5 and 10 m. Indeed, the highest differences are observed close to the source, where the maximum odour concentrations modelled decrease significantly, whereas the shape and the extent of the iso-concentration lines at higher distance from the source look more similar between the different maps.

In order to better visualize the variations in the results relevant to the different conditions tested, the odour concentration values calculated punctually by the model on a set of different receptors, located at different distances from the source, were evaluated. The 98th hourly peak odour concentration values resulting for the different va-

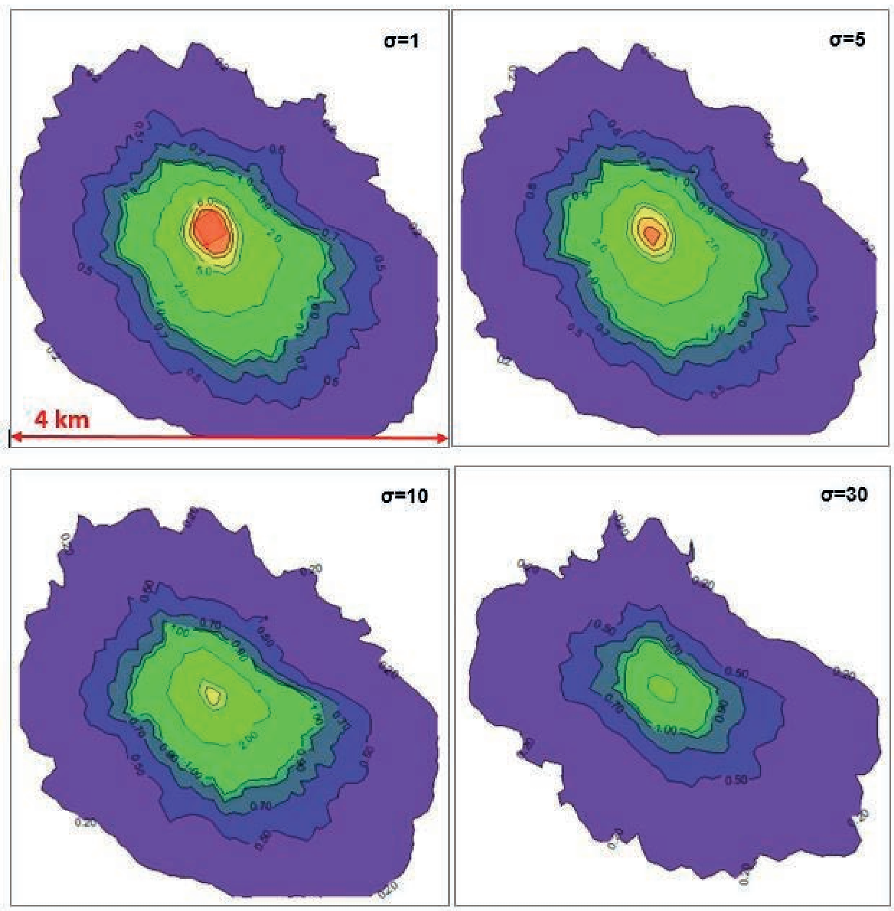

FIGURE 2: Maps of the 98th percentile of the hourly peak concentrations resulting from the model runs performed by setting the source $\sigma_{z,0}$ equal to 1, 5, 10 and 30 m, respectively.

lues of $\sigma_{Z,0}$ on the 32 receptors, selected as illustrated in section 2.3, are reported in Table 2.

The values reported in Table 2 reflect what is observed from the maps. In particular, odour concentrations higher than 5 ou_E/m^3, which is often taken as the reference value for acceptability criteria in Italian guidelines, are highlighted in red. When considering the receptors at 300 m from the source centre, by going from a $\sigma_{Z,0}$ of 1 m to a $\sigma_{Z,0}$ of 30 m, the resulting concentration on the receptor decreases by a factor 3-4. This effect is less pronounced at higher distances from the source, where the decrease is about the half. Nonetheless, the variation factors in the model results obtained with different values of $\sigma_{Z,0}$ are quite high. Assuming that an acceptability criterion of 5 ou_E/m^3 at the closest receptors is fixed for the studied area, then it is clearly visible that the choice of the $\sigma_{Z,0}$ value could cause the result to either stay below or exceed this limit. This consideration is particularly important, since, as stated in the introduction, dispersion models are often used for regulatory purposes.

3.3 Discussion on the evaluation of the $\sigma_{Z,0}$ value

Given that the setting of the $\sigma_{Z,0}$ for landfill sources represents an important degree of freedom in the model implementation, it is very important to analyse the variability of outputs derived from different possible – and reasonable – choices for this value. In this regard, it is important to highlight that, to the best of our knowledge, there is a lack of specific studies addressing this problem and discussing how to choose this parameter properly in order to obtain representative results.

TABLE 2: 98th hourly peak odour concentration values calculated by the model for the different values of $\sigma_{Z,0}$ tested on the 32 discerete receptors selected at 300, 500, 750, and 1000 m from the source centre, respectively.

Receptor	σ=1	σ=2	σ=5	σ=10	σ=20	σ=30
R1_300m	4.67	4.38	3.79	2.79	1.52	1.27
R2_300m	4.61	4.09	2.85	1.98	1.13	0.95
R3_300m	5.25	5.02	4.27	2.80	1.83	1.52
R4_300m	6.68	6.38	5.36	3.80	2.19	1.70
R5_300m	5.99	5.65	4.74	3.22	1.77	1.25
R6_300m	4.28	4.04	3.37	2.43	1.32	0.87
R7_300m	4.44	4.19	3.53	2.60	1.59	0.96
R8_300m	3.73	3.58	3.14	2.45	1.45	1.03
R1_500m	1.78	1.75	1.63	1.29	0.95	0.82
R2_500m	1.24	1.17	1.11	0.81	0.63	0.58
R3_500m	2.69	2.63	2.35	2.01	1.32	0.98
R4_500m	4.07	3.81	3.36	2.29	1.48	1.07
R5_500m	3.35	3.18	2.76	1.92	1.03	0.76
R6_500m	1.99	1.94	1.68	1.29	0.77	0.54
R7_500m	1.98	1.94	1.79	1.48	1.11	0.66
R8_500m	1.63	1.61	1.53	1.29	0.86	0.83
R1_750m	0.69	0.75	0.66	0.62	0.50	0.44
R2_750m	0.75	0.74	0.71	0.58	0.47	0.38
R3_750m	1.40	1.39	1.28	1.16	0.78	0.71
R4_750m	2.10	2.07	1.95	1.47	1.03	0.82
R5_750m	1.89	1.86	1.57	1.09	0.74	0.50
R6_750m	1.06	1.04	0.96	0.75	0.50	0.41
R7_750m	1.14	1.13	1.05	0.95	0.64	0.44
R8_750m	0.96	0.95	0.91	0.77	0.58	0.55
R1_1km	0.49	0.56	0.47	0.40	0.37	0.33
R2_1km	0.50	0.46	0.45	0.35	0.23	0.20
R3_1km	1.05	1.04	0.98	0.89	0.71	0.62
R4_1km	1.37	1.36	1.29	0.99	0.79	0.60
R5_1km	1.27	1.25	1.12	0.90	0.55	0.40
R6_1km	1.73	1.68	1.44	1.01	0.68	0.52
R7_1km	0.73	0.72	0.65	0.62	0.52	0.36
R8_1km	0.61	0.61	0.60	0.54	0.46	0.43

To be able to recommend a certain value, validation in the field would be necessary in order to evaluate the model capability to predict experimental observations. For this purpose, field inspections for the determination of the odour plume extensions represent an interesting opportunity (Capelli et al., 2013; Capelli and Sironi, 2018). Another way of verifying the predictions of dispersion models would be to carry out field monitoring with electronic noses (Capelli et al., 2013). However, model validation was not foreseen in this study because of the high costs involved. Indeed, in the case of odour, the development of an ad hoc trial to validate model predictions is a complicated task (Capelli et al., 2013).

In general, it would be extremely useful to integrate validation efforts with a more theoretical approach, based on the study of atmospheric turbulence over large area sources. For this purpose, further studies will be conducted in the future dealing specifically with the influence that turbulence has on emissions, thus providing more precise indications on ways to select appropriate values for $\sigma_{z,0}$.

Another way to evaluate how to establish an appropriate value for $\sigma_{z,0}$ might be to compare with results from other dispersion models which have a lower degree of uncertainty and do not require the definition of $\sigma_{z,0}$. In this case, the optimal $\sigma_{z,0}$ would be the one that provides results in closer agreement with the other models. However, by definition, air quality models can only approximate atmospheric processes and many assumptions are required to describe real phenomena in mathematical equations (Moussiopoulos et al., 1996). Therefore, agreement with other dispersion models does not necessarily ensure representative results.

It is worth highlighting that the initial vertical dispersion coefficient is a parameter specifically required by the CALPUFF model in cases of area sources. If the landfill is implemented as a point source, the model automatically sets a default value for the $\sigma_{z,0}$ equal to the height of the emission source divided by a factor of 2.15. In that case, the choice of $\sigma_{z,0}$ wouldn't be an issue. Conversely, when dealing with point sources, a proper value for the vertical rise velocity, which is not required in cases of area sources, has to be defined. In the case of a landfill, the estimation of the rise velocity could be done based on the evaluation of the landfill gas flux from the landfill surface. However, due to the large dimensions that are typical of landfills, their approximation as point sources could also be inappropriate. .

CONCLUSIONS

Dispersion models are currently the most common method for assessing odour impacts for regulatory purposes. For this reason, it is important to analyse the differences in the results, which can be produced by making different modelling choices.

The choice of method to obtain the required olfactometric data to characterize the emissions from the landfill surface is particularly critical. Wind tunnels are the sampling method foreseen by Italian regulations on the matter, however, previous studies have proven such systems to overestimate emissions. In this study, we found that wind tunnels produce odour emission rate values that are three times higher than those obtained with flux chambers. This is reflected in the model outputs resulting in concentrations at receptors that are three times higher.

Another critical parameter for area source characterization in the CALPUFF model is the so called "initial vertical sigma" $\sigma_{z,0}$. Given that the definition of the $\sigma_{z,0}$ for landfill surfaces represents an important degree of freedom when implementing the source term in CALPUFF, it is important to be aware of the variability of results that derives from possible – and reasonable – choices for this value. The sensitivity analysis conducted relevant to the proposed case study, involving a landfill with a surface of 55'000 m^2, shows that, for $\sigma_{z,0}$ values ranging from 1 m to 30 m, modelled concentrations at receptors may vary by almost a factor 4 at 300 m from the source and by a factor 2 at a distance of 1 km.

This high variability should not cause distrust in the method, but should emphasize the importance of properly implementing the model. To do that, it is necessary to analyse the physical and chemical mechanisms related to the emission of odours from the studied sources, in order to choose the most appropriate sampling strategy and define the initial dimensions of the emitted plume or puff within a narrow range. Especially in the case of verification of compliance to acceptability criteria, it is important that odour dispersion modelling studies are carried out by experts with appropriate knowledge and understanding of the odour emissions under investigation.

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THE ROLE OF NATURAL CLAYS IN THE SUSTAINABILITY OF LANDFILL LINERS

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ABSTRACT
Engineered synthetic liners on their own cannot protect the environment and human health against landfill leachate pollution. Despite their initial impermeability, they are susceptible to failure during and after installation and have no attenuation properties. Conversely, natural clay liners can attenuate leachate pollutants by sorption, redox transformations, biodegradation, precipitation, and filtration, decreasing the pollutant flux. Depending on the clay, significant differences exist in their shrinkage potential, sorption capacity, erosion resistance and permeability to fluids, which affects the suitability and performance of the potential clay liner. Here, the physico-chemical, mineralogical and geotechnical characteristics of four natural clayey substrata were compared to discuss their feasibility as landfill liners. To study their chemical compatibility with leachate and rainwater, hydraulic conductivities were measured every ≈2 days spread over 7 weeks of centrifugation at 25 gravities. At field-scale, this is equivalent to every 3.4 yrs spread over 80 yrs. All the clayey substrata had favourable properties for the attenuation of leachate pollutants, although different management options should be applied for each one. London Clay (smectite-rich) is the best material based on the sorption capacity, hydraulic conductivity and low erodibility, but has the greatest susceptibility to excessive shrinkage and alterable clay minerals that partially collapse to illitic structures. Oxford Clay (illite rich) is the best material for buffering acid leachates and supporting degradation of organic compounds. The Coal Measures Clays (kaoline-rich) have the lowest sorption capacity, but also the lowest plasticity and have the most resistant clay minerals to alteration by leachate exposure.

1. INTRODUCTION

Leachates produced in municipal landfills constitute a health and environmental problem due to the different pollutants they content. For this reason, liners are required to minimise offsite migration of leachate. Two types of liners are currently used in modern landfills: synthetic liners, typically made of HDPE, and natural liners, typically made of compacted clay (Adar and Bilgili, 2015; Wei et al., 2018). Synthetic liners offer long-term impermeability to leachate but imply a high technology input and can be affected by slope stability, interface shear strength (Kavazanjian et al., 2006) and physico-chemical, thermal and mechanical problems (Kong et al., 2017), which may result in failure within 10 yrs of service (Rowe and Sangam, 2002; Rowe et al., 2003). Compacted clay liners are puncture-resistant and have advantageous reactive properties, but can be unstable in contact with leachate and susceptible to cracking under repeated wetting and drying cycles (Louati et al., 2018; Yesiller et al., 2000).

Because containment liners eventually fail independently of their low permeability properties, landfills are potential "ticking time bombs" that store and isolate waste until the confined pollutants are accidentally released to the environment in leachate. However, the intrinsic reactive properties of clays means they can biogeochemically interact with pollutants in leachates to decrease their availability and potential hazards over time. This reactivity (or attenuation) is enhanced if rainwater is allowed to enter the waste because then waste degradation is boosted, which accelerates its stabilization (Allen, 2001). The attenuation capacity of natural liners results in shorter periods of (1) potential release of pollutants and (2) aftercare monitoring, with subsequently lower landfill cost and less risk of environmental contamination. However, most attention has traditionally

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focused on the impermeability properties of liners as a performance characteristic. Here we focus on both impermeability and attenuation capacities, without relying solely on the impermeability (or containment) role, as a basis to improve liner design and performance. This dual capability can in principle be included in the engineering design of compacted clay liners to manage both organic and inorganic pollutants in leachate (Thornton et al., 1993; 1997).

The attenuation capacity, low cost and ease of implementation of compacted clay liners makes them more attractive than synthetic geomembranes on their own in landfill liner systems. This is particularly important in low-income countries where >90% of waste is openly dumped (Kaza et al., 2018), and in removing organic pollutants (Beaven et al., 2009). However, clays are very diverse in their physico-chemical properties and the suitability of the clayey substratum as a potential liner must be properly evaluated (Widomski et al., 2018). This task is complex and, in order to avoid any adverse effects, must consider the factors and the interactions between them, which affect leachate-liner system. There are many factors involved, such as liner mineralogy, shrink/swell potential, sorption capacity, dispersive/erosion behaviour and fluid permeability. If the clay plasticity is too high, construction of the liner becomes more difficult and the swelling/shrinking/cracking potential more significant as a failure mechanism.

In this study, the feasibility of four natural clayey substrata as landfill liners was evaluated. Their physico-chemical, mineralogical and geotechnical characteristics were studied and the results were discussed in terms of strengths and weaknesses as candidate materials for landfill liners. Finally, the potential for attenuation of pollutants in leachate by each material was evaluated for sustainable landfill applications. The aim was to characterise the relevant properties of the different clays in order to identify those which are geotechnical stable and effective in pollutant attenuation for liner design. This is essential to prevent pollution of the environment and protect human health from leachate spreading over groundwater aquifers or adjacent rivers and lands.

2. MATERIALS AND METHODS

2.1 Materials

Four clayey substrata from the United Kingdom were studied: London Clay (LC), Oxford Clay (OC), and shallow and deep Coal Measures Clays (SCMC and DCMC) (Table 1). The LC originated from shale, greensand, chalk, and lateritic soils during a sea level rise over the Northern Sea Basin. It is a silty to very silty clay, slightly calcareous with disseminated pyrite. The OC, collected from the Peterborough Member, contained many visible fossils (vertebrate and invertebrate), particularly bivalves (Meleagrinella). In contrast to the two previous clays, the Coal Measures Clays originated in a fluvio-deltaic environment and also have a relatively high proportion of iron sulphides (pyrite, marcasite) and gypsum, the latter following pyrite weathering. These Coal Measures Clays consist of interbedded clay, shales, silt and sand, interstratified with coal.

Approximately 100, 73, 25 and 20 kg of respectively LC, OC, SCMC and DCMC were recovered in-situ between June and July 2018. The pore water and cation exchange complex compositions were analysed in several subsamples before oven drying. The exchangeable cations were analysed in air-dried and powder samples (prepared with agate mortar and pestle) after applying 3 cycles of 10 sec of ultrasonification to a suspension of 0.15 mg clay in 50 mL deionized water. The elemental composition and mineralogy were determined after oven drying and grinding to a fine powder. For geotechnical tests the conglomerates were oven dried to remove residual moisture and the dry lumps broken up until a particle size <0.2 cm was achieved. For this, a rammer and several perforated screen trays fitted in a CONTROLS sieve shaker (Model 15 d040/a1) were used and the clays first reduced into approximately 2 cm aggregate lumps. Next, the <0.2 cm particles were recovered separately and the 0.2-2 cm lumps put into a bench soil grinder (Humboldt Co) and broken up to achieve <0.2 cm size. All results are expressed as a function of dry mass.

2.2 Analyses

The concentrations of nitrogen, carbon, hydrogen and sulphur were analysed in duplicate samples ground to ≤0.1 cm (0.005 g) using a Thermo Scientific FLASH 2000 Elemental Analyzer (CHNS). The remaining elements except oxygen and the halogens were analysed using a Spectro-Ciros-Vision radial ICP-OES instrument after acid digestion of 0.031 and 0.094 g at 150°C. For this, 12 mL of aqua regia was applied for 30 min followed by two 1 mL volumes of HF for 10 min, and the resulting solution was eluted up to 50 mL with 1 mass % HNO_3. The mineralogy was determined by X-ray diffraction (XRD) using a Ni-filtered Cu Kα radiation (k = 0.15406 nm) in a Philips X'Pert diffractometer,

TABLE 1: Information about the natural clayey substrata samples.

	London Clay (LC)	Oxford Clay from Peterborough Member (OC)	Shallow Coal Measures Clay (SCMC)	Deep Coal Measures Clay (DCMC)
Location (UK)	North Essex	Northwest Buckinghamshire	West Yorkshire, collected close to the surface	West Yorkshire, collected at a greater depth
Age	Eocene: 47.8 - 56.0 Ma	At the end of the Middle Jurassic: 164-166 Ma	Upper Carboniferous: 310 Ma	Upper Carboniferous: 310 Ma
Origin	Deep marine sediments	Deep marine sediments	Fluvio-deltaic sediments	Fluvio-deltaic sediments
Colour / appearance	Uniform, firm, brown colour	Grey colour with carbonaceous shells and rootlets	Dark grey-dark brown colour	Two-coloured: orange-light brown and dark brown
Selected references	Fannin 2006; Kemp and Wagner, 2006	Fannin 2006; Hudson and Martill, 1994; Scotney et al., 2012	Freeman, 1964, McEvoy et al., 2016	Freeman, 1964, McEvoy et al., 2016

operating at 40 kV and 40 mA, with a step size of 0.016 and a speed of 2 s/step. The samples were prepared by both sprayed random powder (after grinding down to 1-3 μm) and flat oriented slides (after obtaining the <2-μm fraction by dispersant and Stokes' law). Each oriented sample was prepared from a suspension of 0.1 g of the clay-fraction in 2 mL of a solution in three ways: (1) in water and air drying, (2) in water and 550°C drying for 2 h, and (3) in a glycerol solution and air drying (Moore and Reynolds, 1997). The software PDF-4+ 2019 (version 4.19.0.1) and the database v. 4.1903 were used for data interpretation. The content of organic matter, sulphides, hydroxyl groups and carbonates phases was determined by termogravimetrical analysis (TGA). Replicates between 0.015-0.030 g were heated from 30 to 995°C at a rate of 20°C/min with a TGA 4000 Perkin Elmer under two atmospheres: N_2 and O_2 gas (20 mL/min). The results were interpreted in combination with the CHNS, ICP-OES and XRD results. The external specific surface area was measured in 0.2-0.5 g of degassed material (60°C) by the Brunauer-Emmett-Teller (BET) method of nitrogen gas sorption at 77 K in both a Micromeritics Tristar II 3020 and Beckman Coulter SA-3100. The material was prepared from 1 g of original sample gently ground to <400 μm (at least 10 cycles) and discarding the fraction below 64 μm (Bertier et al., 2016). The particle size distribution as volume percent was determined by the Malvern Mastersizer 3000® (double) Laser Diffraction (software version 3.62) assuming the refractive index and density of silica SIO_2 (respectively 1.457 and 2.65 g/cm^3). Samples were dispersed in distilled water by stirring at 2500 rpm and ultrasonic treatment. Measurements of 10 min duration were repeated in the same sample until the results were constant and an average taken.

The pore water chemistry was obtained by mixing 20 mL of deionized water to 10 g of wet clay at room temperature. Sample pH was measured with a pH glass electrode in the water:clay mixture after settling for 24 h and shaking prior to the analysis. The solution was then centrifuged and filtered (0.45 μm) to measure the electrical conductivity with a 0.4-cm sensor, and the soluble elements. The soluble anions and cations were analysed by ion chromatography (Dionex ICS-3000), the alkalinity by titration with H_2SO_4 (HACH digital titrator) and the carbon soluble species using a TOC-V-CSH analyser (Shimadzu ASI-V). The cations in the exchange complex were determined as the difference between the cations extracted with a 1.26 M $SrCl_2$ solution (80 mL) minus the soluble fraction extracted with water (80 mL) after shaking with 5 g of clay for 10 minutes (Edmeades and Clinton, 1981). Due to the high ionic strength of the $SrCl_2$ solutions, sodium, potassium, calcium and magnesium in these extracts were analysed by atomic absorption spectroscopy, AAS (HITACHI Polarized Zeeman Z2300), whereas ammonium was analysed by atomic emission spectroscopy. $LaCl_3$ was added at 20% to standards and samples for the AAS analyses of calcium and magnesium. The cation-exchange capacity (CEC) was determined by copper complex with Cu-triethylenetetramine at pH 7-8, with a photometer at a wavelength for maximum extension of 579 nm (Holden et al., 2012; Stanjek and Künkel, 2016).

To study the consistency and engineering behaviour of the materials, the clay samples were hydrated with different amounts of water for 24 hours in sealed plastic bags prior to index property tests (Head, 2006). The consistency was studied in the <425-μm fraction (250 g) by the determination of two specific water (or moisture) contents: the liquid limit, LL (water content that separates the plastic and liquid states) and the plastic limit, PL (water content that separates the semi-solid and plastic states). The change of clay consistency from plastic to liquid state was determined by the free-falling cone test at a penetration of 2 cm into the wet sample, with duplicates differing ≤0.05 cm (BS 1377:2:4.3, 1990). The change of clay consistency from semi-solid to plastic state was determined by manual rolling wet samples (20 g) until threads of 0.3-cm diameter begin to crumble, with four replicates differing ≤2% moisture content of their PL (BS 1377:2:5.3, 1990 and ASTM D 4318, 2015). To know the range of water content in which the clayey material has a plastic consistency, the plasticity index (PI) was calculated as the difference between the LL and the PL (Head, 2006). All actual moisture contents were determined on a mass % dry basis (Equation 1) after oven drying 5-10 g of material (105°C, 48 h) with duplicates that differ ≤0.5%.

$$\mathrm{MC}(\%) = \frac{m_0 - m_d}{m_d} \times 100 = \frac{m_{0,c} - m_{d,c}}{m_{d,c} - m_c} \times 100 \tag{1}$$

where MC is the moisture content dry basis (%), m_0 is the mass of wet sample before moisture removal (g), m_d is the mass of sample after drying (g), $m_{0,c}$ is the mass of wet sample plus container before moisture removal (g), $m_{d,c}$ is the mass of sample plus container after drying (g) and m_c is the mass of the container (g).

The optimal condition of the clays at which the susceptibility to settlement is reduced was studied by applying the same compactive effort in different hydrated samples (240-540 mL water in 1600-1800 g clays). The compaction was placed into a mould of 5.25 cm radius by 11.55 cm height, in three equal layers subjected to 27 blows each one, by a 2.5 kg rammer of 2.5-cm radius that dropped from a height of 30 cm (BS 1377:4:3.3, 1990). The optimum moisture content (OMC) was selected on the basis of the maximum dry (bulk) density (MDD) after the compaction. To calculate the particle density of the solids, the specific gravity (G_s) was obtained by triplicate tests at 20°C, in desiccated materials of <0.2 cm size (50 g) using air-dried pycnometers of 50 mL (BS 1377:2:8.3, 1990 but 20°C instead of 25°C). The porosity (n) was calculated as a percentage following Equation 2 (Equation 3 for the optimal conditions). The slight difference between n and void ratio (e) is that the latter measures the void volume (the sum of V_{air} and V_w) in relation to the volume of the solid instead of the total volume. Thus, e (normally expressed as a ratio) can be >1, but n cannot be higher than 100%.

$$n(\%) = \frac{V_{air} + V_w}{V_T} \times 100 = \frac{V_T - V_s}{V_T} \times 100 \tag{2}$$

$$n_{opt}(\%) = \frac{V_T - \frac{m_{d,OMC}}{G_S}}{V_T} \times 100 \tag{3}$$

where V_{air} is the volume of the air (mL), V_w is the volume of the water (mL), V_T is the total volume (mould of 1000 mL),

V_s is the volume of the solid particles (mL), $m_{d,OMC}$ is the dried mass of sample at the OMC (g) and Gs is the specific gravity (unitless).

Assuming that volumes of voids filled with air are constant ($\eta_{air,i}$), lines at different air void ($\eta_{air,1}$, $\eta_{air,2}$...) and saturation values can be drawn as a function of dry bulk density ($\gamma_{d,i}$) relative to the moisture content ratio (Equation 4). The difference between air void line (η_{air}) and saturation (s) is that the latter ratio measures the volume of water in relation to the void volume instead of the total volume. The zero-air void line (η_{air} = 0) corresponds to the maximum saturation ratio (s = 1).

$$\gamma_{d,i} = \frac{(1-\eta_{air,i}) \cdot G_s \cdot \gamma_w}{1+MCi \cdot G_s} \text{ being } \eta_{air,i} = \frac{V_{air,i}}{V_T} \quad (4)$$

where $\gamma_{d,i}$ and MC_i are the dry bulk densities (g/cm³) and the moisture contents (ratio) corresponding to the air void constant i, $\eta_{air,i}$ is the line corresponding to an air void and saturation constant i related to the total volume (ratio), G_s is the specific gravity (unitless) and γ_w is the density of water (1 g/cm³).

The specific value for the air void and saturation line after compaction at the optimal conditions ($\eta_{air,opt}$) was calculated with the $\gamma_{d,i}$ and MC_i equal to MDD and OMC (ratio), respectively (Equation 5).

$$\eta_{air\ opt}(\%) = \left(1 - \frac{MDD\ (1+OMC \cdot G_s)}{G_s \cdot \gamma_w}\right) \times 100 \quad (5)$$

The potential swell (S %) was determined based on its relationship with the Atterberg limits by the well-defined empirical Equation 6 (Seed et al., 1962):

$$S(\%) = 216 \cdot 10^{-5} \cdot PI^{2.44} \quad (6)$$

where PI is the plasticity index (%).

The hydraulic conductivity or permeability coefficient (K) was measured in aluminium permeation cylinders by centrifuging at 25 gravities (Regadío et al., 2020). Model liners were compacted into of 5 cm radius by 10 cm height cylinders after being hydrated with tap water at the OMC. Leachate from a municipal solid waste landfill and rainwater were used as permeating fluids. Rainwater was normally used for permeating model liners previously permeated with landfill leachate. Eleven cylinders on average were assembled per centrifuge test, each one connected to an intake line. All intake lines came from a common tank that provided continuous permeation to all the cylinders by applying a pressure of 1.1 bar. The tests were conducted under a 50% CO_2/N_2 anaerobic gas (for leachate permeation) or under compressed air (for rainwater permeation). The fluid head in the tank that provided the permeating fluid was measured every ≈2 days during 2.5 weeks under leachate permeation, or 4.5 weeks under rainwater permeation. The K measured in the centrifugal permeability tests ("experimental model") had a 25-fold enhanced gravity. To calculate the corresponding real value in the field (K "prototype"), the scaling law (Ng, 2014) was applied in the falling head equation for less permeable soils (Head, 1994) adapted to this method (Equation 7):

$$K = \frac{K_{exp.m} \cdot n}{n^2} = \frac{\frac{aL}{At}\ln\left(\frac{h_0}{h_1}\right)}{n} \text{, being } h_1 = \frac{h_0 - (h_0 - h_{1\#})}{\#} \quad (7)$$

where $K_{exp.m}$ is the hydraulic conductivity in the experimental model (m/s), n is the enhanced gravity applied in the centrifugal experiment (25), a is the cross area of the tank (0.06158 m²), L is the lengh of the liner specimen (0.10000 m), A is the cross area of the liner specimen (0.00785 m²), t is the time period considered for the calculation (seconds), h is the head in the tank at the initial (if subscript 0) and final (if subscript 1#) points, # is the number of model liners connected to the tank.

3. RESULTS

3.1 Elemental composition and mineralogy

The four clayey substratum were mainly composed of silicium and aluminium, followed by iron, potassium, hydrogen and magnesium (Figure 1). This agreed with the expected high presence of clay minerals, potentially higher in Coal Measures Clays (see below). Silica, potassium, titanium, and especially magnesium were higher in the LC. The OC was notable for its high content in calcium, carbon, sulphur (g/kg) and strontium (mg/kg), with lower silica, iron and phosphorus content. The Coal Measures Clays were notable for their high concentration of aluminium, nitrogen and manganese, whereas the concentration of calcium, sodium, potassium, strontium and boron were the lowest within the four samples. In the case of SCMC, there was more carbon, nitrogen and sulphur than in DCMC. (Figure 1).

All samples contain smectite, illite, kaolinite and chlorite but in different proportions (Figure 2). Smectite was most important in LC, illite together with kaolinite in OC, and kaolinite (followed by illite) in the Coal Measures Clays. To a lesser extent, phlogopite mica was detected in LC, chlorite in both LC and OC, and interstratification (mixture of layers) of illite/smectite in OC and the two Coal Measures Clays.

Quartz and feldspars were the most important phases in all materials. These are accompanied by oxides except in the OC, which mainly contained calcium carbonate and iron sulphide (calcite and pyrite). Also relatively high levels of sulphides were found in SCMC, whereas, there were fluorides and oxide-fluoride in DCMC (Figure 3). The mass loss through heating due to dehydroxylation (associated with the mass of clay minerals) was the highest in the Coal Measures Clays (5-9%), while the highest mass loss due to decomposition of organics, sulphides and carbonates phases corresponded to OC (2, 4 and 5% respectively). Particulate organic matter as an associated material in these clays was especially high in the OC. It varied from 1.1 to 3.5% in the OC, in contrast to LC, SCMC and DCMC, which gave steady values of 1.7, 1.5 and 1.8%, respectively. The variability in the particulate organic matter content in the OC was due to its presence in many diverse forms: disseminated organic matter, coarse lignite fragments and fossils. Similarly, the carbonate phases in the OC varied from 4.5 to 6% due to the spread of carbonaceous shells.

3.2 Pore water and mineral surface characteristics

The LC had the highest natural moisture content followed by the OC (39 and 25 ± 2%, respectively), whereas SCMC and DCMC had the lowest values (10 and 12 ± 1%, respectively). This indicates a decreased water absorption and porosity from LC > OC > Coal Measures Clays. The pore water composition of the clayey materials of the OC was

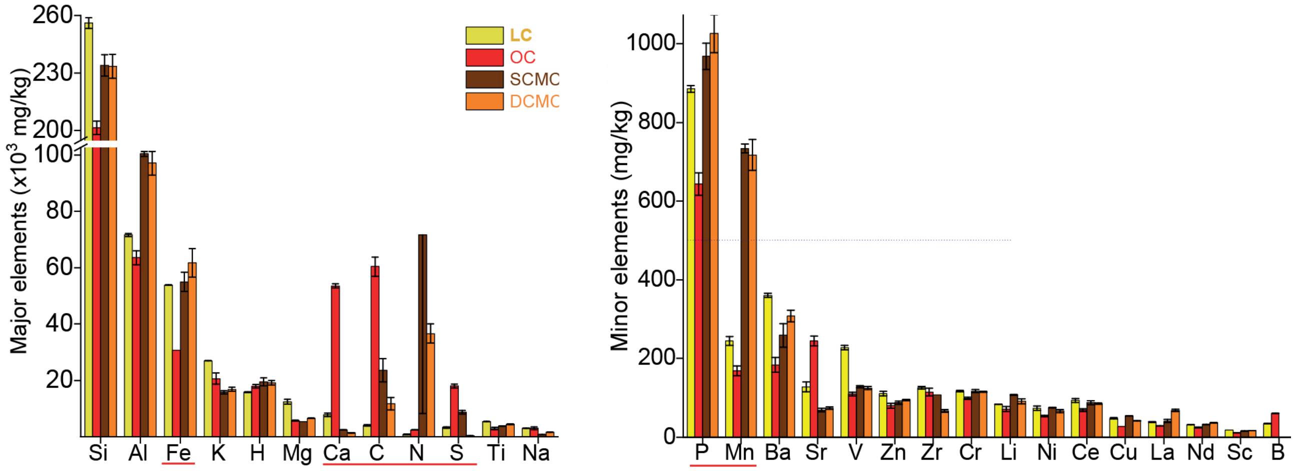

FIGURE 1: Elemental composition of London Clay (LC), Oxford Clay (OC), shallow Coal Measures Clay (SCMC) and deep Coal Measures Clay (DCMC).

the most basic due to the presence of calcium carbonate phases (pH 9.0) and the LC was close to neutral (pH 7.3). In contrast, the SCMC and DCMC were acidic (pH 3.8 and 5.4, respectively). Consistent with this, alkalinity was only present in the OC (10.3 mmol/kg as $CaCO_3$) and LC (1.7 mmol/kg as $CaCO_3$). The total dissolved ion content in terms of electrical conductivity in aqueous extracts (L/S 1:2.5) was between 1-3 mS/cm, except for DCMC (0.1 mS/cm) and for OC (5 mS/cm). The predominant soluble anion in all samples was sulphate (SO_4^{2-}), mainly balanced by calcium (Ca^{2+}) and sodium (Na^+) in both the LC and OC, by magnesium (Mg^{2+}) and Ca^{2+} in the SCMC and mostly Na^+ in the DCM. In all cases the concentration of potassium (K^+) was very low and ammonium (NH_4^+) was not detected. Only the OC had a significant content of soluble carbon in the pore water (10 mg/g).

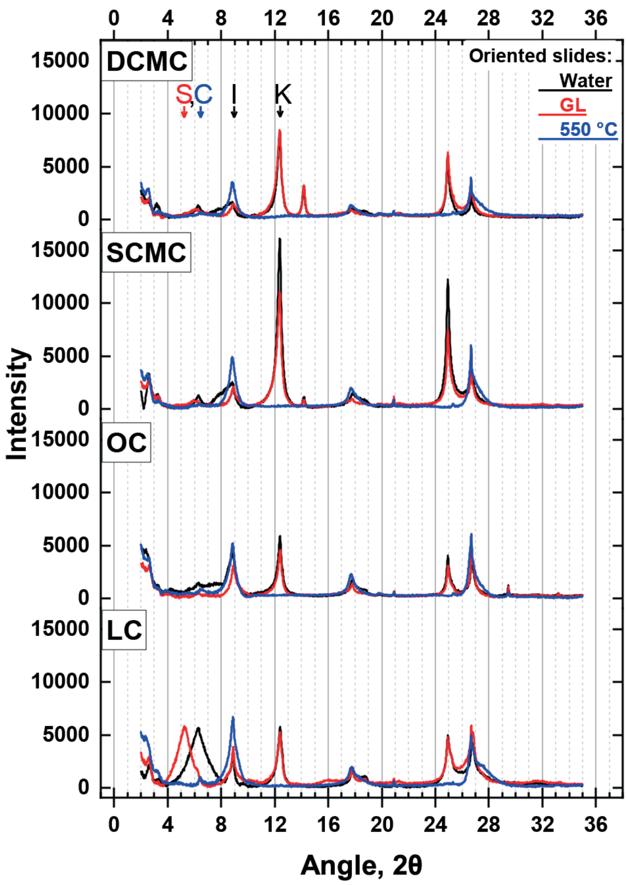

FIGURE 2: Sheet-silicate mineralogy of London Clay (LC), Oxford Clay (OC), shallow Coal Measures Clay (SCMC) and deep Coal Measures Clay (DCMC). Water: water and air drying preparation, GL: glycerol and air drying preparation, 550°C: water and 550°C drying preparation. S: smectite d001 reflection under GL preparation, C (chlorite) and I (illite) d001 reflections under all three preparations. K: kaolinite d001 reflection under water and GL preparations.

As expected the exchangeable cations on the negatively charged sites of the clays and particulate organic matter were similar to the most abundant in the pore water. The sum of exchangeable cations often exceeded the total charge of the clay (CEC), due to high concentrations of Ca^{2+} released by dissolution of carbonate minerals. Thus, the CEC was measured directly instead of estimating this from the sum of exchangeable cations, to avoid bias from Ca^{2+}. The CEC at pH 7-8 decreased in the order: LC (26 cmol+/kg) > OC (16 cmol+/kg) > Coal Measures Clays (13 cmol+/kg). The external specific surface area increased in the order: LC < OC < DCMC < SCMC (9.3 ± 0.3, 12.8 ± 3. 6, 31.0 ± 1.0, 53.7 ± 1.0 m^2/g, respectively). The highest values corresponded to the material with the highest amount of hydrous aluminium phyllosilicates minerals (Coal Measures Clays), followed by the material with the highest amount of particulate organic matter (OC). Nonetheless, the total surface area (external plus internal) of the LC may be one of the largest due to its higher content of expandable clay minerals (illite + smectite) than non-expandable ones (kaolinite + chlorite).

3.3 Consistency classification and properties

The moisture contents at PL and LL were determined to identify clays susceptible to dispersion and excessive shrinkage in the field (Table 2). These parameters can distinguish between silt- and clay- size, and organic or inor-

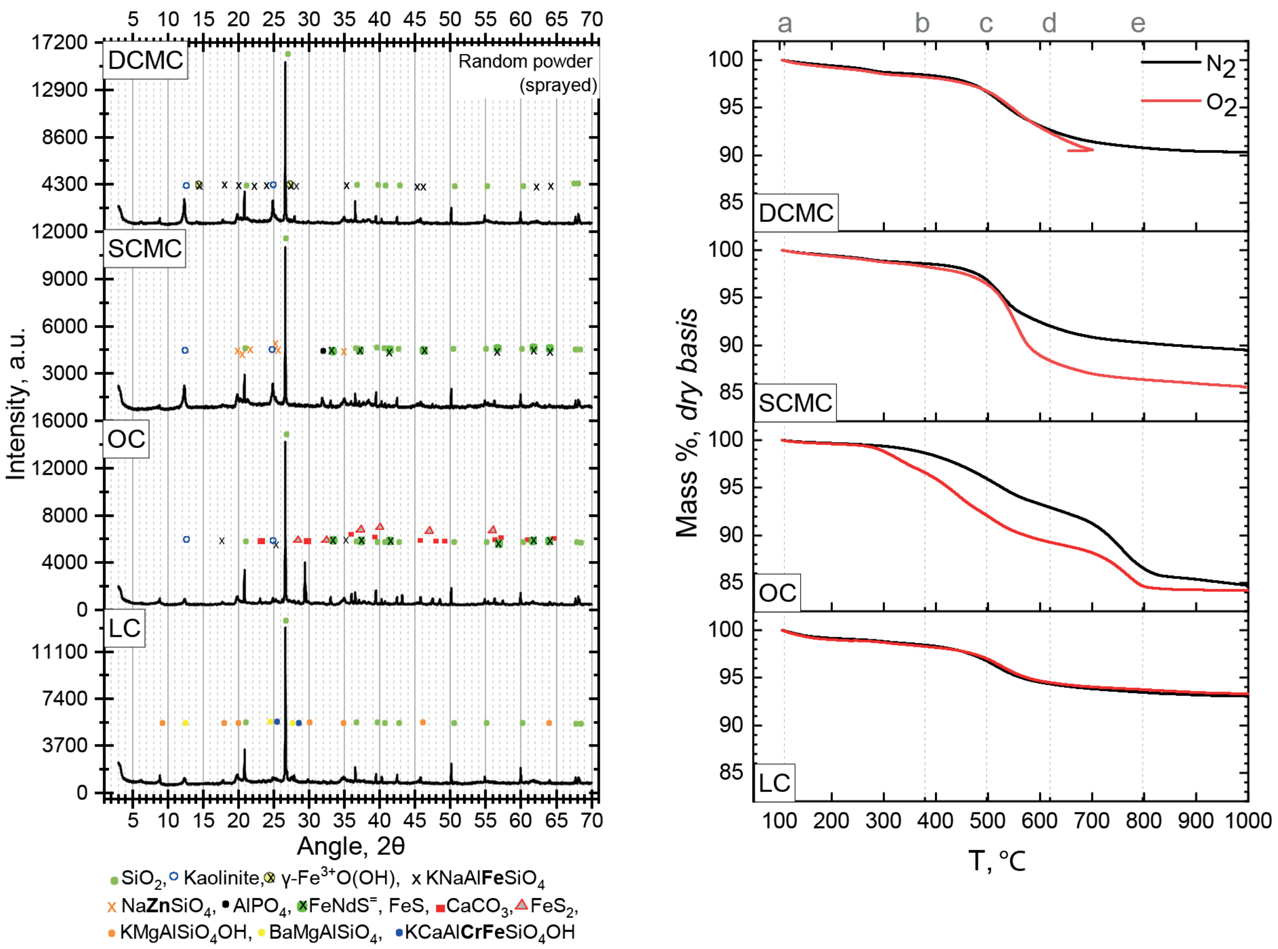

FIGURE 3: Left: Global mineralogy by sprayed random powder of London Clay (LC), Oxford Clay (OC), shallow Coal Measures Clay (SCMC) and deep Coal Measures Clay (DCMC). Right: TGA curves with mass relative to the mass after dehydration (removal of interlayer water): (a-b) mass loss mainly due to thermal decomposition of organic matter, (b-c) mass loss due to thermal decomposition of sulphides, (c-d) mass loss due to dehydroxylation, (d-e) mass loss due to thermal decomposition of carbonate and (e-end) materials vitrification and formation of new phases (recrystallization).

ganic character. All clays had a LL >20%, confirming that they were cohesive materials. The LL and PI varied in the order: LC > OC > Coal Measures Clays, consistent with the dominant sheet silicate in each material: smectite, illite/kaolinite and kaolinite, respectively. The LC and OC had high plasticity, high toughness and high to very high dry strength (Figure 4). The high plasticity of LC was notable for the broad range of water contents at which this clay had plastic consistency (from 28 to 79%, i.e., PI = 51%), twice that of the OC. This is due to the presence of smectite (expandable clay mineral) and the higher content of clay-size particles in the LC compared with the other clays with a higher silt content. Ninety vol% of the LC was composed of particles ≤52 μm, whereas 90 vol% of the DCMC, SCMC and OC were made up of particles ≤130 μm, ≤136 μm and ≤185 μm, respectively. The two Coal Measures Clays gave similar results: intermediate plasticity, medium toughness and high dry strength. The potential to swell decreased from LC >> OC > SCMC > DCMC (30.2, 5.3, 2.6, 2.4%), as expected from the mineralogy and PI results. All studied materials had no dispersive clay fines as these do not occur in clays from intermediate to high plasticity with smectite. On the contrary, dispersive clays typically appear in soils classified as clayey of low plasticity (CL), sometimes also in silty and/or sandy soils with low plasticity (ML, CL-ML) (Figure 4).

The “A-line” on the plasticity chart (Figure 4) denotes the empirical boundary between inorganic materials and clays (above line) and organic clays and clastic silts (below line). The OC fell on the dividing line between inorganic and organic categories, while Coal Measures Clays and the LC fell above the line in the inorganic region, being the DCMC close to the organic silts, and the LC the most inorganic clay.

3.4 Compaction and permeability behaviours

Clays are normally compacted for placing and constructing the clay liners because to increase the shear strength and bearing capacity, which limits future settlement. In addition, the void ratio and permeability is decreased, and variations in volume change are less pronounced. Consequently, clays are less susceptible to cracking that would offer preferential flow paths for leachate leakage and groundwater seepage. To optimise this, clays should be compacted close to the OMC, the quantity of water nec-

TABLE 2: Moisture contents corresponding to the Atterberg consistency limits (<425-μm fraction).

	Linear regression		LL, %	PL, %		PI, %
	Slope	Intercept	Value (X when Y= 20)	Value	RSD, percent	Value
LC	0.552	-23.339	79	28	4.4	51
OC	1.136	-40.404	53	29	4.3	24
SCMC	1.758	-49.944	40	22	4.0	18
DCMC	1.368	-40.052	44	25	0.9	19

Linear regression: relationship of the cone penetration (Y-axis in mm, as a reverse measure of the shear strength) on the moisture content (X-axis in %), LL (or WL): liquid limit, PL (or WP): plastic limit, PI (or Pi): Plasticity index, RSD: relative standard deviation (the standard deviation divided by the average and multiplied by 100), LC: London Clay, OC: Oxford Clay, SCMC: shallow Coal Measures Clay, DCMC: deep Coal Measures Clay.

essary to achieve the maximum dry (bulk) density. Under the same compaction effort, the OMC followed the order LC > OC > Coal Measures Clays, while the maximum dry (bulk) densities followed the inverse sequence (Figure 5). The clay sequence for OMC agreed with those for air void lines and for porosity (both after compaction at the optimal conditions), and with the higher plasticity of LC, followed by OC, which could accommodate more water to achieve their maximum dry densities than the Coal Measures Clays. The particle density of the solids, in terms of specific gravity, followed the order LC > Coal Measures Clays > OC due to the higher particulate organic matter content of the last one (Figure 5). Both Coal Measures Clays had very similar consistency and engineering behaviour (Figure 4 and Figure 5).

The K for leachate and rainwater through the most plastic clays (LC and OC) amended with sandy materials was measured over time. In all three tests run with landfill leachate, K slightly increased from initial values of 0.1 and $0.3 \cdot 10^{-9}$ m/s to a maximum of 0.6 and $0.8 \cdot 10^{-9}$ m/s after

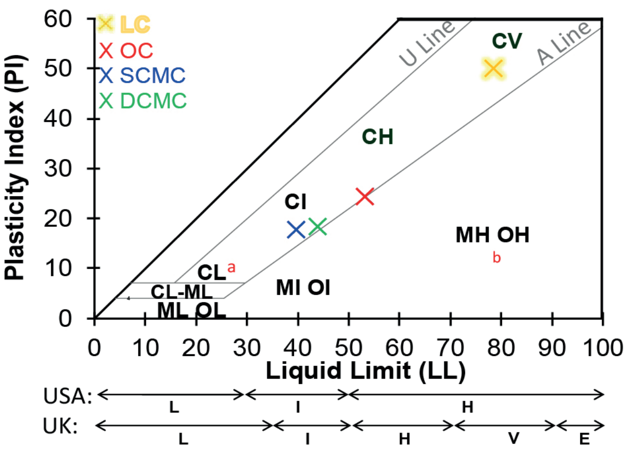

FIGURE 4: Plasticity chart for soil classification (<425-μm fraction) of London Clay (LC), Oxford Clay (OC), shallow Coal Measures Clay (SCMC) and deep Coal Measures Clay (DCMC). Divisions of plasticity in: L: low, I: intermediate, H: high, V: very high and E: extremely high, according to USA and UK. O: significant organic material; C: clayey; M: silty and/or sandy (Unified Soil Classification System, USCS). [a] dispersive clay fines, [b] non dispersive clay fines. U-line: upper reference bound of PI for natural soils defined by two equations: PI = 7 if LLs ≤16, and PI = 0.9 (LL- 8) if LLs >16. A-line: reference boundary of PI between the clay soils (above line, mostly inorganic) and the silt soils (below it) defined also by two equations: PI = 4 if LLs ≤25.5, and PI = 0.73 (LL- 20) if LLs >25.5 (Casagrande, 1947).

an initial time equivalent to 8-15 yrs. Then, K decreased to comparable values at the start (Figure 6). Only on three separate occasions during leachate permeation through liners of 20% sand, K exceeded the maximum legal limit: in the beginning (between 0 and 3.1 yrs), in the middle (between 8.2 and 10 yrs) and towards the end (between 25.4-27.1 yrs). However, the average of K measurements taken in 9 different periods over a total modelled time equivalent to 32 yrs prototype of leachate through the liners of 20% sand was $0.8 \cdot 10^{-9}$ m/s (± $0.6 \cdot 10^{-9}$), and lower through the liners of 10% sand ($0.3 \cdot 10^{-9}$ m/s (± $0.2 \cdot 10^{-9}$)). When changing to rainwater though liners of ≈6% sand, there was also in the beginning a slight increase of K with time up to $0.5 \cdot 10^{-9}$ m/s, followed by a decrease with a stabilization around $0.2 \cdot 10^{-9}$ m/s (±$0.1 \cdot 10^{-9}$) from year 44 onwards (30 yrs of leachate permeation followed by 14 yrs of rainwater, Figure 6). The average of K for rainwater permeation taken over 16 periods of time within the total time modelled (53 yrs prototype) was $0.3 \cdot 10^{-9}$ m/s (±$0.1 \cdot 10^{-9}$) though the liners of ≈6% sand. The accumulated K calculated as a single measurement over the entire test time (≈19 days each test) was very close to the average K calculated with all intermediate measurements taken every ≈2.3 days over the 19-day tests (Table 3). This together with the small standard deviations of the K in liners with 10% or less sand, denotes that their K variation describe above was not very significant.

4. DISCUSSION

Based on the previous analysis, the feasibility of the four natural clayey substrata to attenuate landfill leachate is discussed below. Although its composition varies, landfill leachate always contains high concentrations of Na^+, K^+, bicarbonate and chloride, with significant NH_4^+ and organic compounds. The heavy metal content is generally relatively low, often of no major concern and limited to chromium, nickel and zinc (Aucott, 2006; Kjeldsen et al., 2002). As NH_4^+ and K^+ are major elements in landfill leachate and virtually absent in these Ca-clay mineral liners, both can be used as tracers in leachate migration studies. The differences between the clayey substrata on porosity, density, sorption, surface, plasticity, permeability are due to their origin, particle size and mineralogy (Table 1 and from Figure 1 to 3). The presence of carbonaceous material is characteristic of clays formed in alluvial or shallow waters, as is the case with Coal Measures Clays (Bain, 1971). Smectite is often found interstratified with illite and in mixtures with chlori-

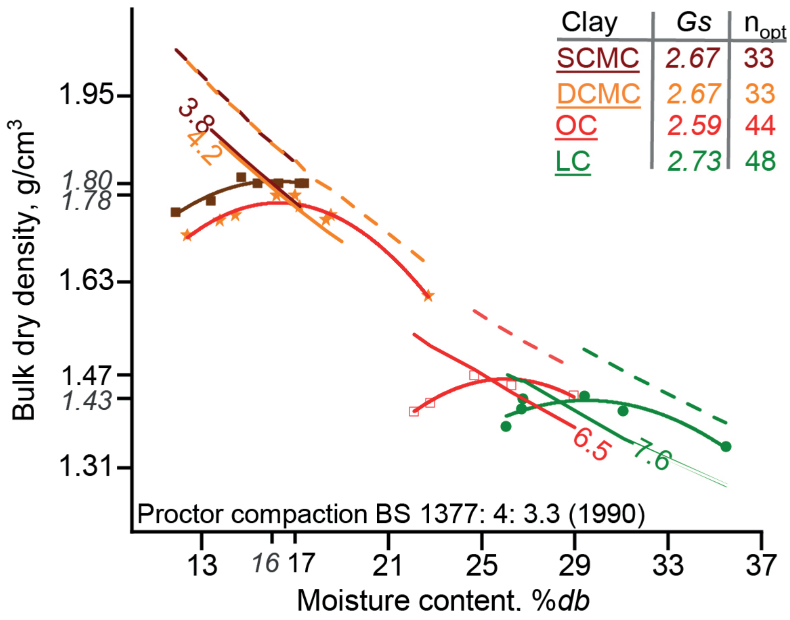

FIGURE 5: Air void lines (air vol% of the total volume) and compaction curves under Proctor BS 1377:4:3.3 (1990) to estimate the (optimum) moisture contents at which the dry bulk densities are maximum. Dashed straights: zero air line or full (water) saturation lines (s=1). Solid straights: air void lines at the optimum. n_{opt}: porosities at the optimum (air plus water vol% of the total volume). Gs: specific gravity (unitless). LC: London Clay, OC: Oxford Clay, SCMC: shallow Coal Measures Clay and DCMC: deep Coal Measures Clay.

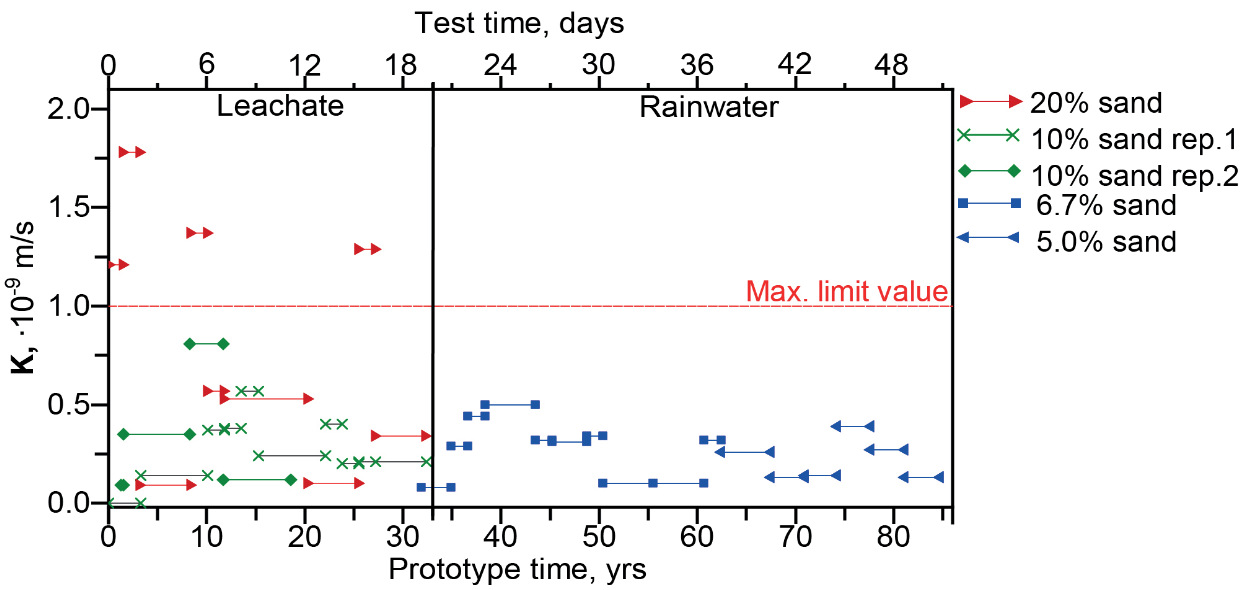

FIGURE 6: Hydraulic conductivities (K) of landfill leachate (from year 0 to 33) and rainwater (from year 33 to 82) through model liners (≈11) of averaged compositions made of clays and mixtures of clays with sandy non-cohesive materials. The mixtures were used to decrease the plasticity of London and Oxford clays and therefore its associated risk of shrinkage.

te and sometimes kaolinite in deep sea marine sediments, as for the LC and OC. A high silica-to-aluminium ratio is characteristic of clays with smectite minerals (Weaver and Pollard, 1973), agreeing with that the LC had the highest values of this ratio and of smectite.

4.1 Evaluation as attenuation liners

In addition to the low permeability that they provide, compacted clays can attenuate leachate pollutants by sorption, dilution, redox transformations, biodegradation, precipitation and filtration (Allen, 2001; Griffin et al., 1976; Thornton et al., 1993). Attenuation here refers to a reduction of the mass of pollutants by naturally-occurring processes (Regadío et al., 2015). These attenuation processes occur simultaneously and can affect more than one pollutant in leachate. By sorption, pollutants are attached to mineral phases or particulate organic matter by a physical or chemical process, which encompasses ion exchange, adsorption, absorption and chemisorption. By redox transformations, organic and metal compounds are converted

TABLE 3: Hydraulic conductivities in m/s (K) as a single accumulated measurement within the entire test and as an average of the intermediate measurements taken every 2-3 days throughout the test.

Average liner composition	Permeating test time	Permeating fluid	One accumulated measurement	Average (± standard deviation) of intermediate measurements
Clays with 20% sand	From day 1 to 19	Landfill leachate	$0.45 \cdot 10^{-9}$	$0.81 \cdot 10^{-9}$ ($\pm 0.61 \cdot 10^{-9}$)
Clays with 10% sand (1)	From day 1 to 19	Landfill leachate	$0.21 \cdot 10^{-9}$	$0.25 \cdot 10^{-9}$ ($\pm 0.17 \cdot 10^{-9}$)
Clays with 10% sand (2)	From day 1 to 19	Landfill leachate	$0.29 \cdot 10^{-9}$	$0.27 \cdot 10^{-9}$ ($\pm 0.33 \cdot 10^{-9}$)
Clays with 6.7% sand	From day 19 to 36	Rainwater	$0.25 \cdot 10^{-9}$	$0.28 \cdot 10^{-9}$ ($\pm 0.14 \cdot 10^{-9}$)
Clays with 5.0% sand	From day 36 to 48	Rainwater	$0.22 \cdot 10^{-9}$	$0.22 \cdot 10^{-9}$ ($\pm 0.11 \cdot 10^{-9}$)

(1) and (2) are replicates.

into less toxic or immobile forms by electron transfer reactions. By biodegradation, organic pollutants are chemically decomposed by microorganisms. By precipitation, metallic pollutants become less bioavailable or mobile. By filtration, larger pollutants such as metal-organo complexes in the leachate remain physically trapped within the liner fabric.

The surface of soil particles is critical for the chemical reactions, sorption, colloid filtration, and transport of contaminants. All clayey materials and especially the OC contained particulate organic matter (Table 1, Figure 3 right, Figure 4) which has a large surface area and CEC. Particulate organic matter is important for the attenuation of contaminant molecules by sorbing them to its surface or fostering microbial communities that would breakdown the contaminants to less toxic or nontoxic compounds (see biodegradation below). The CEC in particulate organic matter and also in clay minerals is especially important for sorption. In this case cations in the pore water are sorbed by clays to neutralize their negative charge created by unbalanced substitutions of their structural cations. Sorbed native cations can be replaced by cationic pollutants in the leachate. Illites (present in the four clays here) have high affinity for selective sorption of NH_4^+ and K^+ due to their size compatibility with the interlayer (exchange) sites in this clay lattice (Griffin et al., 1976). Smectites (in the LC) also fix these cations but this destabilizes smectitic minerals, resulting in illitization, i.e., partial collapse of smectites with their subsequent conversion into illite. In the case of larger cations, organic cations or organometallic complexes in leachate, smectites sorbed these species preferentially relative to smaller, inorganic or uncomplexed metals (Koutsopoulou and Kornaros, 2010). This is because for the same valence these weakly hydrated cations are the easiest to sorb in the exchange sites than stronger hydrated small cations (Teppen and Miller, 2006), and only smectites have an exchanger interlayer space large enough to accommodate them. Smectite, along with vermiculite (interlayer Mg), has a high CEC, while illite has mid-range values and kaolinite very low values. Thus, the capacity to reduce the concentration of cationic pollutants in leachate by cation exchange reactions follows the order LC > OC > Coal Measures Clays. CEC generally increases with pH due to the development of greater negative charge (average pH in leachate is 7-8).

Anion sorption (bicarbonate, chloride, sulphate from leachate) is less efficient and very similar for the different clay minerals (kaolinite, smectite). It occurs at OH^- ions exposed on the mineral edges and is enhanced by positively charged iron-oxide colloids (present in LC and Coal Measures Clays) associated with clays (Raymahashay, 1987). Bicarbonate is the major inorganic anionic compound in leachate and largely determines the acid-base neutralisation potential of the system. This is good for Coal Measures Clays which have no pH buffering capacity to attenuate acidic episodes caused for example by oxidation of sulphides (pyrite) by infiltrating rainwater (Thornton et al., 2001). This oxygenated water can re-oxidized sulphide phases, resulting in the release of previously attenuated metals that precipitated earlier in such immobile phases (Regadío et al., 2013). Bicarbonates in the leachate itself would provide acid neutralization capacity to Coal Measure Clays which lack calcite. The acidity in Coal Measures Clays most likely arises from oxidation of pyrite in the upper, weathered zone, which produces a low pH, gypsum and amorphous iron oxides as by-products. Chloride is not significantly attenuated and mainly diffuses through the clay liner, together with Na^+ and the cations displaced from the exchange sites of clays (usually Ca^{2+} and Mg^{2+} substituted by NH_4^+, K^+) (Regadío et al., 2012; Zhan et al., 2014). These elements are diluted by the receiving groundwater and are generally not a problem due to its low toxicity even at relatively high concentrations. Sulphate in leachate is attenuated by anaerobic microbial reduction, a common redox process in landfills (Batchelder et al., 1998). All the studied clayey substrata contain redox-sensitive species, the most important being pyrite in the OC and SCMC, and iron oxides in the LC, SCMC and DCMC. As a result, these clays support the metals to precipitate as sulphides in the liner and the sulphate is in low concentration in leachate.

Biodegradation is also accompanied by changes in redox potential in the landfill, which results in transformation of organic and inorganic species by reactions under aerobic and a range of anaerobic conditions. Depending on the specific redox conditions in the landfill and liner (aerobic, nitrate-reducing, iron-reducing, sulphate-reducing, etc.), one or other organic compounds can be biodegraded (Bright et al., 2000). The concentration of the oxidising agents and their reduced species in the leachate indicates the redox conditions (Taylor and Allen, 2006). Differences in clay minerals have a minor effect on the biodegradation of organic pollutants than on the sorption of inorganic pollutants. This is because organic pollutants are attenuated mainly by anaerobic biodegradation (Thornton et al., 2000; Bright et al., 2000; Adar and Bilgili, 2015), rather than by sorption to clay minerals, in which case only smectite, chlorite and vermiculite would show significant organic sorption (Koutsopoulou and Kornaros, 2010). The decomposition of organic compounds down to water, methane and carbon dioxide, depends on the establishment of an appropriate microbial population. The native particulate organic matter of clay materials is essential to support in situ biological activity in liners, with the highest organic matter content found in the OC (followed by SCMC), sampled from a fossiliferous location (Martill et al., 1994). Anaerobic degradation in the liner is also sustained by the microbial inoculum in the leachate itself.

The attenuation of heavy metals in leachate is associated with particulate organic matter and mineral phases, including clay minerals in liners, and occurs by a combination of sorption, redox transformation and precipitation processes (Fannin, 2006). These processes are supported by specific mineral phases such as sulphate-bearing species (pyrite, gypsum), iron and manganese oxides and oxyhydroxides, and clays (mainly smectite and illite) (Fisher and Hudson, 1987). The studied materials all contain a high content of clay minerals which assists retention of heavy metals. The OC and SCMC contain pyrite, and the LC, SCMC and DCMC contain iron/metal oxides. The high native particulate organic matter content of the OC favours sorption of metals, whereas the dissolved organic compounds in leachate favour the formation of soluble metal-organo complexes. Despite the fact that metal-organo complexes are dissolved in leachate and therefore mobile, they can be attenuated by filtration due to their larger size (Christensen et al., 1996; Gregson et al., 2008). However, a proportion of metals complexed with dissolved organic matter or associated with colloids in leachate may not be attenuated (Thornton et al., 2001).

4.2 Evaluation as containment liners

None of the clays here were identified susceptible to dispersion in the field. Dispersive clays resemble normal clays but can be highly erosive and susceptible to severe damage or failure. Soils of high plasticity silt (MH in the USCS classification, Figure 4) and smetite-rich materials (LC) rarely contain dispersive clays. In the case of the LC, smectites are responsible for the adhesion forces between particles, which helps to prevent dispersion and thus soil erosion. The locations of the clays on or above the "A-line" on the plasticity chart (Figure 4), denotes a relatively low silt content so they are easy to compact well, resulting in low erodibility. The high OMCs of the OC and LC (25-29% with maximum dry bulk densities of 1.43-1.47 g/cm^3), indicate the abundance of clay-sized particles (heavy clays) as silt-rich soils have medium values and sandy materials have very low values. Coal Measures Clays had 16-17% of OMC with maximum dry bulk densities of 1.78-1.80 g/cm^3, being more characteristics of sandy-clay materials. In the case of shrinkage potential, only LC presented high risk because of the smectites. Concordantly, the LC was classified as high shrinkage (average shrink limits of 16.8±4.8%) compared to the OC as medium shrinkage (14.5 ±2.0%) in Hobbs et al. (2019). In addition, smectites can also sorb larger quantities of water that decrease the soil strength, causing destructive landslides and slope failure (Borchardt, 1977; Wagner, 2013; Yalcin, 2007). To ensure landfill liner stability, clays should have PIs of 15-30% (25% is good) and clays with PI >40% should not be used on their own.

The LC and OC are plastic clays and thus contain little sand and much clay, with a particular abundance of swelling minerals (illite and smectite). These are expandable sheet silicates with desirable properties such as erosion resistance, low permeability and excellent ability to attenuate pollutants, due to high surface area and CEC (e.g., the LC). Thus, they have been used globally to improve compacted soil liners (Ruiz et al., 2012) and to achieve permeabilities in geosynthetic clay liners in the low range of $0.10 \cdot 10^{-9}$-$0.01 \cdot 10^{-9}$ m/s (Egloffstein, 2001). A key limitation is that smectites are plastic minerals very sensitive to the cation occupying the hydrated interlayer, which results in a high potential for swelling or shrinkage in water or leachate, respectively. This property can induce instability and cracks in compacted clays and increase leakage through liners (Borchardt, 1977; Wagner, 2013; Yalcin, 2007). This risk can be reduced by compaction and by addition of sand (Tanit and Arrykul, 2005; Varghese and Anjana, 2015). No consensus exists though on whether applying a water content lower (Widomski et al., 2018) or higher (Benson et al., 1999) than the optimum, will limit the shrink potential and thus desiccation cracking, ensuring K values $\leq 1 \cdot 10^{-9}$ m/s. A further drawback of smectites is that their alteration to newly formed illite or even kaolinite results in much less chemo-mechanical stable materials than kaolinite-rich and illite-rich samples that are not originally derived from smectite (Zhao et al., 2007). This illitization at the expense of the smectite content can occur after ammonium and potassium sorption from the landfill leachate (Regadío et al., 2015), reducing the CEC of the clay by ≤10%.

The Coal Measures Clays were easily compacted until negligible air was present in their voids (4%), which is convenient to achieve a low permeability in the liner. The OC and LC can achieve the lowest K (Maritsa et al., 2016) due to their high plasticity, but also have a higher shrinkage risk, with consequent risk of increased K due to desiccation cracks. This is especially critical in the LC as its PI is >30%: the low K of compacted clay liners with such high plasticity could increase above the design specification after repeated cycles of shrinkage-by-drying and swelling-by-wetting, and never recover its initial value even after rewetting (Widomski et al., 2018). Conversely, low plasticity clays have a K that remains nearly constant and within the design specification, even after several drying/wetting cycles. Another advantage is that non-plastic clays exhibit predominant vertical instead of horizontal deformation, the latter being predominant in plastic clays. Vertical deformation presents a lower risk of desiccation cracking in a compacted clay liner. Thus, for the centrifuge permeability tests the sandy materials were added to both clays to decrease their plasticity (Mansouri et al., 2013; Tanit and Arrykul, 2005; Varghese and Anjana, 2015).

Adding non-cohesive materials decreases the LL and swell index, but should be done with caution to avoid an excessive increase in K (Lee et al., 2005). The K varies depending on the solid properties (surface area, particle sizes, porosity, tortuosity…), and many factors such as:

- Lab or field measurements (Allen, 2000; Benson et al., 1999; Shackelford and Javed, 1991);
- Compaction (Herrmann et al., 2009);
- (liquid) saturation ratio (Benson et al., 1999; Widomski et al., 2018);
- Other minor construction variables (Benson et al., 1999);
- Permeating liquid (di Emidio et al., 2017; Francisca and Glatstein, 2010; Jo et al., 2001; Lee et al., 2005; Singh and Prasad, 2007; Stepniewski et al., 2011; Uma Shankar and Muthukumar, 2017);
- Methodology (Sandoval et al., 2017);
- Passing of time and wet-dry seasonal variations (di Emidio et al., 2017; Egloffstein, 2001; Mitchell and Jaber, 1990; Stepniewski et al., 2011; Widomski et al., 2016).

The low K measurements ($0.2\text{-}0.8 \cdot 10^{-9}$ m/s) showed that these clayey substrates are chemically compatible with landfill leachates and promising candidates for use in the design of landfill bottom liners to minimize leachate migration as dual impermeability-attenuation barriers. The K values in the clay liners with ≤10% sandy materials under long-term leachate and rainwater permeation were below the most common maximum regulatory criterion ($1 \cdot 10^{-9}$ m/s) over a time equivalent to 85 yrs. These experimental results are in line with the graphical and multivariate regression of Benson et al. (1994), which estimates K values of $<1 \cdot 10^{-9}$ m/s for materials with at least 20% LL, 7% PI, 30% fines and 15% clays. No significant differences were found between the K values measured between different periods of time. The little variation is most likely due to the not complete (but almost) saturation of the compacted liners at the beginning (Darcy, 1856). This results in measurements of unsaturated K whose values are typically lower than those of saturated K as the water would be strongly attracted by the tension of the dry soil. The possible loss of the hydraulic connection when the pore water at the bottom of the model liner is transferred to the collector during spinning would also promote unsaturated conditions with lower K. Additionally, there are other processes that can also be affecting K. The leachate, with a high concentration and valence of ions, would decrease the net particle charge (Chorom and Rengasamy, 1995) and thickness of the Diffuse Double Layer (di Emidio et al., 2017; Schmitz, 2006; Stepniewski et al., 2011) in an initial stage. The former is due to the decrease in the dispersion of clays and the latter is relevant for the high porosity of freshly compacted soils. For Ca-clay minerals like here, the maximum dispersion occurs at pH 6.5-7.7 (Chorom and Rengasamy, 1995), which is the pH for most leachates. As a result, the transport of charged species in clays with high plasticity is enhanced, resulting in an increase of K in the first years. This supports earlier observations of an increase of K with the leachate concentration (Mitchell and Soga, 2005). In a later stage the precipitation of mineral phases and the growth of microbial activity may contribute to pore clogging (Francisca and Glatstein, 2010; Stepniewski et al., 2011) and therefore the decrease of K after its maximum during the previous stage (Figure 6). Calcite is likely to precipitate within the liner due to the basic pH, the high leachate bicarbonate concentration and additional dissolved calcium released over time by cation exchange reactions with the liner (de Soto et al., 2012; Thornton et al., 2001).

5. CONCLUSIONS

The performance of four natural clayey substrata as potential landfill liners was assessed by measuring their physico-chemical properties and stability and alterability upon contact with leachate, followed by rainwater. The attenuation of pollutants in leachate depends on the pollutant species and liner mineralogy. Potassium, ammonium, (dissolved) organic compounds and heavy metals (chromium, nickel and zinc) are the most representative leachate pollutants, according to their concentration, toxicity or persistence. All studied clayey materials are useful for the attenuation of leachate pollutants in sustainable waste landfills. These pollutants are mainly attenuated in the clayey materials by anaerobic biodegradation and sorption mechanisms, especially cation exchange. Chloride and sodium in leachate and native cations released from exchange sites on the clay liner after sorption of pollutants can be diluted by groundwater. However, different management options should be applied depending on the clayey material. The LC is the best material based on the sorption capacity and erosion resistance. However, the LC has a large plasticity (high susceptibility to excessive shrinkage) and easily alterable smectite clay minerals that partially collapse to illitic structures. Illitization has less impact on the CEC of the liner than on its chemo-mechanical stability and could be countered by compacting and mixing LC with sands. The OC is also plastic but to a lesser extent, with an acceptable PI. This substratum has a significant sorption capacity and is the best material for buffering acid leachates (due to native calcite) and supporting biodegradation of organic compounds. On the negative side, Coal Measures Clays have the lowest sorption capacity and zero neutralization power. However, they have the lowest plasticity and the most resistant clay minerals (kaolinite accompanied by illite) to alteration by exposure to leachate. In addition, both Coal Measures Clays are easily compacted until negligible air voids, which favours the achievement of a low K. The SCMC contained sulphate-bearing species (resulting from oxidation of pyrite) that enhance the retention by precipitation of heavy metals through bacterial sulphate reduction in the liner. The DCMC had very low mineral phases or inorganic salts that are readily dissolved in water. This is advantageous as it results in less mobilization of leachable salts from the liner itself. The LC and Coal Measures Clays have associated iron/metal oxides and oxyhydroxides that can enhance anion exchange and the removal of metals by sorption. Redox-sensitive species such as pyrite (OC and SCMC) and iron oxides (LC and Coal Measures Clays) can enhance the removal of metals by bacterially-mediated redox transformation and precipitation processes. The presence of pyrite and iron oxides also determines to a large extent the acid-base neutralisation potential, together with native carbonates in OC and bicarbonates in the leachate. After permeation with landfill leachate and rainwater during several weeks (equivalent to years under field conditions), the model liners achieved long-term sustainable low K, that rarely surpassed the maximum value specified for liner design.

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FIRST WORLDWIDE REGULATION ON SUSTAINABLE LANDFILLING: GUIDELINES OF THE LOMBARDY REGION (ITALY)

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Final sink

ABSTRACT
The environmental sustainability principle, since the Rio de Janeiro Conference (1992) and the Kyoto Protocol (1997), has produced a marked change in environmental protection strategies. In waste management practices this trend is reflected in the passage from a linear to a circular approach, where strong attention is paid to the recovery of resources from waste, with a dramatic reduction of untreated waste landfilling. Deposition of waste on soil still plays a crucial role in acting as a final sink for closing materials loop in Circular Economy. Paradoxically the regulations of landfilling at international level appear obsolete, not taking into account the environmental sustainability concept, still promoting unsustainable approaches, with environmental protection measures mainly based on physical barriers, without any consistent control of long term emissions of contaminants which last longer than the barriers themselves. “Guidelines for Sustainable Design and Management of Landfills” issued by the Lombardy Region in 2014 represents the first official regulation which introduced systematically the principle of environmental sustainability. They highlight the modern role of landfilling as a final sink and promote measures and procedures for controlling the mobility of the potential contaminants in the waste, until reaching, within a generation time, a Final Storage Quality in equilibrium with the environment. The aim of this paper is to illustrate and discuss the main aspects introduced by the Guidelines, offering an interesting base for a future spread of the practical application of the sustainability concept to landfilling.

1. INTRODUCTION

1.1 Objective of the Guidelines and organisation

Current legislation on landfilling in Italy (Law n° 36/2003) stems from the European Directive 1999/31/EC. The main drawback of this regulation is that the environmental sustainability concept is not taken into account. This is evident when considering that the main tool to control contaminants emission is the physical barrier (synthetic and mineral liner) which lasts shorter than landfill emissions (Cossu et al., 2020).

As a matter of fact, when considering the origins of the contaminated sites in the Lombardy Region (Italy), a significant number of cases are related to legally authorized landfills, permitted in compliance with the national regulations (Cossu et al, 2017).

Despite the Lombardy Region strongly promoted Circular Economy programmes achieving a dramatic reduction in landfilling (nowadays only 2% of MSW and 18% of Industrial waste are landfilled), this system is still necessary.

Considering the potential risk of contamination posed by traditional “legal” landfilling, particularly during the aftercare phase, and recognizing that most of the available areas suitable for landfilling are situated within environmentally sensitive areas, the main goal of the “Guidelines for Sustainable Design and Management of Landfills” issued by the Lombardy Region was to introduce preventive measures by promoting sustainable landfilling. This means adopting any sort of treatment (before or during landfilling) and carrying out management activities that can control long term emissions and achieve environment equilibrium within a generation time (25-30 years).

Additional aims of the Guidelines were the following:

- to introduce homogeneous procedures in the whole Region, combining and integrating different regional regulations and synergizing experiences gained by all the provinces;
- to better define a number of features of the national regulations which are not sufficiently clear or detailed

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(e.g. leachate recirculation) or which need to be integrated (e.g. additional physical barriers in sensitive areas).

These Guidelines are the result of two years of collaboration between the Lombardy Region, the University of Padua, the twelve Provinces of Lombardy and ARPA (the Regional Agency for Environmental Protection which carries out on-site inspection in the Lombardy Region).

The Guidelines, written in pursuance of the national law, represents a model adopted by an institutional body for issuing regulations by involving the scientific community and discussing them with all interested stakeholders.

The Guidelines have been developed according to the following steps:

- The first draft was a collection of critical elements deriving from the experiences gained by the institutional bodies (Region, Provinces, ARPAL) in implementing the national regulations;
- The second draft was the result of the collaboration with the University of Padua (18 meetings and workshops between 2012 and 2014);
- The draft text was then presented and discussed at the "Sardinia 2013" Symposium;
- Official presentation and discussion in a public meeting with more than 400 participants, held in Milan, Lombardy Region Congress Hall, on 2nd April 2014
- Open consultation of a collection of written comments by the stakeholders.
- Revision of the text following all the acceptable remarks and transmission of the final version of the Guidelines to the Regional Assembly for final approval.

The Guidelines issued in the form of Regional Law (D.g.r. n. X/2461) were published on 7th October 2014 in the Official Bulletin of the Lombardy Region.

For the first time at an international level, the principle of environmental sustainability was systematically introduced in an official regulation. No legislation worldwide, in fact, to our knowledge, considers the sustainability concept in regulating landfilling (Cossu, 2016; Butti et al., 2019).

The role of landfilling as a final sink for closing the materials cycle has been highlighted and measures and procedures for reducing the mobility of the potential contaminants in the waste have been promoted.

1.2 Implementation and legal issues

After the approval, the Guidelines were applied to more than 20 cases, including new landfill sites, the extension of existing ones and the closure of completed landfills with final capping (Cossu et al., 2017).

There have been no problems in applying the technical requirements set by the guidelines, as is the case of the additional confinement barrier, even if this causes a certain loss of storage volume.

The main problem has been that the sustainability principle is still approached more theoretically than practically, without a detailed and consistent planning of all the necessary measures for controlling the long term emissions.

The applicability of the FSQ (Final Storage Quality) value is still uncertain especially when landfilling of special industrial waste is considered.

In December 2015, roughly one year after the approval of the Guidelines, a landfill operator carried out a judicial appeal to the Regional Court of Lombardy Region (TAR-Lombardia) for the cancellation of the guidelines. The Court cancelled the Guidelines (sentence of 17th March 2016, n.522), upholding the action brought by the landfill operator, because only the National Government can legislate on environmental issues, and regional governments cannot make additional laws even if written in accordance with national law. Although the Lombardy Region made an appeal for a revision of the Regional Court decision, in 2017 (9th June, 2017, sentence n. 2790/2017) the final decision of the State Court confirmed the cancellation of the guidelines for the lack of authority of a Regional Government in issuing regulations on Environmental Protection.

Although the compliance is no longer compulsory, the Guidelines, nonetheless, represent a vital technical reference document for landfill designers and public administration officers responsible for permitting.

Excerpts of the Guidelines (highlighted in italics) that concern the most innovative features pertaining to Legislative Decree 36/2003 are reported in the subsequent paragraphs.

2. PRINCIPLES, SCOPE AND WASTE ACCEPTANCE CRITERIA

The Guidelines (GL) of the Lombardy Region on landfills begin by centering on how they fit into the context of regional planning, and on the Circular Economy, "based on integrated management focusing on prevention, preparing for reuse, recycling and recovery. In such a framework, landfill represents the closure of the materials cycle, whereby appropriate techniques and procedures are adopted in order to immobilize elements and substances contained in waste, rendering them harmless and, conversely, to minimise the movement and reactivity of such elements and substances."

2.1 Principles

The "Driving Principles" are the cornerstone of the Guidelines. The convergence of politicians, administrators, technicians and citizens on these principles represents an unequivocal basis for the development of regulations directed at promoting environmental sustainability.

- Landfills must be designed, built and managed in compliance with the principle of environmental sustainability, by achieving the Final Storage Quality (FSQ) of the landfill (concerning emissions and biological and mechanical stability of waste) in equilibrium with the environment, within a generation time of no more than 30 years from the closure of the landfill or active sectors.
- In order to ensure environmental sustainability, the Final Storage Quality (FSQ) to be achieved during the period of post-operational management, is indicated by the target values (Annex B of the guidelines), which

project, operational and post-operational management must pursue.
- To attain environmental sustainability, waste to be landfilled should be pre-treated to reduce in advance the mobility of the elements and substances contained therein. In situ treatment or the combination of both in situ- and ex situ-treatment can be alternatively applied to reach the same objective.
- During the planning stage, the methodologies and technologies required to realize environmental sustainability must be justified, described, sized and computed in detail.
- Landfills can be designed to receive homogeneous fractions of waste in order to facilitate possible subsequent landfill mining.
- Landfill must not represent the final use of the site. instead, it should be designed to achieve environmental reclamation with a specific intended use, in compliance with broader local land planning.
- The work on local infrastructure and other work required to achieve the desired use must be specifically planned, and carried out coherently with the various construction stages of the landfill.
- Landfill designs that do not meet environmental sustainability criteria are no longer acceptable.

2.2 Scope of Application

The Guidelines have been issued using the same scope of application and in compliance with the national law (Legislative Decree 36/03), establishing the same minimum requirements for planning, authorisation, construction, operational and post-operational management.

2.3 Waste types and admissibility

Waste types that are not admitted to landfill (WEEE, packaging, wastes from separate collection, etc.) as well as the conditions and the quantities for the acceptance for other specific wastes, (eg. Waste mixtures) fixed by the guidelines are consistent with Italian and EU regulations as described in Annex A of the guidelines (Admissibility Criteria for Waste in Landfills).

3. WASTE TREATMENT

3.1 Objectives

"The treatment of waste before landfilling (ex situ) and, if necessary, during landfilling (in situ) should be carried out to reduce the environmental impact in the short and medium-long term, in accordance with the principle of environmental sustainability.

The choice of the type of treatment must be justified and differentiated according to various aspects: goals, process type, technology and level of advancement, references, expected results. Waste treatment can combine ex situ and in situ treatment".

This is a very important point in the Guidelines as the concept of environmental sustainability should be perceived in terms of consistent planning activity, and not simply be interpreted as a couple of fancy words on paper!

3.2 Ex situ treatments

The following are examples of ex-situ pre-treatment proposed by the guidelines, depending on the type of waste:
- manual and/or mechanical selection and recovery of fractions with economic value;
- mechanical-biological treatment;
- chemical and/or chemical-physical treatment;
- thermal treatment;
- washing;
- specific treatment options as a combination of the above.

3.3 In situ treatment

In situ treatment might occur at different stages of landfill life, both during the operational phase and after closure. The following different typologies of in situ treatment are mentioned in the Guidelines:

- aerobic processing of the landfill, by using natural (semi- aerobic) or mechanical (in situ aeration) systems, possibly following a period of anaerobic processing with landfill gas generation;
- natural (open landfills/flushing) or mechanized (water infiltration/leachate recirculation) washing out;
- combined aerobic-anaerobic systems, etc.

These indications are quite important as they reflect the most advanced scientific and technical developments taking place in recent years, which have often been neglected, if not contradicted, by local Authorities.

4. DESIGNING AND PLANNING

The Guidelines stipulate that all planning and project actions and the location of the landfill site must comply not only with the provisions of current legislation, but also with the terms already present in the Regional and Provincial Plans of specific reference.

4.1 Project documentation

The accompanying documentation to the project (i.e., administrative documents, technical drawings; an environmental compatibility study; technical, hydrogeological/geomorphological reports; operational and post-operational management; surveillance, control and financial plans; a restoration report, as well as environmental and landscape design documents) is listed, with detailed content.

The Technical Report in particular must fulfill the following requirements:

- Describe and design in detail all the planned treatments of waste prior to, during or after the landfill disposal; describe, on the basis of literature, the expected results of the proposed processing;
- Define and estimate the forecast trends of emissions quality over time, focusing on sustainability targets;
- Describe the technical characteristics of the collection and disposal system for wastewater and rainwater, and the discharge point (e.g. sewers, surface water runoff, collection system etc.);
- Assess the mechanical stability of the embankments,

the working face, the cover and the internal embankments, taking into account a possible earthquake;
- Assess the carrying capacity of the bottom of the landfill (including details of the calculation methods) and calculate the forecast subsidence in order to assure the proper functioning of the leachate collection and of the drainage system in maintaining the gradient of the bottom;
- Calculate the total capacity of the landfill, with reference to the forecast degree (%) of waste compacting and planned use of various sectors;
- Adopted criteria for the allocation in landfill of the received waste;
- Calculate the potential surface runoff and leachate production compared to the time needed to construct the surface cover system, and the size of the leachate collection systems and storage tanks;
- Assess the functioning of the leachate collection wells and of the possible liquid drained between different liners (control methods, measuring and production, pump characteristics);
- Include information on the recirculation and/or other treatment systems in situ and/or ex situ for the treatment of leachate, describing the adopted criteria and proposed targets, and referring to detailed maps and project drawings/charts;
- Include information on systems for monitoring and ensuring the long-term functioning of leachate collection, drainage and storage systems;

In addition, when leachate recirculation is planned it has to be well motivated. The technical aspects have to be appropriately engineered by taking into account the following requirements:

a) Definition of the optimal waste moisture content to be reached and maintained through recirculation. Calculation of the required recirculated volumes and comparison with the estimated leachate volumes being produced and stored;
b) Forecast of the maximum biogas production during recirculation and control of the efficiency of collection, extraction and energy recovery facilities;
c) Assessment and monitoring of the efficiency of the barrier system and of the leachate collection and extraction system in order to ensure the minimum hydraulic head on the bottom, at a level compatible with the pumping and extraction systems;
d) Description of any pre-treatment of leachate deemed necessary to optimise the process;
e) Description of the planned recirculation system (measurement and transfer system from the tanks to the landfill, sizing and positioning of piping system) with detailed descriptive maps/plans
f) Details of the planned additional monitoring required, on top of what has already been authorised, subsequent to the start of recirculation.

The Guidelines enable the use of waste materials for constructing landfills under the following conditions:

a) Total quantities for which authorisation is being sought, expressing them both in m^3 and the maximum daily total in tons;
b) Results of the tests and analyses proving compliance with the technical and environmental characteristics indicated in Annex D (Use of Waste in Landfill Construction), and a statement of compliance with those conditions;
c) EWC codes, description chemical-physical quality and composition of the waste, its origins and any treatment required to ensure technical suitability for recovery.

Finally, the report on environmental and landscape restoration and adjustment must contain:

a) Overview of the area and surrounding zones regarding the morphology, geomorphology, geology, hydrogeology, climate, soil use, surface hydrogeology, woods, vegetation, management of farming and animals, history and existing facilities;
b) Landscape and environmental quality analysis, maps and, possibly, an analysis of any human settlement and infrastructure;
c) Targets and restrictions on environmental and landscape recovery, considering the timeframe and recovery methods involved;
d) Intended use and final layout of the area, with the study of morphological trends, the layout of the surface area and the relations to the surroundings. Illustration of the work to be done, justification for the chosen solution, resolution of problems tied to the feasibility analysis, including alternatives. In the case of work on a limited area, the report must indicate the architectural details;
e) Planning of the green areas, including indications of trees and bushes to be used and their related agronomic characteristics, as well as broad details about the need for water and maintenance of such plants. The project must illustrate the timing of the interventions in the different phases of the landfill, up to the final restoration.

It is clear that these indications add substance to the principle that the landfill must not in itself constitute a destination of use, but rather represents a viable tool for building and developing destinations of use. In other words, the volume occupied by the waste can represent a precious resource for planning interventions in the area.

Essentially, it is not necessary to design landfills but parks, golf courses, recreation centers, etc., to be built using the waste, and inserting these uses into the territorial planning of a given area.

With these Guidelines "environmental restoration and adjustment" cease to be, in the design documentation, a useless design exercise or an unrealistic pipe dream, but, rather, become an integral part of the whole project.

5. BASE BARRIER SYSTEM

The Guidelines shed light on the fact that the base barrier system (lining and drainage) refers to both bottom and side slopes, without any structural differentiation in mineral layers. Specific solutions for the drainage system on side slopes might be accepted in particular cases.

5.1 Background for the containment system

The background for the containment barrier - i.e. artificially created low permeability mineral layers should respect the following criteria:

- Control of bearing capability (for the Plate Bearing Test - Swiss Method - the value of the Modulus of Deformation on a 30 cm plate must be equal to or greater than 50 N/mm^2 , calculated with the pressure in the range between 0.15 and 0.25 MPa in the first loading cycle);
- Slopes necessary to guarantee the drainage of the leachate that take into account the expected settlement of the bottom (minimum transversal slope of at least 2% and a longitudinal slope of at least 1.5%);
- For confined aquifers, the distance between the top of the aquifer and the level of the base barrier, or the formation level for any structure built into the ground to contain leachate must be at least 1.5 m. For unconfined aquifers, this same distance must be 2 m starting from the maximum level of the aquifer, except for inert landfills where the law sets this distance at 1.5 m).

Referring to the last point, having specified the definition of confined aquifer and groundwater, it is still important to foresee the construction of systems (which will remain effective over time) to capture and remove any water that may come into contact with the waste.

The Guidelines also set out criteria for the construction of artificial, not saturated layers and for the evaluation and control of inhomogeneous conditions in the background soil.

5.2 Barriers

This is an important and highly sensitive point of the Guidelines which specify the requirements defined by national legislation that have often been the subject of controversy and misinterpretation.

Specifically, where the natural geological barrier established by Legislative Decree no. 36/03 is not existing (e.g. in a permeable area), then an equivalent artificial confinement barrier should be installed. The calculation of the equivalence should refer to the time criterion (based on Darcy's law), which is calculated as the ratio between the thickness of the layer and the permeability of the material that the layer must uniformly be made of.

In addition to the barrier indicated above, for landfills of hazardous and non- hazardous waste, a second and additional confinement barrier is required. According to the above-mentioned criteria the required structures for the different landfill typologies are described in Figure 1.

6. DRAINAGE AND LEACHATE COLLECTION SYSTEM

In the structuring of a landfill, the drainage system is one of the most delicate points and yet one of those to which little attention is paid, both at a technical and regulatory level. In addition to recalling the role of the drainage system to "favor the most rapid removal of the leachate", the Guidelines outline the strategic objectives to be observed in its design, namely:

a) to prevent leachate accumulation in the landfill, keeping to a minimum the water head within the waste mass.
b) to reduce the clogging phenomena
c) to allow easy video inspections of the system.

Point a), as observed earlier, has a similar importance to the base barrier: a high leachate head, according to Darcy's law, is equivalent to a higher barrier permeability. Point b) brings into play the quality of the leachate, which depends on the management methods and on the quality

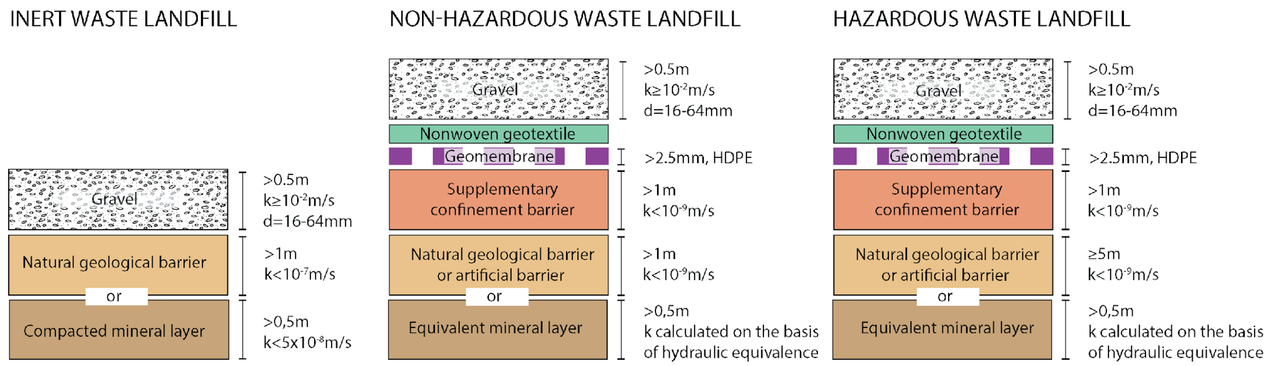

FIGURE 1: Required structures for the different landfill typologies according to the Lombardy region Guidelines. The compacted and equivalent mineral layers have to be artificially made of material from classes A6 and A7 in the HRB AASHTO classification system. The gravel around the drainage piping must be classes A1 and A3 of the HRB AASHTO classification system. It should be a washed aggregate with the CE mark (indicatively, gravel with pieces 16- 64 mm) with low carbonate content (< 35 %), <3% passing through ASTM 200; with uniform size distribution, a flattening coefficient < 20 (UNI EN 933-3 standard) and minimum diameter d > 4 times the size of the openings of the drainage piping; minimum height of 0.5 m above the crown of the perforated pipe and no less than 2 m wide at the base. Supplementary confinement barriers have to be made of materials from classes A6 and A7 in the HRB AASHTO classification system; HDPE geomembrane (artificial sealing liner), thickness ≥ 2.5 mm, conforming to the UNI 11309 standard for smooth geomembranes and the UNI 11498 standard for improved geosynthetic barriers; nonwoven geotextiles (minimum resistance to traction in the two longitudinal and transversal directions: 60 kN/m – UNI EN ISO 10319 standard; minimum static puncture resistance: 10 kN – UNI EN ISO 12236 standard; minimum mass per unit area: 1200 g/m^2 - UNI EN 9864 standard) or other suitable protection for the geomembrane.

of the waste, while point c) points to the need for a design that does not interfere with the inspection (and reparability) of the system through external interventions (video cameras, descaling tools, washing etc.).

In relation to point b), in order to avoid blockages, the Guidelines stipulate that "No synthetic and/or natural materials, acting as filters, should be placed between the drainage system and the waste if their hydraulic conductivity and porosity levels are below that of the drainage layer".

6.1 Primary and Secondary drainage pipelines

The Guidelines dictate, for the main perforated drainage pipelines, minimum nominal diameters that are governed not by hydraulic calculations, but by the dimensions of the cameras and inspection and control equipment that may need to be inserted from the outside. As the length to be inspected increases, the overall dimensions of the inspection equipment also increases and consequently the diameter of the pipe must be modified to better adjust to the variation.

The values for the main parameters to be considered when resizing the drainage and collection pipes are given in Table 1.

Further important design measures to be taken are as follows:

- Where the site morphology makes it possible, inspection wells for the drainage pipes must be placed upgradient and downgradient from the collectors and/or outside of the landfill body.
- The secondary drainage pipes must not be connected to the primary collectors so as to not compromise the stability and ability to inspect the latter. Since such piping is not watertight, but offers a preferential drainage route, the pipes do not need to be connected, especially as a connection point might impede inspection or else break due to settling.
- The slope values of the pipelines must refer to the operating conditions,following the complete settlement of the bottom of the landfill.
- The perforations in the pipes must be no smaller than 10 mm to prevent clogging.
- The drainage piping system must be placed on the geomembrane protection layer and must be tested for mechanical, thermal, chemical and biological stability under the forecast load and operational conditions.
- The bearing capacity of drainage pipes must be determined in a specific calculation taking into account the pressure of the waste column and the pipe covering layer, at the same time taking into account the physical, mechanical and chemical actions.

TABLE 1: Values of the main parameters for dimensioning primary and secondary pipes in the drainage systems.

Drainage pipes	Minimum diameter (mm)	Minimum density (m/ha)	Pipes distance (m)
Primary	315	170	60
Secondary	200	500	20

6.2 Leachate collection wells

The Guidelines, in order to avoid any failure or malfunctioning in leachate collection wells, prescribe the following:

- Leachate collection wells must allow easy inspection of the primary leachate drainage collectors.
- Leachate collection wells within the landfill body should be placed preferably on the slopes, with the exclusion of those special cases where there is a need to use vertical wells. For wells located outside the landfill, the isolation system of the well, of the connection to the landfill and of any crossing system of the bottom barrier must meet the equivalent conditions for tightness (sealing) and protection as regards the impermeabilisation system.
- The leachate extraction wells must be designed in order to avoid the potential accumulation of any biogas produced by the landfill.
- The opening size of the leachate collection well must be sufficient to allow inspection and an easy removal of the pumping system for maintenance.

6.3 Leachate extraction and hydraulic head

For the functional requirements of the leachate drainage and collection system mentioned above, adequate measures must be taken as follows:

- The system must always be designed so that the pumps start automatically as soon as the lowest technically possible levels of hydraulic head are reached;
- The well must be created so that the bottom is below the level of the drainage network to ensure the proper removal of the leachate;
- The waste mass must have adequate drainage capability or specific facilities should be installed for extracting leachate;
- The waste storage areas must be divided into hydraulically separate basins, with areas of roughly 10,000 m^2, measured on the bottom.
- The pumping system must be dimensioned taking into account the surface of the given sector at the start of waste deposing activities; it must ensure the emptying of the sector within 48 hours of the onset of an extreme rainfall event (return period of 10 years and a duration of 48 hours).

6.4 Leachate Storage

The leachate storage system must be designed to store the maximum generated amount, calculated by taking into account, as an input datum, the rainfall value in the conditions referred to above - i.e. an extreme rainfall event, lasting 48 hours, with a return period of 10 years. The system must also have a reserve volume of 10% of the total capacity.

The structure of the storage tanks and the control system should respect the technical requirements set out in a specific national regulation (D.d.g. 07/01/1998 n. 36).

7. TOP COVER

Having reviewed the main functions required of a waste covering system in a landfill (seclusion of waste from the top ambient, control of water infiltration into the waste and control of biogas fugitive emissions), a subdivision of the feasible types of covering is proposed (daily, temporary, permanent), which has multiple design and management implications, to be reviewed later.

7.1 Daily Covers

The Guidelines provide specific technical requirements for the materials to be used for daily covers. Specifically, hydraulic permeability should be a constant $k>10^{-3}$ m/s. These requirements should also be the technical reference for the acceptability of waste derived materials in daily covering according to the requirements often presented by landfill operators. Additional requirements are the following:

- Quality compatibility with the given landfill type and related admissibility criteria for the permitted waste;
- The amount should be considered within the permitted waste volume;
- Waste delivery should use the permitted disposal methods (D1/D5).

7.2 Temporary Covers

Prior to the installation of the final top cover, the Guidelines, in line with the national regulations, allow for a temporary cover for the time required to reach the mechanical and biological stability indicated by the project. Compared to the current legislation which considers only mechanical stability ("settlement"), the Guidelines also include biological stability, on which, in the case of urban waste, the settlements themselves depend. A final cover installed before the waste is reasonably biologically stabilized could slow down its degradation and prolong contaminant emissions over time. From a design point of view, the Guidelines clearly state that the structuring of the surface cover must be compatible with the type of landfill and that, in addition to it being well described and sized in the technical documentation, it "must always, keep the waste separate from the surrounding environment (allowing for the movement of gas and/or liquids, as outlined in the project), guarantee proper runoff of surface water and ensure a balanced landscape integration of the landfill, taking into account the duration for which the temporary cover will be in place."

7.3 Final Capping: General Technical Specification

For the final covering, the Guidelines refer to the requirements of the national legislation as regards its role and its structure, in accordance with the different types of landfill. The Guidelines then go on to highlight how the multi-layer structure, regardless of the landfill type, must include geotextiles for separating materials with different grain sizes (Figure 2). In order to drain precipitations, the Guidelines prescribe a minimum slope of the top cover, once settling is expired ("less than 3% for inert landfills and 5% for other landfill types, in a year").

8. LEACHATE AND RAINWATER MANAGEMENT

8.1 Rainwater management

The different types of rainwater that fall on the functional areas of the landfill are considered in the Guidelines (surface cover, roads, parking lots, etc.) which together with the wastewater produced by different services (tyre washing, toilettes, etc.) must be properly managed (drained, collected, treated, recycled, etc.). Here, an important aspect needs to be communicated in big, clear lettering: "Infiltration of precipitation in a landfill might, depending on the waste type, be necessary to improve biological stabilisation and waste leaching." This should be considered as a "leit-motiv" in the design of sustainable landfills, because waste needs water to stabilize and therefore landfills cannot be sealed, thus preventing water from entering the waste body.

8.2 Leachate Management

The quantity of leachate foreseen in the various life stages (operational and post-operational) of the landfill must be calculated meticulously by identifying and considering all the parameters that can influence the hydrological balance in the given area and the design solutions

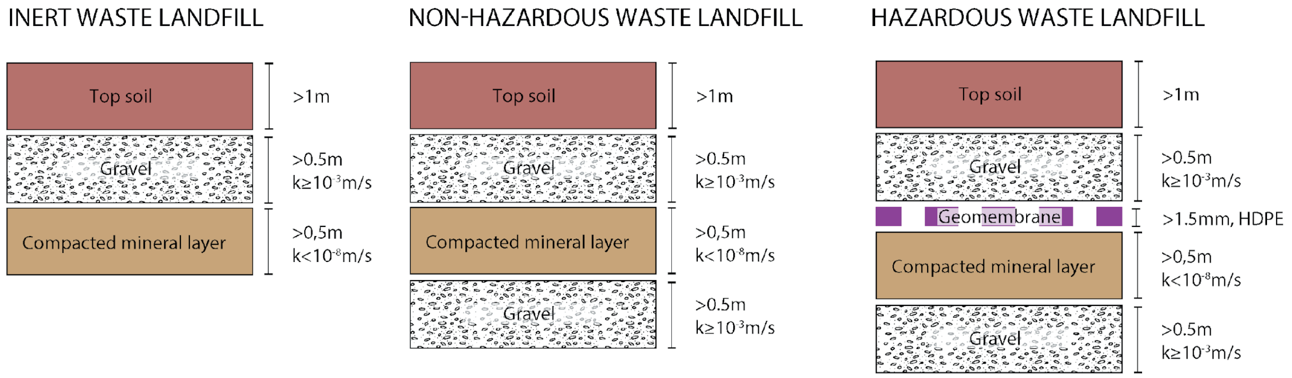

FIGURE 2: Final top cover for Inert, non-hazardous and hazardous waste landfills. The water and gas gravel drainage layer have to be made up by aggregate with the CE Mark, belonging to classes A1 and A3 in the HRB AASHTO classification system; the HDPE geomembrane has to conform to the UNI 11309 standard for smooth geomembranes and the UNI 11498 standard for improved geosynthetic barriers.

adopted (meteo-climatic characteristics, characteristics of the waste to be deposited, degree of compaction, type of covering, materials used, slopes, surfaces used, storage activities in place, methods for removing surface water, characteristics of a possible greening, etc.).

The Guidelines state that:

- During all stages of the landfill it is important to detect and record the main meteorological-climatic parameters and to verify the estimates of quantity and quality of the leachate produced as the influencing factors vary.
- During the planning stage, final decisions must be made on how the leachate will be managed and how on-site operations will be defined
- The identification of the treatment system must take into account the changes over time in the quality of the leachate, adopting the best techniques available.
- The management of the residues from leachate treatment (sludge, condensates, spent activated carbon, etc.) must be defined. These residues can only be placed in the landfill if they meet the admissibility criteria for waste entering the landfill. This will prevent the accumulation in the landfill of mobile elements and substances that are not degradable, which would otherwise contradict the principle of environmental sustainability.

8.3 Leachate Recirculation

This is one of the points that, as already mentioned, has always been the subject of controversy and has resulted in uneven management practices between one Region and another. Undoubtedly, the Guidelines show that recirculation is feasible and permissible only if it is "functional to achieving specific operational targets that have been consistently described in the project" In other words, leachate recirculation must be a technical tool available to the designer to make the landfill more sustainable according to the "Bioreactor landfill" model, and not an expedient to avoid the works necessary for the treatment of the leachate. The Guidelines also contain a series of technical recommendations as follows:

- Recirculation must be compatible with rainfall levels.
- Leachate recirculation must be performed in such a way that uniform distribution can be guaranteed, thus avoiding the formation of aerosols and odours, perched aquifers or preferential infiltration paths.
- The techniques used to distribute the leachate must be defined in the project on the basis of the recirculation targets, taking into account the quality of the leachate and the quantity.
- Leachate recirculation can only commence in the sector of the landfill where waste delivery has been completed. Such recycling is only feasible during the waste delivery stage in specific, suitably justified cases (e.g. to prevent dust spreading)
- Leachate can be recycled untreated or following treatment, depending on the defined targets.
- In traditional anaerobic landfills, recirculation can only commence once the biogas collection system has begun operating.
- Leachate recirculation must be planned and managed so that it does not interfere with gas circulation in the landfill.
- Where a landfill has already been closed, the commencement of leachate recirculation must be carried out so that the cover can be restored without compromising its characteristics.
- Recirculation can only be achieved if the bottom barrier can adequately safeguard the groundwater.
- Leachate injection systems must be placed under the cover to avoid atmospheric emissions, especially bad smells, and to optimise the distribution of leachate in the waste mass.
- The safeguards for the leachate injection system must include a system to monitor and regulate the pressure so that the leachate flows under atmospheric pressure.
- The competent authority can, based on monitoring and specific checks, require that changes be made to recirculation or that additional precautions/measures be adopted.

In addition to the above, the Guidelines set out further requirements to be observed during the operational phase which recognize the importance of:

- Measuring flow rate to determine the volumes of leachate, separating recirculated leachate from disposed leachate;
- Recognizing that only the leachate from the landfill itself can be recirculated;
- Halting recirculation in case of malfunctioning or problems in the bottom barrier system (impermeabilisation and drainage) or in biogas extraction system. The recirculation of the leachate must be halted as a precaution and can only recommence once clearance has been obtained from the competent authority

The Guidelines also provide information about authorization procedures to be adopted in the event that the request for recirculation occurs after the approval and authorization of the landfill.

9. BIOGAS MANAGEMENT

As regards the management of landfill gases, the Guidelines clearly specify that the main objective is to minimize the environmental problems that the gas can cause (emissions of greenhouse gases, volatile contaminants or odorous substances). They also suggest that only if there are the right conditions and if economically viable, the gas can be exploited energetically.

Therefore, a very important aspect is sanctioned from the point of view of environmental sustainability, namely that the management of the landfill "must prioritise acceleration in the production of biogas so as to reduce the potential organic matter content of the leachate".

Following conventional indications on the sizing of the biogas management system in line with the landfill cultivation methods, and on structuring the collection and transport network, the Guidelines suggest that the gas not being exploited energetically can be oxidised not only in flares or external biofilters but also, when below a production equal to 0.001 $Nm^3/m^2/h$, can be treated in "bio-oxidation covers that must be appropriately sized and designed ". To support this choice, two citations (Rachor et al., 2011; Pedersen et al., 2011) from the literature have been included in the Guidelines referring to experiences where a high oxidation of methane has been achieved with flow rates five times higher than the one indicated above.

10. USE OF WASTE IN LANDFILL CONSTRUCTION

The use of waste materials in landfill construction is one of the most salient points of the Guidelines. In the past, it was left to the scrutiny of the Authoritative Bodies to assess this possibility. Precise indications are given on the possible use of waste materials in the construction of landfills, in terms of admissibility, quality requirements, and consistency of performance over time. These indications are accompanied by detailed technical specifications relating to the different works of the landfill (Annex D - Use of waste for the construction of landfills) and administrative procedures (Annex E).

The main criteria to be observed are the following:

- Waste can be used as the sole material or mixed with other kinds of waste or raw materials. Where mixed waste is used, the environmental protection characteristics and compliance with the definition of "inert waste" must be verified for each type of waste employed, while the geotechnical characteristics must be verified on the mixed material prior to use in the construction of the landfill.
- Settlement forecasting of the material under the predicted loads on the layer must be assessed to guarantee the minimum thickness of the layer and its geotechnical characteristics.
- Permeability must be assessed, under the forecast load after completed closure of the landfill.The geotechnical characteristics must be verified under the most stringent operating conditions.
- The layers must be assessed as part of inspection procedures in compliance with the national regulations.
- The use of waste to form the bottom barrier is not allowed to intrinsically ensure optimal functionality of this key component of a landfill and to be certain of the constant quality of the material used.
- End-of-life tyres can be used as engineering materials.
- The use of inert waste in the construction of landfills is not subject to the specific regulations for inert landfills as already set out in the national regulations "This decree does not apply: (...) to the use of inert waste for development or reconstruction purposes in landfills".
- Recovery activities inherent in waste management must be explicitly authorized in compliance with national regulations.
- The European directive 2008/98/EC classifies the recovery operations into 13 categories from R1 to R13; R5 represents the recycling/reclamation of inorganic materials, other than metals, including soil cleaning resulting in recovery of the soil and recycling of inorganic construction materials. Recovery of waste materials in the construction of landfills must be considered within the framework of R5 mining operations, as it involves the use of waste to replace raw materials (sand and gravel, clay, earth). Considering that the recovered waste is used as a substitute for raw materials, the admissible quantities must be consistent with what is technically necessary for the construction and efficiency of the layer, within the volumes already foreseen by the permitted project.
- The use of such waste is not subject to the payment of the "landfill tax" as it is an R5 recovery operation which is exempt from the application of Legislative Decree 36/2003, unlike the waste used in the layers of landfill cover which is still subject to the "landfill tax".

11. OTHER DESIGN FEATURES

Other design features that are considered in the Guidelines are:

- Staff Organisation, with an indication of professional profiles and roles, opening hours of the plants, etc..
- Infrastructure (Entry control systems, Security systems, internal and external access routes with relevant signposting, Weighing point, Wheel cleaning point, Devices (potentially not fixed) to measure radiation levels of incoming waste, Fire-safety systems, Staff block, including change rooms, Vehicle storage area, autonomous generator capable of supporting all landfill users in the event of a power failure of the central electrical unit);
- Monitoring and control of groundwater quality (preliminary investigations for the authorization, organization and structuring of the groundwater control network, construction characteristics of sampling piezometers, frequency of checks, etc.)
- Biogas monitoring network (possible installation of spy wells to control any leakage of biogas outside, groundwater monitoring wells, measures for emergency intervention measures in the electrical substation,);
- Compliance verification tests.

12. CLOSURE PROCEDURES, ENVIRONMENTAL RECOVERY AND RELEASE OF THE FINANCIAL GUARANTEE RELATING TO OPERATIONAL MANAGEMENT

In addition to the procedures for the closure of the landfill (essentially based on the stabilization of settlements), the Guidelines also indicate a list of minimal checks, to positively assess the release of the financial guarantee relating to post-operational management (Article 14, paragraph 3, Legislative decree 36/03).

a) Positive assessment of the environmental recovery work in the area, as per the approved project;
b) Absence of subsidence, fracture or depressions in the cover.
c) Absence of contamination in the groundwater due to the landfill.

d) Leachate levels in the waste, in the leachate extraction wells and (where applicable) in biogas extraction wells, or in any other wells built no higher than the draft heads of the extraction systems;
e) Properly functioning leachate collection and storage system;
f) Properly functioning biogas extraction and treatment system (where applicable);
g) Properly functioning safeguards (piezometers, fencing, gate, masking);
h) Properly functioning systems/devices (irrigation, fire prevention, road network).

13. FINAL STORAGE QUALITY (FSQ)

The most important point of the Guidelines regarding environmental sustainability is the following: "The design and operation of the landfill must be done taking into account the FSQ, as per the expected timetable indicated in the project, which cannot be more than 30 years". Therefore, within the thirty- year timespan, not only financial provisions are forecasted (as set out in the current legislation), but more importantly, the drafting of concrete objectives according to which the designer must guarantee timely and congruous interventions are made clear.

Once the stability of the settlements and the presence of suitable slopes to guarantee the surface runoff of rainwater have been verified, the Guidelines then consolidate the concept of the Final Quality of the Landfill by identifying the target values "which the project, operational management and post-operational management must pursue". These values are shown in a Table inserted in the Guidelines text as Annex B (FSQ target values), reproduced here in full (Table 2).

14. TECHNICAL AND FINANCIAL REPORTS AND RELATIONSHIPS

The Guidelines also provide a series of clarifications aimed at resolving some non-univocal interpretations relating to a number of critical aspects of the contents of the Plans according to Legislative Decree 36/03, listed below:

- Operational Management Plan
- Post-Operational Management Plan
- Financial Plan
- Environmental Recovery Plan
- Monitoring and Control Plan
- Annual Report

The Annual Report "must specifically indicate if the planning forecasts for the various parameters leading to the FSQ are accurate or not, and provide details of any corrective measures".

The same report should also contain the following information:

- quantity of leachate recirculated and sent for disposal, compared to the estimated volumes indicated in the permitted technical documentation;
- assessments of the effects of recirculation on the quantities and characteristics of biogas, characteristics of the leachate, settlement of the waste;
- estimated water balance - assessing rainfall infiltration, waste moisture, quantity of leachate produced by the degradation of waste and recirculation, evaporation, leachate sent for disposal - that verifies the absence of significant infiltrations of leachate into the subsoil and the efficacy of the collection and transportation system.

TABLE 2: Target values for the parameters describing the Final Storage Quality (FSQ) in a landfill, Annex B of Lombardy Region Guidelines

Matrix	Parameters	Target values (mg/L)
Leachate	COD	1500
	BOD_5/COD	0,1 (adim)
	Ammoniacal-N	50
	Al	1
	As	0.5
	B	2
	Cd	0.02
	Cr	2
	Cr VI	0.2
	Cu	1
	Fe	2
	Hg	0.005
	Mn	2
	Ni	2
	Pb	0.2
	Se	0.03
	Sn	10
	Zn	3
	CN^-	0.5
	SO_4^{--}	1000
	SO_3^-	1
	F^-	6
	Nitric-N	20
	Total Hydrocarbons	5
	Phenols	0.5
	Aromatic organic solvents	0.2
	Nitrogenous organic solvents	0.1
	Organophosphate pesticides	0.1
	Total pesticides (excluding Organophosphate)	0.05
	Chlorinated solvents	1
Biogas	Surface emissions with extraction plant off	0,5 Nl $CH_4/(m^2 h)$
Solids (alternative methods)	RI_4 (Respirometric index after 4 days)	2 (mg O_2/gST) in 4 d
	IRD (Dynamic Respirometric index)	100 mgO_2/kgSV/h
	BP_{21} (Biogas production in 21 days)	5 (Nl/kgST) in 21 d

15. SPECIFIC INDICATIONS FOR MONO-WASTE LANDFILLS OF CEMENT-ASBESTOS

Finally, the Lombardy region Guidelines, in compliance with the national regulation (D.M. 27 September, 2010) provide minimal requirements for the construction and operation of landfills for mono-landfilling of asbestos-containing waste. The requirements are to deal with the quality of the acceptable materials, the structure of the barrier systems, biogas and leachate management, gate control and quality inspection, emergency procedures, sampling, depositing procedures, daily, temporary and final cover of the deposited waste, etc.).

16. CONCLUSIONS

Whilst all European Union countries are heavily involved in promoting the Circular economy, the "nasty and unsightly landfills" continue to play the fundamental role of closing the materials cycle in waste management. While it is well documented that landfills could be a source of a wide range of environmental problems (e.g., groundwater pollution, odors, greenhouse gases), the national landfill regulations provide technical requirements only for the construction of "traditional" landfill sites, completely overlooking any accomplishments provided by the impressive scientific and technical developments that have taken place in the context of sustainable landfilling in recent years (Cossu, 2016).

Interestingly, in the European Landfill Directive (EU, 1999), which is the mother of all the Member Countries national regulations, the issue of the long term emissions control in the post-operational phase is addressed only in economic terms (art. 8 and art. 10). The Directive sets out for landfill managers a financial provision relating to the covering of after-care costs for a period of at least 30 years. It then decrees, in art. 13 that "…(c) after a landfill has been definitively closed, the operator shall be responsible for its maintenance, monitoring and control in the after-care phase for as long as may be required by the competent authority, taking into account the time during which the landfill could present hazards… (d) for as long as the competent authority considers that a landfill is likely to cause a hazard to the environment and without prejudice to any Community or national legislation as regards liability of the waste holder, the operator of the site shall be responsible for monitoring and analyzing landfill gas and leachate from the site and the groundwater regime in the vicinity…"

As no environmental criteria have been established to determine when "a landfill is likely to cause hazards," the input of data during the planning and design stage relating to how to achieve the aims of Final Storage Quality has not been considered. We all know that the long-term impact of traditional landfilling (the conceptual landfill referred to in the regulation) far exceeds the 30 years covered by the financial provision and, even worse, far exceeds the lifespan of the physical barriers. As a result, the designing of landfills that comply solely with the European Directive might generate contaminated sites, as observed in the Lombardy Region experience.

Despite some controversial legal issues, the history and the technical value of the Lombardy Region Guidelines show that regulations aimed at sustainable landfilling are possible and could be fundamental in turning the general public's negative attitude towards landfilling into a responsible acceptance, considering the unavoidable role of landfilling in any modern waste management strategy.

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CONSTRUCTION WASTE MANAGEMENT PERFORMANCE IN GREEN BUILDING: CONTEXTUALISING LEED IN CHINA

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ABSTRACT

Construction waste issues have raised considerable concern in recent decades. Green building (GB) has been adopted around the globe as a strategy to curtail building-related environmental issues, including construction waste. Particularly in China, with the soaring construction activities tied to urbanization and urban regeneration, massive construction waste has been generated, imposing tremendous pressure on the industry and beyond. China is also vigorously pursuing a national GB strategy, but its effects on construction waste management (CWM) are yet to be confirmed. This paper evaluates CWM performance in GB by putting the dyads into China's particular Political, Economic, Social, and Technical (PEST) context. By analysing a total of 310 LEED (Leadership in Energy and Environmental Design) accredited GB projects in China, it is surprisingly discovered that GB does not prominently improve CWM. The paper goes further to understand the causes of the mediocre CWM performance, by conducting ten semi-structured interviews with GB and CWM practitioners in China. Finally, a comprehensive PEST analysis is conducted to discuss the situation in the context of China. Factors such as (a) incomplete CWM regulations in China, (b) lack of economic incentives, (c) lacklustre awareness about CWM, and (d) lack of advanced technologies, caused the CWM performance in GB. Based on the PEST analyses, some targeted strategies are also recommended. This study is of benefits to both researchers and practitioners in the GB industry.

1. INTRODUCTION

Construction plays a pivotal role in materializing the built environment and maintaining the national economy in China, revealed from its prodigious portion contributing to its Gross Domestic Product (GDP) with a contribution of consistently over 6% in recent years (NBS, 2019). However, construction activity by nature is hostile to the natural environment. It can lead to a series of negative impacts, including land depletion and deterioration, energy consumption, solid waste generation, dust and gas emissions, noise pollution, and consumption of non-renewable natural resources (Shen et al., 2007; Lu and Yuan, 2011; Yang et al., 2019; Yang et al., 2020), which have been frequently witnessed in the past decades in China. Amongst the contributions leading to environmental degradation, the solid waste generated from construction activities has plagued China for decades.

The solid waste arising from construction, renovation, and demolition activities is called construction waste, or sometimes, called construction and demolition (C&D) waste (Kofoworola and Gheewala, 2009; Wu et al., 2017). In recent years, a considerable amount of construction waste has been generated in China due to the mounting construction activities tied to urbanization and urban regeneration. C&D waste has incurred negative social-economic and environmental effects (Ye et al., 2012; Wu et al., 2019). The world generates over 10 billion tons of construction waste every year and about one quarter of the total waste is generated in China, reaching up to 2.3 billion tons (Lu et al., 2016; Ajayi et al., 2016; Zheng et al., 2017). Lu et al. (2016) estimated that approximately 1.13 billion tons of C&D materials were generated in China during 2014. Conventionally, the disposal methods for construction waste are landfill and incineration, which not only considerably occupy valuable land resource but also causes environmental deterioration. Therefore, how to tackle construction waste problems, termed as construction waste management (CWM), has become a discipline in its own right. In the past decades, CWM has been receiving increasing attention in China due to the growing awareness of embracing sustainability. To address the issues arising from construction waste needs various grand, systematic initiatives. Among them, a promising initiative is to embrace the green build-

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ing movement in China.

Here, green building (GB) refers to an environmentally responsible and resource-efficient structure or site throughout its lifecycle (EPA, 2016). Amidst the global trend of achieving sustainable development, GB has been elevated to the top of many construction-related institutions, including China. An array of governments, professional institutions, and independent organizations have developed and launched green building rating systems (GBRSs) to outline GB standards and label certifications. There are eight categories of project performance defining the assessment criteria of most prevailing GBRSs, covering project management, site, energy, water, materials, emission and storage of hazardous materials, and indoor environment quality (Gou and Lau, 2014; Wu and Low, 2010). CWM, as a pivot indicating the degree of sustainability, has also been formulated in most prevailing GBRSs. It has been found that credits aiming for CWM are typically under the "Materials" category, comprising 8-12% of all the attainable credits in a GBRS (Tam et al., 2004; Lu et al., 2016). Substantial studies have been undertaken to examine the effects of GB on carbon emission, energy saving, occupant comfort, and property market price (Shuai et al., 2018; Castleton et al., 2010; Zhang and Altan, 2011; Fuerst and McAllister, 2011). Nonetheless, rather limited studies have explored the effectiveness of GB on CWM, even though CWM also plays a critical indicator of sustainability.

Hence, this study aims to evaluate the effects of GB on CWM in China, and then understand the causes of the effects by conducting a series of semi-structured interviews and a comprehensive political, economic, social, and technological (PEST) analysis. According to the PEST identified, a series of targeted strategies are also recommended to improve the CWM performance. This study will focus on the GBRS of U.S.-developed Leadership in Energy and Environmental Design (LEED) because it is the most prevailing and widespread GBRS globally. A large number of GB projects in China have also been accredited by LEED. The deliverables of this study are not only beneficial to researchers and practitioners in the GB industry, but also provide a good reference for exploring whether GB can be used as a tool to improve the CWM performance in China.

2. LITERATURE REVIEW

2.1 Green building movement

The concept of GB can be traced back to 1960s when the energy crisis and environmental pollutions concerns were increasingly anabatic due to the over-exploitation and overuse of fossil energy (Kibert, 2004). In the 1980s, with the increasing embracement of sustainability in various industries, the call for sustainability in the construction industry has become extremely strong with the consciousness of the building industry as a predominant contributor to energy consumption and environmental pollution (Lu and Tam, 2013). Under this background, UK created the first GBRS in the world, BREEAM (Building Research Establishment's Environmental Assessment Method), which represents a milestone of the world GB movement (Cohen et al., 1998). Afterward, the GB movement has entered the track of rapid development (Shen et al., 2012).

Nowadays, GB, also known as sustainable building, refers to a practice of creating structures and using processes that are environmentally responsible and energy efficient throughout a building's life cycle from planning, design, construction, operation, maintenance, and deconstruction (EPA, 2016). Green buildings are created to relieve the adverse impacts of the built environment on both human and natural environment (EPA, 2016). In recent decades, GB development has witnessed an unprecedented surge, and numerous GBRSs have been issued by an array of governments, professional bodies, and independent organizations to pioneer sustainability of the construction industry (Chi et al., 2020). GBRSs are tools to evaluate a building's performance in conformity to a series of criteria, such as site, energy, water, air, and materials (Wu and Low, 2010). Usually, GBRSs are created by a panel of experts and stakeholders (Gou and Lau, 2014). A study conducted by Vierra (2014) estimated that there are more than 600 GBRSs globally. Amongst them, LEED developed by the U.S. is the most widely spread GBRS. According to USGBC (U.S. Green Building Council) (2019), there have been more than 94,000 LEED-certified projects in 167 countries and territories globally.

In terms of the GB movement in China, the emergence of GB is later than the west until 2005 when the first "international green building conference" was held in Beijing. After one year, in 2006, the Chinese national GBRS, Green Building Evaluation Label (GBEL) was formally issued, which significantly catalysed the GB movement in China. Even though the inception of GB in China is later, the permeation of it is incredibly in-depth into the market. It is ambitious of the Chinese government that by 2020, 50% of new residential buildings will be certified by GBRSs according to its "Construction Industry 13th Five-Year Plan" (Zhang and Kang, 2018). Amongst the GBRSs prevailing in the Chinese market, LEED possesses the leading position among the clients despite the fact of the existence of its national GBRS, GBEL. It is announced by USGBC (2018) that there have been more than 1,211 LEED-certified projects (47.16 million m^2 GFA) until 2017, second only to the U.S. globally. That is why LEED in China is the research focus of this study.

2.2 Construction waste management

Construction waste, sometimes termed as construction and demolition waste (C&D waste), refers to the solid waste resulting from any construction activities, such as new construction, renovation, and demolition (Roche and Hegarty, 2006; Lu et al., 2019). Construction waste can be generally classified into two generic portions, inert materials and non-inert waste depending on whether it has stable chemical properties (EPD, 1998). Inert materials, such as soil, earth, slurry, rocks and concrete accounts for the vast majority of the construction waste and are suitable to reuse and/or recycle for different purposes, e.g., road formation, land reclamation, and recycled aggregate. The non-inert waste mainly comprises bamboo, plastics, glass, wood, and paper, contaminates the surrounding environment significantly. Therefore, it is not considered for reuse and/or recycling, and it is normally disposed of at landfills (Poon, 2007). Landfilling not only gives rise to negative social-economic impact but also leads to environmental degradation due to anaerobic decay of the materials disposed of and thus the production of carbon dioxide, methane, and leachate (Lu et al., 2015). It also rapidly exhausts invaluable land resources.

In recent decades, vast amounts of construction waste have been generated, which has raised worldwide attention. The need to tackle the construction waste issues gradually fosters the emergence of a distinct discipline, termed as construction waste management (CWM). CWM can be generally guided by the '3R' principles, i.e., reduce, reuse, and recycle according to their desirability (Peng et al., 1997). The 3R's highest priority, the reduction has been examined extensively, and substantive measures have been proposed. Such measures generally involve either (1) adopting low waste technologies; (2) reducing waste by design; (3) raising practitioners' attitudes towards waste reduction; (4) developing an effective waste management system; or (5) reducing waste by government legislation (Lu and Yuan, 2011). Reuse means using the same material in construction more than once, including using the material again for the same function (e.g. formwork in construction) (Ling and Leo, 2000) and new-life reuse for a new function (e.g. using the cut-corner steel bar for shelves; using the stony fraction for road base material) (Duran et al., 2006). Recycling is considered the final option before disposal. Through recycling, a variety of new materials can be made from construction waste (Bao et al., 2019).

Compared with other Western developed countries, construction waste issues in China are even worse. China is already the world's largest waste generator and by 2030 its volume of waste is projected to be double America's volume of municipal solid waste (MSW); nearly 40% of the MSW generated is construction waste, consuming about 40% of natural resources and energy (WEF, 2018; Wang et al., 2008). Nowadays, around three-quarters of Chinese cities are facing the dilemma known as "waste siege". The construction waste issues in China are even more grievous as a result of poor CWM performance (Bao and Lu, 2020). The reuse and recycling rate of construction waste in China is only 5% compared with that of 70%-95% in some developed countries (Huang et al., 2018). With the further urbanization and urban renewal in China, the construction waste issues would be even worse and thus, it is imperative for China to devise some effective strategies to tackle the construction waste issues. That is why this study aims to evaluate the effectiveness of GB on CWM to see whether GB can be applied as a strategy to tackle the construction waste issues in China.

2.3 GBRS and CWM dyads

Considering that CWM is pivotal in the process of fostering sustainability in the construction industry, some scholars have tried to explore the potential of GBRSs to tackle construction waste issues. For example, Lu et al. (2019) did a study trying to compare the CWM performance among the three GBRSs and unveiled that poor CWM performance is in common in the three GBRSs. Additionally, Lu et al. (2018) analysed the effects of BEAM Plus, GBRS developed by Hong Kong, on CWM performance with big data analytics applied and the results indicate that CWM performance shows a notable reduction only in demolition projects. Chen et al. (2018) furthered the study of the nexus between BEAM Plus and CWM and discovered that BEAM Plus has a negligible influence on CWM performance. Furthermore, Wu et al. (2016) compared the CWM related items in the five GBRSs and uncovered that GBEL, Chinese national GBRS pays the most attention to the CWM problems, while GBI (Green Building Index), GBRS developed by Malaysia emphasizes the CWM the least. To conclude, even though there have been some studies trying to investigate the potential of GBRSs to tackle the CWM, minimal research, if any, has probed into the effectiveness of LEED specifically in China, although China is the largest construction market and LEED is the most widespread GBRS. This is the research gap this study intends to fill.

3. RESEARCH METHODS

This study adopts a mixed method, including three phases, i.e., archive analysis, semi-structured interview, and PEST analysis. These three phases are related and succeeded mutually, which are elaborated individually below. To help the readers better understand the research methods adopted in this study, a framework is made (see Figure 1).

3.1 Archive analysis

This study begins with a series of desktop archive analysis to examine how GB projects certified with LEED influence CWM. The version of LEED v2009 New Construction is selected for this study due to the fact that the number of projects certified under the most recent version, the amount of LEED v4 is still limited during the study period. The authors first analysed where the CWM-related credits are allocated in the LEED. It is found that the credits related to CWM in this rating system normally reside under the category of 'materials & resources'. After finding out where the CWM-associated credits exist, the authors also identified the CWM-related credits by probing into the evaluation criteria by item by item. Afterward, a total of 310 GB projects accredited LEED including different certification levels in China has been sourced from the U.S. Green Building Council (GBC) in this study. By calculating and comparing the average rate of credits attained under the category of CWM and other performance categories, it is intuitional to discover how the GB affect CWM.

3.2 Semi-structured interviews

Following the archive analysis, the study conducted a series of semi-structured interviews to explore the reasons for the effect of GB on CWM (Wang et al., 2019). Therefore, a succession of semi-structured interviews was undertaken by collecting the qualitative data with LEED and CWM experts, consultants, researcher, project managers based in China. The detailed profile of the interviewees is shown in Table 1. In total, 10 experienced and professional in both LEED and CWM construction industry practitioners were

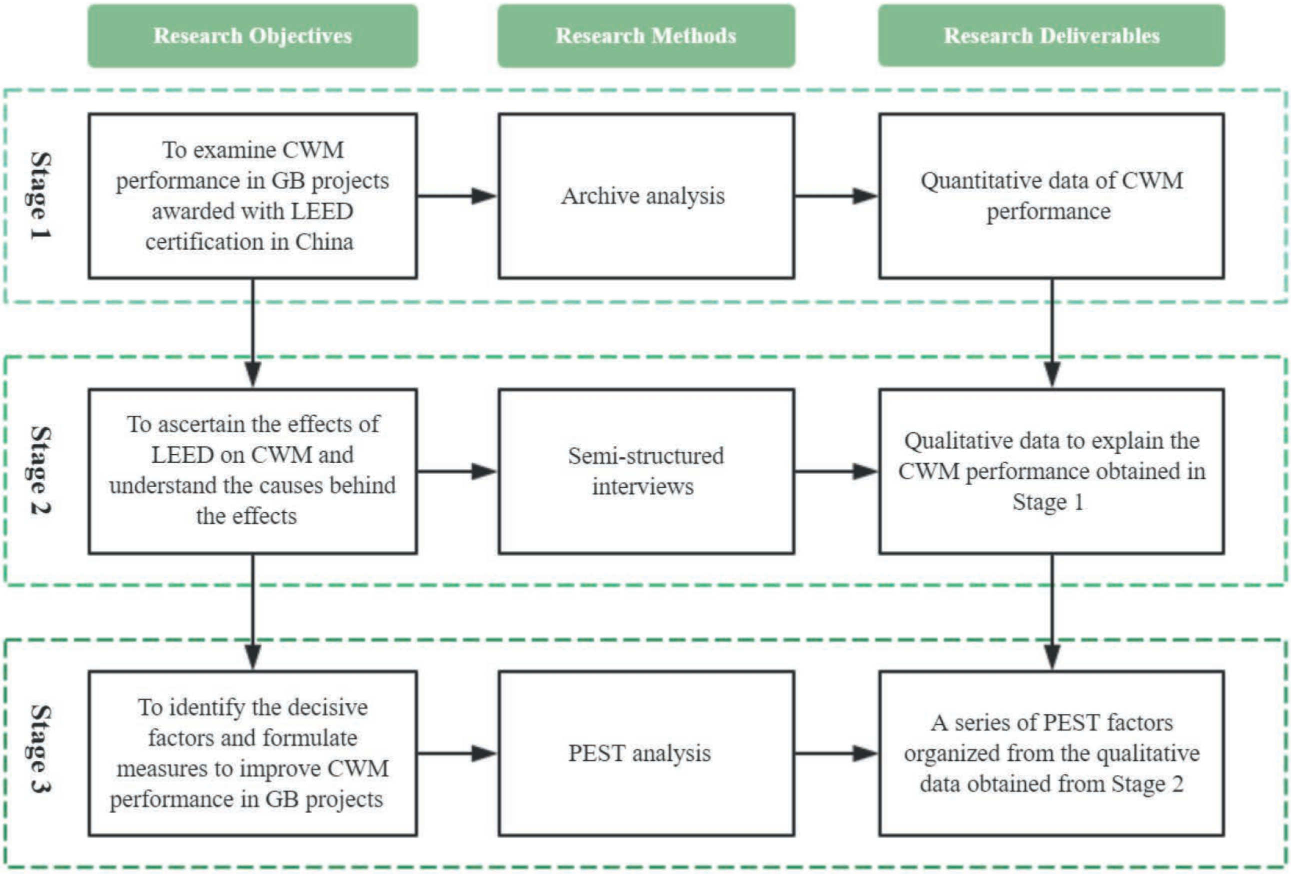

FIGURE 1: Framwork of research: objectives, methods, and corresponding deliverables.

interviewed. These interviewees, with deep industry experience, have been engaged in a great many GB projects awarded by LEED.

The interviews were started by asking some open-ended questions by eliciting the interviewee's overall opinions on the LEED itself in the context of China. Then the questions were extended to CWM in China, e.g.:

- How is the CWM situation in China generally?
- What do you think is the cause of the CWM situation in China?
- In your opinion, is the current CWM-related credits allocation in the LEED rational enough to better improve the CWM performance in China?

Afterward, the questions were even more targeted at the specific item in the LEED and asked credit by credit, e.g.:

- Which credits are relatively harder to achieve?
- Why the credits are difficult to obtain?
- Based on your practical experience, is there any effective approach to deal with the corresponding difficulty?

For each of the interview, it lasted between one to two hours. The tape-recording was conducted for some of the interviews, while if the interviewee did not agree to the recording, a detailed note was taken accordingly. If some notes taken during the interviews were not clear, the telephone call was dialled with the interviewee to check, which ensures the validity of the data.

3.3 PEST analysis

By collecting the qualitative data from the semi-struc-

TABLE 1: Profile of the interviewees.

No.	Role	Relevant working experience
1	GB expert in an architecture firm, architect, LEED AP	> 12 years
2	GB consultant in a world-class engineering consultancy firm, LEED AP	> 8 years
3	GB consultant in a local GB consultancy firm, LEED AP	> 5 years
4	GB consultant in a national architecture institute, LEED AP	> 6 years
5	GB consultant in a regional comprehensive design firm, engineer	> 15 years
6	GB expert in a regional architecture institute, LEED AP, engineer	> 15 years
7	Researcher in a regional architecture institute	> 5 years
8	GB expert in a regional GB council, architect, LEED AP	> 8 years
9	Project manager in a state-owned construction firm, engineer	> 20 years
10	Director in a real estate development firm, engineer	> 12 years

tured interviews, a comprehensive PEST analysis was conducted. The PEST refers to four factors, political, economic, social, and technological. It is a powerful and widely used tool for understanding strategic risk, identifying the change and the effects of the external macro environment (Sammut-Bonnici and Galea, 2015). PEST analysis has two functions. The first is to identify the external environment where the company or organization operates, while the other is to provide data and information to enable the company to predict situations and circumstances and formulate strategies (Yüksel, 2012). PEST was firstly developed to apply in the discipline of market research, but in recent years, it has been widely applied across the disciplines due to the awareness of its significance. Adapting the PEST analysis into this study, it can assess how GB affect the CWM in the context of China from four external factors, political, economic, social, and technological. The results of the analysis can inform the practitioners for reference to formulate the strategies to improve CWM in the future.

4. RESULTS AND FINDINGS

4.1 Archive analysis

From the archive analysis, Table 2 shows the credits allocated to CWM in the LEED. The study identified CWM associated credits by going through all the criteria item by item. The LEED version 2009 for New Construction includes seven aspects of assessment, i.e., sustainable sites, water efficiency, energy & atmosphere, materials & resources, indoor environmental quality, innovation, and regional priority credits. It is examined that pertinent credits relating to CWM lie on the performance category of materials & resources as listed in Table 1. The materials & resources category receives 14 credits out of total 110 credits, in the interim, 11 points can be attained by CWM. The attainable point(s) of each CWM related item, as well as its relative weight in the overall grade, were presented in the last two columns of the table, from where all CWM associated points account for 10% of the total LEED score. It can also be perceived that this rating system allocates dissimilar emphasis on different aspects of CWM performance when devising the crediting mechanism.

This study has sourced the scoring data of all the 310 projects (as of 30 April 2019) certified for green building certification as LEED (see Table 3) in China. Table 3 shows the detailed credit distributions of LEED-certified projects at different levels of certification. It is noticed that GB projects obtained low scores in the category of materials & resources (MR) where CWM related items are allocated. Based on the identification shown in Table 2, the authors created one more column depicting how CWM is performed in real-life LEED evaluation shown in the last column. The analysis so far indicates that less than 32% of the total applicable credits in CWM are attained, which are relatively lower in each level of certification. It can be induced that the designated performance of enhancing CWM via LEED has largely unattained. Furthermore, the detailed scores of each CWM-associated credit in the LEED-accredited buildings are elaborated in Table 4. It would be intriguing and necessary to keep digging the cause of this anomaly.

TABLE 2: CWM related credits in the LEED.

Overall assessment framework			CWM related credits			
	Performance category	Attainable points	Credit item		Attainable points	Attainable rate in overall grade
1	sustainable sites (SS)	26	MR1.1	Building reuse - Maintain existing walls, floors, and roof	3	2.73%
2	water efficiency (WE)	10	MR1.2	Building reuse - Maintain existing interior non-structural elements	1	0.91%
3	energy & atmosphere (EA)	35	MR2	Construction waste management	2	1.82%
4	materials & resources (MR)	14	MR3	Materials reuse	2	1.82%
5	indoor environmental quality (IEQ)	15	MR4	Recycled content	2	1.82%
6	innovation in design	6	MR6	Rapidly renewable materials	1	0.91%
7	regional priority credits	4				
	Total	110			11	10%

TABLE 3: The credits distribution of LEED-certified projects in China.

Certification Level	No. of projects	Average Overall Grade	SS (%)	WE (%)	EA (%)	MR (%)	IEQ (%)	ID	RP	CWM (%)
Platinum	45	82.11	82.56	95.56	74.48	44.29	42.13	5.16	3.98	37.58
Gold	147	64.64	75.20	91.09	37.84	39.94	36.17	4.63	3.90	32.53
Silver	89	54.16	68.41	77.30	23.18	37.00	31.95	4.27	3.65	28.91
Certified	29	45.07	66.31	60.00	17.04	31.28	24.43	3.59	2.17	25.39
Average	-	-	73.12	80.99	38.14	38.13	33.67	-	-	31.10

Note: SS = sustainable sites; WE = water efficiency; EA = energy & atmosphere; MR = materials & resources; IEQ = indoor environmental quality; I = innovation in design; RP = regional priority
Data Source: Public data posted on the official website of the U.S. Green Building Council

TABLE 4: The CWM related credits distribution of LEED-certified projects in China.

Certification Level	MR1.1 (%)	MR1.2 (%)	MR2 (%)	MR3 (%)	MR4 (%)	MR6 (%)
Platinum	0	0	97.78	1.11	91.11	33.33
Gold	2.72	0.68	78.91	0	78.91	1.36
Silver	1.12	0	91.01	0	66.29	0
Certified	2.3	0	84.48	1.72	50	0
Average	1.54	0.17	88.05	0.71	71.58	8.67

4.2 The performance of CWM explained

Apart from the quantitative data collected above, this study also conducted ten semi-structured interviews with GB and CWM experts and practitioners. A combination of the qualitative analysis and quantitative analysis can help understand the rationales, i.e., the causes behind the poor performance with respect to CWM in real practice and assessment.

4.2.1 MR1.1 Building reuse - Maintain existing walls, floors, and roof & MR1.2 Building reuse - Maintain existing interior non-structural elements

These two credits are aimed to encourage developers to reuse the existing, previously occupied building components including structural and nonstructural elements embedded in the first two items respectively. According to the requirements, the thresholds are set as 55% and 50% for the percentage of salvaged portion correspondingly. It can be noticed that minimal buildings can earn these credits even for the platinum-certified projects. Regardless of the certification levels, the average percentages of the projects that attained the credits MR1.1 and MR1.2 are only 1.54% and 0.17% respectively. In total, there are only six projects thereof that attained the credits among total 310 cases. As informed by the interviewees, the majority of GB projects are new construction, especially in China. These two credits are more applicable to reconstruction projects, which are more commonly seen in some developed countries, such as U.S. As such, it is scarcely for the new building to triumph the thresholds devised in the system. Additionally, achieving these credits is heavily reliant on the plan of demolition works in great details. The loss of previous design documentation, the tight project schedule, as well as the time cost expenditure can be the hindrance to living up the ideal demolition works as per the high LEED's standards. In that case, many developers and GB consultants will lower the priority of these two credits.

4.2.2 MR2 Construction waste management

To reduce the amount of construction waste going to the landfill and incineration facilities, the evaluation criteria are highly dependent on the data and necessary supporting documents submitted by the project contractors, indicating the percentage of the waste is recycled or salvaged. As shown in Table 4, the average percentage of the projects under the four certification levels reaches up to 88.05%. Although the data shows GB projects got the highest rate of points under this criterion, it reveals one issue derived from the interviews. In China, only a few cities launch their regional regulations to specify the waste disposal procedure appropriately, but they still do not specify the sensible way to calculate the amount of waste recycled or salvaged which LEED does demand. As reflected by interviewees, the documentation requested by LEED basically relies on the estimation by figuring the number of dumpers and contractors' experience. Due to the lack of verification mechanism in the LEED evaluation process, the data may be thus vague, which does not mirror the actual performance in the real-life projects.

4.2.3 MR3 Materials reuse

This credit requires to use salvaged, refurbished or reused materials, the sum of the cost for using such materials should be reaching at least 5% of the total value of projects materials usage. It is found that the credits are scored rarely with the average percentage calculated only 0.71% from Table 4. There are three major reasons for this phenomenon, according to the interviews. Firstly, the quality and durability of reusable/recycled materials are the primary concerns from project stakeholders. To this end, they are reluctant to undertake unnecessary risks arising from the quality of the recycled products. Secondly, both developers and clients are unenthusiastic reusing old building materials, partially due to their traditional Chinese mindset, fond of the new and tired of the old. Due to this mindset, the adoption of recycled products is bound to affect the selling price of the projects and then the profitability of the stakeholders, particularly for some commercial and residential projects under the background of the current increasingly heated real estate market in China. Thirdly, the cost savings for applying the criteria might not considerably economize the project cost, since the selection of qualified recycled materials can increase the extra labor cost and time cost and may not be able to perform price soar of the property to be sold. In that case, this credit may not be a good candidate for scoring.

4.2.4 MR4 Recycled content

As reflected from Table 4, the average percentage of the projects that attained the credit MR4 is 71.58%, which manifests that the majority of projects achieve the requirement of employing building products incorporated with recycled content, thereby decreasing the usage of natural virgin materials. As mentioned before, the validation of such credit depends on the evidence provided by the project developer/applicant. To obtain the credit, the developer or contractor should collect corresponding proper documentation to prove the proportion of recycled component reaches the thresholds from product suppliers. In the current market, there appears peculiar paucity of building materials originally labeled with recycled content value within a material. Therefore, the contractors should re-identify eligible material suppliers, or provide a template of document for material suppliers to fill in the information that LEED requests. However, there is no standard operating procedure of data collection to ensure the authenticity of the information. As reflected by interviewees, making sure the authenticity of documentation is still the loopholes in the GB evaluation process. Some interviewees even admitted that the situation of the data fabrication to attain the credit is not uncommon in China.

4.2.5 MR6 Rapidly renewable materials

The average percentage of the projects attaining this credit is calculated as 8.67% from Table 4. There appears to be a scarcity of GB projects that earned this credit because there are limited options for the materials specified in the requirement. According to the potential strategies mentioned in the indicator, it is suggested to consider using materials such as bamboo, wool, cotton insulation, linoleum, wheatboard, strawboard, and cork. Through interviews, the most commonly used types of rapidly renewable materials are bamboo and wood. For ordinary buildings, it is improbable to install these kinds of materials accounting for 2.5% of the total cost of building materials used in the whole project. Therefore, only some pilot demonstration projects that aim to achieve the big-league performance of LEED certification are planning to employ these kinds of certain materials in the very beginning usually at the stages like design or selection of suppliers, so as to get to the credit. Because of this condition, only platinum/gold-certified projects scored the point.

4.3 PEST analysis

From the results obtained from archive analysis and semi-structured interviews, CWM-related credits attained in the LEED-accredited projects in China were the lowest compared with other performance categories, inducing GB does not have distinct effects on improving regular CWM performance in China. A PEST analysis was conducted to analyze further and understand certain situations that China would face when promoting CWM in further ascension ahead. Four external perspectives (political, economic, social, and technological) are elaborated individually below (see Table 5).

4.3.1 Political dimension

Poor CWM performance in the GB projects can be attributed to the incomplete CWM regulations in China concerning the political dimension. Very few cities, like Beijing, Shanghai, and Shenzhen, have issued their regional CWM related regulations. A vast majority of cities still do not have any regulation to stipulate CWM issues. Even for the cities like Beijing, Shanghai, and Shenzhen, the regulations are far from complete compared with western countries and other developed economies. The current regulations are just enough to improve construction waste treatment at a very general stage. As a consequence, in terms of a specific situation, the regulations are difficult to follow. As echoed with Yuan (2013), comprehensive regulations could form a concrete basis to implement CWM. Nevertheless, the neighbors of (Mainland) China, e.g., Japan and Hong Kong, where the relatively mature and all-around CWM related political mechanism is implemented for addressing construction waste issues, and thereof have yielded notable effects on CWM. From the view of the political system, the primary endeavor is to formulate a series of systematic CWM related regulations at both national and regional levels. Besides, more efforts can be input in China to learn a well-rounded CWM system and practical experience from advanced countries/regions.

4.3.2 Economic dimension

In terms of the economic dimension, the lack of financial incentive gives rise to the subpar CWM performance in the GB projects. To have a better CWM performance in the GB projects, the cost imposed would be increased in various aspects. As mentioned by many interviewees, to obtain more CWM-related credits in the LEED-accredited projects, it is required to have efficient management on site and extra manpower for CWM, e.g., on-site sorting, which will surely increase the cost. Additionally, to obtain some credits, such as MR.3, it requires to use the recycled materials to diminish the consumption of virgin resources. However, the market for recycled building products in China is still in its infancy. The price of recycled building materials is even higher than virgin/ordinary materials, which economically hinders the stakeholders from adopting recycled materials. It makes sense that the environment is generally in the lower priority compared with the cost, quality, duration, and safety in the Chinese construction industry

TABLE 5: Summary of PEST analysis.

PEST dimensions	Specific factors
Political dimension (P)	P1: limited regulatory enforcement on CWM in a handful of cities; P2: imperfect CWM-relatad political system by the national and regional authorities; P3: inadequate political support in promoting the adoption of recycled construction materials.
Economic dimension (E)	E1: addtional investment / cost in CWM (e.g. labor cost, on-site sorting); E2: premature market for construction waste recycling industry; E3: lack of economic motivation in promoting the adoption of recycled construction materials.
Social dimension (S)	S1: weak public awareness about developing better CWM; S2: excessive worries on recycled construction materials from project stakeholers; S3: undertrained on-site workers.
Technological dimension (T)	T1: lack of core competente on recycled construction materials manufacture; T2: deficient information and tools to implement proper demolition / deconstruction; T3: insufficient funds for supporting research in field of CWM.

(Shen et al., 2006). Therefore, some strategies could be formulated from an economic perspective. For example, the government can provide direct financial incentives if the stakeholders use recycled materials to a certain level in the projects.

4.3.3 Social dimension

Considering the social dimension, a relatively low level of stakeholders' awareness about CWM in China results in the poor CWM performance in these projects. The contractors are reluctant to conduct CWM because, in the traditional Chinese construction industry, more concerns have been given to cost, quality, duration, and safety instead of the environment. For instance, on-site sorting is regarded as an effective approach for dealing with construction and demolition waste. However, in the real practice, the waste generated is usually divided into "can be sold", e.g., steel, metallic materials; and the remaining "cannot be sold" waste is muddled up together, then going to landfills without being separated. Moreover, generally in China, the on-site workers are not well educated, which causes their awareness on CWM is very low if there is not enough training provided for them. The behaviors of workers may directly impact the effectiveness of CWM in the execution. Therefore, the targeted strategy from the social perspective is that the government should provide a series of training courses for construction industry practitioners to enhance their awareness about the environment, as well as launch certain award as an incentive for stakeholders to actively involved and perform in CWM-associated events, so as to arouse public consciousness.

4.3.4 Technological dimension

Regarding the technological dimension, the interviewees stated that the current technology in China is still not advanced enough to support to have a better CWM performance. Some interviewees explained that most equipment in China to produce recycled construction waste materials is imported from western countries, significantly increasing the cost of recycled materials in the market. In turn, higher price hinders the stakeholders from using it in the projects. The above challenges have been largely responsible for the immature market for construction waste recycling, which will eventually affect the effectiveness of CWM performance. Additionally, there appears no specific and clear standard concerning the demolition or deconstruction work where a significant amount of waste generated. Demolition wastes are heterogeneous mixtures of building and decoration materials that are usually contaminated with lots of chemical components. As reflected by interviewees, a proper sequence and approach of demolition may primarily simplify the treatment of waste at source. Starting from this point, the proposed strategy is that the government should invest a considerable amount of funds to support the CWM related research, research and development (R&D) in the fields of construction and demolition waste minimization, construction waste treatment guidelines, lean construction, prefabricated building, recycled building materials, sharing or trading of construction waste.

5. DISCUSSION AND RECOMMENDATION

Based on the results from PEST analysis as summarized in Table 5, some targeted recommendations have been proposed in accordance with the political, economic, social, and technological dimensions, which are elaborated individually below.

5.1 Political dimension

A well-developed CWM regulatory system with a well-defined hierarchy from national to regional should be established and emphasized in more cities as currently, only limited cities have formulated the CWM-related regulations. Also, the regulations formulated should be as detailed as possible so that they are straightforward to follow and enforce. Additionally, the conflicts of the regulations at the national and regional level should be avoided. Finally, once the CWM regulatory system has been enacted, the enforcement should be strictly executed.

5.2 Economic dimension

In the economic dimension, a mature construction waste recycling market should be cultivated, which can also provide some financial incentives for stakeholders to conduct the CWM behaviors, e.g., on-site sorting, partially if not fully covering their additional cost. Additionally, the government, either regional or national can allocate a specific fund to promote the adoption of recycled construction materials in construction, for example, different levels of subsidies can be set when different percentages of recycled construction materials are adopted in construction.

5.3 Social dimension

In terms of the social dimension, the government should use various public media tools to raise awareness of the public about the importance of CWM, such as newspapers and television programs. Moreover, the government can provide regular training courses for the practitioners involved in the CWM, including architects, engineers, contractors, and workers. Through the proper training, on-site workers can have better knowledge and skills on how to achieve better CWM performance and excessive worries on recycled construction materials from stakeholders can also be relieved or even eliminated.

5.4 Technological dimension

Regarding the technological dimension, the government should provide more funds to support the research of CWM in different manners. For example, the proper techniques for conducting demolition activities can reduce construction waste most in different situations. Moreover, the various equipment to achieve better CWM performance, such as on-site sorting equipment and the equipment for the storage of sorted construction waste should be developed as soon as possible so that the cost of stakeholders to use the equipment can be reduced. Additionally, the compilation of the standards of recycled construction materials, e.g., recycled aggregate for concrete is also of great importance.

6. CONCLUSION

Construction waste issues have raised worldwide attention due to their negative social-economic and environmental effects. Particularly in some developing countries, massive construction activities have been undertaking to support their economic growth, leading to profuse construction waste generation. A promising strategy to address the issues is to promote green building. Some past studies have been conducted to investigate the potential of GBRSs to address CWM, but rather limited studies have probed into the effectiveness of LEED specifically focusing on China, hence calling for more studies to be conducted due to the extreme seriousness of CWM issues in China. To this end, this paper evaluates the effects of totally 310 GB projects accredited by LEED on CWM in China. From the archive analysis, it is discovered that CWM performance in these LEED certified GB projects is rather poor. The causes behind this phenomenon have been pointed out from item to item in the LEED based on the semi-structured interviews with GB and CWM experts and practitioners. Afterward, a comprehensive PEST analysis was conducted to probe into this phenomenon in the context of China.

Generally, the multiple causes, incomplete CWM regulations in China, lack of economic incentives of CWM, inadequate awareness concerning CWM and lack of advanced technologies from political, economic, social and technological (PEST) perspectives respectively hinder contractors and developers from achieving excellent CWM performance in practice. Finally, some strategies have also proposed to improve the CWM performance based on the PEST analyses. The study is beneficial to both researchers and practitioners in the GB industry. It also provides an important reference for exploring whether GB can be used as a tool to improve the CWM in China.

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AN INVESTIGATION OF THE GENERATION AND MANAGEMENT OF CONSTRUCTION AND DEMOLITION WASTE IN VIETNAM

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Vietnam

ABSTRACT

As a developing country in the context of globalization, Vietnam is experiencing a boom in its economy, characterized by a rapid rate of urbanization nationwide. Together with its benefits, this process also puts pressure on the environment, among which the increasing generation of construction and demolition waste (CDW) is an urgent issue. In this study, the authors conducted an in-depth investigation of various aspects in the generation and management of CDW in Vietnam. Firstly, part of the overall picture of CDW management in Vietnam was revealed in detail from the perspective of demolition contractors, one of the important stakeholders in the field. Their insights provide valuable information on current situation, practice, and attitude towards CDW recycling. Secondly, this paper reports the generation rate of different categories of materials from demolition sites of building structures in Hanoi, Vietnam, with the main focus on crushed concrete and crushed brick (CC-CB) and steel as they make up the majority of the generated waste. In order to achieve two mentioned goals, close contact was constantly kept with a key collaborator – a renowned demolition contractor in Hanoi throughout the investigation. The collaborator provided proper introduction to enable interviews with various other contractors, together with information of buildings being dismantled.

1. INTRODUCTION

According to official statistics, the total amount of CDW generated in major cities in Vietnam is approximately 1.46 to 1.92 million tons per year (MONRE, 2011). Together with the low recycling rate of only 1% to 2% (MONRE, 2011), this enormous and increasing source of solid waste is also ill-managed due to the lack of legislation, awareness, technology, and various other reasons. In various other countries, CDW has been studied and proven to be an effective substitute for natural materials. For instance, the first systematized research on CDW as aggregate for concrete was conducted in Germany soon after World War II (Nixon, 1978). Should the main content of CDW (brick, concrete, metal, wood, etc.) be reused and recycled, it would not only be economical but also reduce pressure on the urban environment. The status of the annual CDW generation and treatment in some advanced countries is summarized in Table 1 to compare with the situation in Vietnam.

Various studies converge on the fact that CDW poses as one of the major concerns to the environment even in developed countries due to its enormous percentage in the total amount of solid waste. In the United States, 534 million tons of CDW were generated in 2014 (EPA, 2016) which made up 67.4% of solid waste. Respective numbers in the EU-28 in 2016 were reported to be 924 million tons and 36.4% (Eurostat, 2018); whereas in China in 2013 were 2.36 billion tons and 30-40% (Zheng et al, 2017; Huang et al, 2018). The topic of management of CDW has been intensively studied in many countries. It is possible to mention recent papers in European countries such as Germany (Hiete et al, 2011), Norwey (Bersgdal et al, 2007), Portugal (de Melo and Goncalves, 2011; Coelho and de Brito, 2012), Spain (Coronado et al, 2011; Mercante et al, 2012; Saez et al, 2013), Greece (Fatta et al, 2003; Banias et al, 2010); in Asian countries and regions such as China (Hu et al, 2010; Lu et al, 2011; Xiao et al, 2012; Jin et al, 2017), Hong Kong (Deng et al, 2008; Tam, 2011), Taiwan (Hsiao et al, 2002), Kuwait (Kartam et al, 2004), Malaysia (Hussin et al, 2013), Thailand (Koforowola and Gheewala, 2009; Luangcharoenrat et al, 2019); as well as in many others around the world. All countries strongly encourage recycling CDW as one of the sustainable solutions and some have achieved high recycling rate as summarized in Table 1.

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TABLE 1: Estimated amounts of CDW generated and recycled percentage in some countries including Vietnam.

S/N	Country	Year of data	Amount of CDW (Million tons/year)	Recycled percentage (%)	Source
1	Japan	2012	72.69	96	MLIT (2017)
2	Korea	2009	67	36	Bansal and Singh (2014)
3	Germany	2012	89	85	Deloitte (2017)
4	France	2012	76	93	Deloitte (2017)
5	United Kingdom	2012	47	70	Deloitte (2017)
6	Italy	2012	40	79	Deloitte (2017)
7	Spain	2012	28	98	Deloitte (2017)
8	Netherlands	2012	26	83	Deloitte (2017)
9	Vietnam	2009	1.5-1.9	1-2	MONRE (2011)

The CDW situation in Vietnam is gradually getting worse with all existing research agree on an overall rising trend of the amount generated. Lockrey et al (2016) projected the total amount of CDW generated in 2020 in Vietnam to be 6.3 million tons, and in 2025 to be 11 million tons. Inadequate management of CDW is causing Vietnam three major problems: illegal dumping, hazardous waste, and depletion of natural resources (Nguyen et al, 2018). In recent years, the Vietnamese government has enforced strong measures through the Ministry of Construction and Ministry of Natural Resources and Environment in order to realize the 'National Strategy for Management of Solid Waste up to 2025, and Vision towards 2050.' The mentioned strategy, issued by the Prime Minister, pursuant to Decision No. 2149/QD-TTg on 17 December 2009 states: "In 2025, the aim is recovering 90% of the amount of CDW, and recycling 60% of such amount".

Literature review reveals that studies on CDW in Vietnam are limited in number, it is even possible to state that this is one of the new frontiers of research in the country. Statistics of CDW are available in periodic Reports on National Environment by the Ministry of Natural Resources and Environment (MONRE), and serve as good references for researchers. Naturally, CDW is grouped in the solid waste category, and sometimes is considered hazardous. Nguyen (2009) briefly touched CDW among the studied hazardous industrial waste. By summarizing existing knowledge, the paper presented an overall view of the situation in Vietnam including practices and regulations, and proposed a number of future directions for treating this general type of waste. Tong et al (2013) presented the experience of reusing and recycling CDW as recycled aggregates for concrete in the world, and suggested employing this method for CDW in Vietnam. The authors compared studies of recycled aggregate concrete in various countries, and summarized data of important mechanical properties such as strengths, elastic modulus, and water absorption to give insights on its viability as a construction material in Vietnam. Also targeting the prospect of reusing and recycling CDW, Lockrey et al (2016) conducted a careful assessment on opportunities and challenges. The authors employed known data to project the total CDW generated in Vietnam and in Hanoi; a number of stakeholders (policy makers, Urban Environment Company, contractors) are then interviewed to analyze the CDW flow in Hanoi, current constraints to recycling CDW, and the stakeholders' opinion on the economic attractiveness of CDW. A thorough study by Nguyen et al (2018) presented an overall view of CDW management situation in Vietnam. Within the paper, a number of topics were discussed such as: CDW generation countrywide and trend, responsible stakeholders and current practice in managing CDW, legislations and regulations on CDW, together with recommendations for sound CDW management in Vietnam. It is clearly observed that most existing literature mainly stopped at overview level, with the exception of Lockrey et al who provided some field data through interviews. An in-depth view on the complicated state of CDW management in Vietnam is rather lacking.

This paper intends to shed further light on such situation by attempting to achieve two objectives. First, to gain an overall understanding of demolition contractors in the two largest cities – Hanoi and Ho Chi Minh City (HCMC). Being one of the important stakeholders in the CDW management process, they can reveal the practices, awareness, and feasibility of CDW recycling in Vietnam. Second, to come up with an estimation of waste generation rate from demolition of structures in Vietnam (case study in Hanoi). This value can effectively aid in future projection of demolition waste by relating it to construction rate. An important clarification must be made between "construction waste" (CW – generated from new constructions) and "demolition waste" (DW – from dismantling or renovating old constructions) in the context of Vietnam in order to specify the scope of this study. It is later found out through interviews that most demolition contractors also operate in the field of construction, and many of them accept to transport both CW (hired by construction contractors) and DW (from their own demolition sites) since they possess the necessary equipment. Hence, the first objective of this study targets both CW and DW. The second objective, however, only targets DW from dismantling buildings in Hanoi.

2. RESEARCH METHODOLOGY

It was fully acknowledged that to sketch a clear picture of CDW situation in the two largest cities of Vietnam, the party that directly involves in the process of handling CDW – namely demolition contractors – must be reached out for. This approach was also maintained in the obtainment of building structures to be surveyed in the quantification of DW. To attain the two intended targets specified in previous section, the authors made a complete list of demolition contractors in Hanoi and HCMC, and kept close contact with them throughout the course of the study. Field methods such as interview or survey were then employed to collect data from such party. One particular contractor in Hanoi was also identified as key collaborator and introduced the authors to survey various demolition sites. This step was the most crucial to the success of this study since the CDW business in Vietnam in general, and in Hanoi in particular, is rather complicated. Normally, it is challenging to gain access to demolition sites since all contractors are extremely cautious. Only with the introduction of a renowned contractor were the authors allowed to conduct site visits and ask further data and information.

2.1 Interview methodology

Face-to-face interviews were conducted with a total of 46 demolition contractors currently operating in the two largest cities in Vietnam, Hanoi and HCMC. There were 29 contractors in Hanoi and 17 in HCMC who agreed to be interviewed, the number of employees among which ranged from several dozen to approximately two hundred. The companies participate in a wide range of activities as will be shown in the subsequent results and discussion part of this paper; however, they are directly involved in demolishing structures, and/or transporting CDW, and/or collecting and treating CDW.

The contractors were first contacted by email and telephone to set up meetings. Interviews in Hanoi were carried out continuously from March 2018 through January 2019; while those in HCMC were conducted in December 2018. Because of the complicated nature of the demolition business in Vietnam, an agreement was reached with the interviewees, in which the two basic codes of conduct are:

- Results collected in the interview is to be used for research purposes only; and
- Identities of interviewees are kept confidential.

Therefore, interviewees are identified by a code number. There is no association with the code number and the sequence of appearance of contractors in the data statistics.

The set of interview questions were designed to target three main areas of information:

- Basic information on contractors: number of employees, fields of operation, experience in the demolition business, and average number of demolition contracts per year;
- Awareness of CDW policies in Vietnam; and
- Challenges in implementing compulsory classification of all CDW.

2.2 Methodology for quantifying CDW

2.2.1 Overview of different methods to quantify CDW

An in-depth and thorough study by Wu et al. (2014) summarized the main groups of methods to quantify CDW as presented in Table 2.

It is noted that the amount of CDW in groups of methods 2 and 3 can be determined either by performing a quantity survey on as-built drawings, or by requesting it from contractors.

Methods in group 1 have the advantage of being easily visualized and specific; the shortcoming, however, is that they only produce raw data. The second group of methods (GRC) is also the most popular among researchers as it provides CDW generation per unit (whether per capita, or dollar, or square meter) which can be compared, contrasted, and conclusions drawn from. Groups of methods 3, 4, and 5 are relatively modern and require a rather complete database on the building (group 3) or the CDW management system as a whole (groups 4 and 5), which can be challenging to conduct in developing countries such as Vietnam.

2.2.2 Proposed method of survey

Considering the real scenario in Vietnam, the authors adopted a combination of the indirect measurement method (SV group) and the area-based calculation method (GRC group). These two methods complement each other well

TABLE 2: Groups of methods to quantify CDW.

S/N	Group of method	Method	Description
1	Site Visit – SV	Direct measurement	Calculate CDW stockpile by geometry
		Indirect measurement	Record number of transport trucks
2	Generation Rate Calculation – GRC	Per-capita multiplier	Amount of CDW ÷ Population Unit: kg/capita/year
		Financial value extrapolation	Amount of CDW ÷ Building value (USD) Unit: kg/USD
		Area-based calculation	Amount of CDW ÷ Gross Floor Area (m^2) Unit: kg/m^2
3	Lifetime Analysis – LA	Building LA	Amount of CDW ÷ Building age (year) Unit: kg/year
		Material LA	Amount of CDW ÷ Material age (year) Unit: kg/year
4	Classification System Accumulation – CSA		Based on GRC, considering effects of the classification system
5	Variables Modelling – VM		Model the CDW generation process by a function of influencing factors

since they ensure the provision of specific and precise amounts of DW together with comprehensive per square meter data.

In the first step of the procedure, the gross floor area (GFA) of each building was determined by direct measurement on site and verification with information from as-built drawings.

Throughout the demolition process, DW generation was monitored closely. The authors aimed at two main categories of DW, CC-CB and steel. Let the total bulk volume of CC-CB after being crushed to separate reinforcing steel be , this volume is then treated in two ways:

- A major portion is transported to landfills, denoted; and
- Another portion is employed as backfilling material on site, denoted .

Volum is determined by recording the number and type of transport trucks, and was used for calculating the generation rate of CC–CB as it is the portion of DW that is discarded, and hence exerts pressure on the environment.

Volume V1 is converted into mass by multiplying it with the bulk density of CC–CB, (Mália, 2013):

$$m_{CC-CB} = V_1 \times \rho_{CC-CB} \quad [kg] \tag{1}$$

The generation rate of CC–CB is then calculated by the following formula:

$$G_{CC-CB} = \frac{m_{CC-CB}}{GFA} \quad [kg/m^2] \tag{2}$$

Because steel (as well as other metals) is always separated for scrap sale, its mass can be easily obtained. The steel generation rate, therefore, can be determined as follows:

$$G_S = \frac{m_S}{GFA} \quad [kg/m^2] \tag{3}$$

A few photos illustrating the procedure of the survey method are shown in Figure 1.

3. RESULTS AND DISCUSSION ON CDW MANAGEMENT IN VIETNAM

3.1 Scale and business of demolition contractors in Vietnam

Among the 29 demolition contractors in Hanoi, 26 also operate in the field of construction; 15 also transport CDW, among which two companies (Bắc Quân and Tài Lợi) specialize only in transporting CDW. The manpower distribution of these contractors is displayed in Figure 2.

In HCMC, 16 out of 17 demolition contractors simultaneously take on construction works, and seven of them also transport CDW. The manpower distribution of HCMC demolition contractors is shown in Figure 3.

In Vietnam, there are no professional demolition contractors. Interviewees from some companies revealed that they also operate in non-engineering fields such as interior design (HN-25), importing beverages (HN-11), or selling air tickets (HCMC-04).

Compared to Hanoi, the number of demolition contractors in HCMC is significantly less. This was confirmed by interviewee HCMC-10 as he explained: “Small demolition works in HCMC are carried out by individual groups of workers, who do not bother to register a company as Southerners work quite casually”.

From the manpower distribution graphs, it is noted as follows:

- The overall trend is that demolition contractors have a low number of engineers (0-7) and many fewer permanent workers than temporary workers (average ratio is 1:2 to 1:4);
- The only two companies that have low numbers of temporary workers are Bắc Quân and Tài Lợi, as they specialize in transportation of CDW, and thus hire only permanent truck drivers;
- Companies that also take on construction consultation work (Dr. House, Bắc Việt, and Thăng Long in Hanoi, and ITSCO 68 and Hoàng Gia in HCMC) have more engineers (15-20).

Many of the surveyed contractors have had more than 5 years of experience in the demolition field (28 out of 49, accounting for 57.14%). The number of demolition contracts that they take on per year varies widely from 20 to 200. From the data collected, it can be estimated that there are 1500-2000 buildings (large and small) being demolished in Hanoi annually. In HCMC, there are approximately 900 large works demolished per year. These numbers are also increasing with the urbanization of Vietnam. Hence, it is crucial that CDW management strategies are formulated and implemented to control such pressure on the urban environment.

FIGURE 1: Survey of CDW generation (a) Determining GFA; (b) Scrap buyers measuring the amount of aluminum; and (c) Counting the number of CC–CB transport trucks.

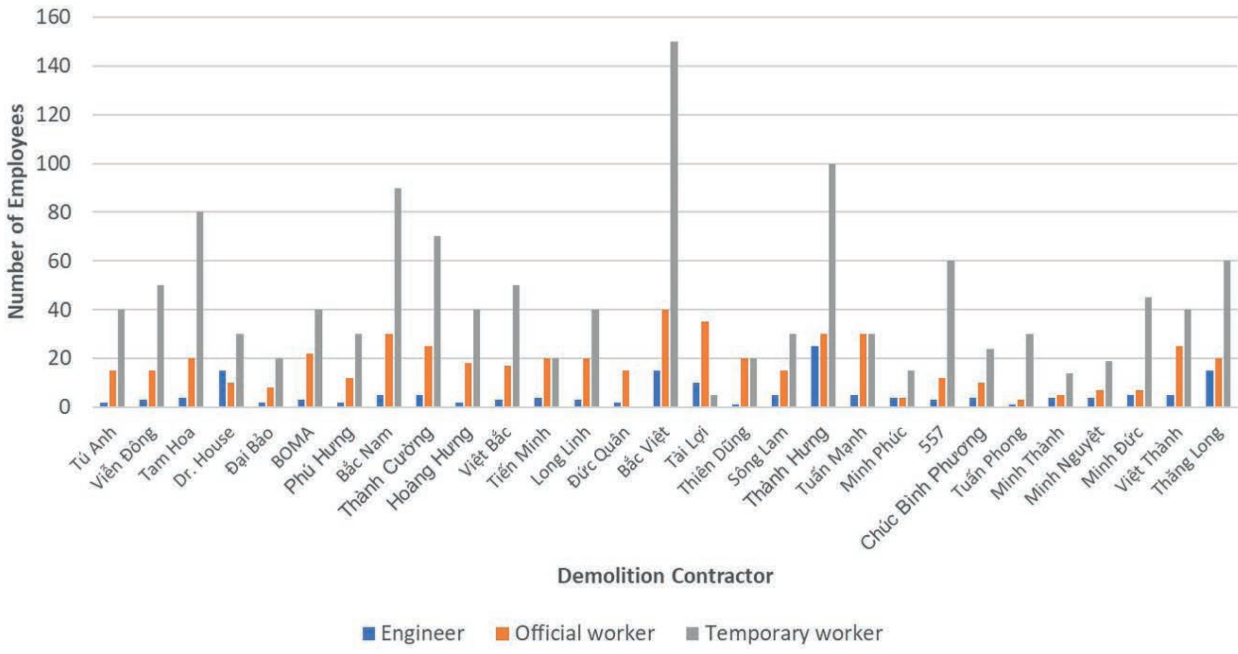

FIGURE 2: Scale of manpower of demolition contractors in Hanoi.

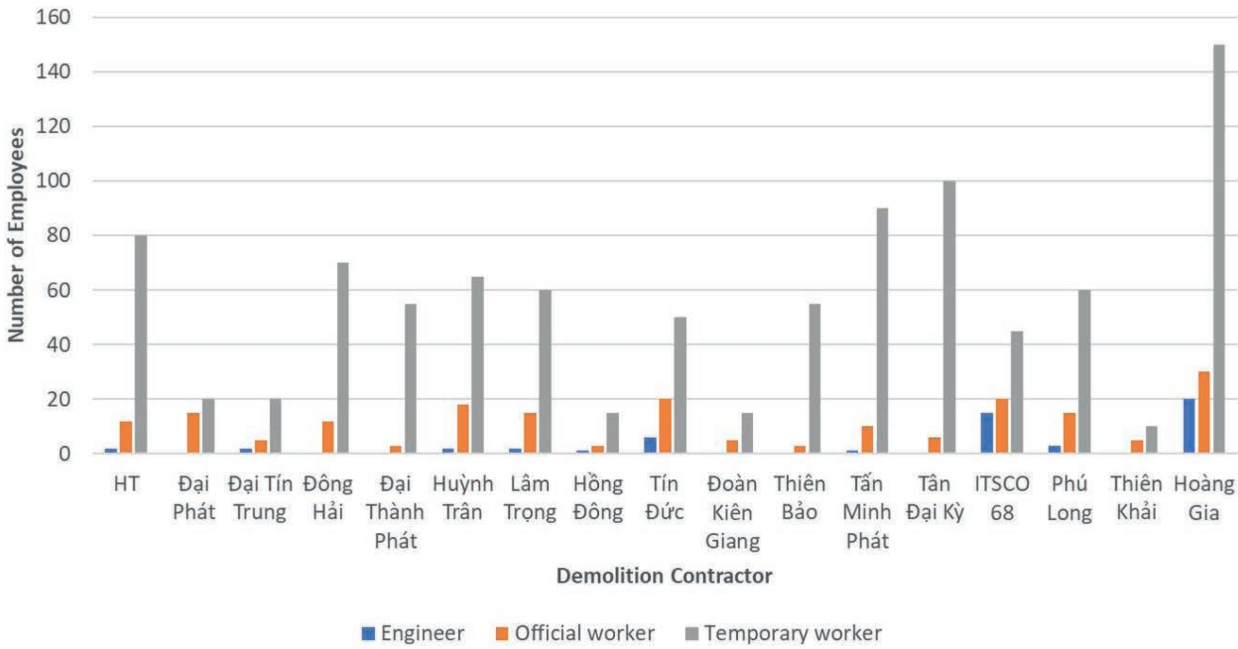

FIGURE 3: Scale of manpower of demolition contractors in Ho Chi Minh City.

3.2 Awareness of CDW policies

Currently in Vietnam, there is a set of legal documents regulating solid waste management in general, some of which have attempted to include CDW in their scopes; a summary of these documents can be seen in Table 3 (Nguyen et al., 2018).

Among the items in Table 3, Circular No. 08 directly aims towards CDW, and it is expected to be closely related to the operation of demolition contractors. However, 100% of interviewees were unaware of any legal document on CDW management, let alone Circular No. 08.

The interviewees were also asked for their opinions on current legislations on CDW in Vietnam; in particular: Are current regulations 1) Easy to follow, 2) Appropriate, 3) Economically efficient, and 4) Necessary. The answers were ranked on five levels, from totally disagree to totally agree. By adopting fuzzy logic theory, linguistic variables were converted into numerical values (Zimmermann, 2001) with the scale as shown in Figure 4 for data analysis.

The results of interviews are presented in Table 4. It is obvious that demolition contractors in Vietnam are neutral, even tend to disagree with the first three items because

TABLE 3: Vietnamese legal documents relevant to CDW management.

S/N	Type and name of document	Year of approval
Lawsuit		
1	Law on Construction	2014
2	Law on Environmental Protection	2014
3	Law on Urban Planning	2009
4	Law on Public Investment	2014
Decree		
5	Decree No. 59/2007/ND-CP on Solid Waste Management	2007
6	Decree No. 12/2009/ND-CP on Management of Investment Projects on the Construction of Works	2009
7	Decree No. 38/2015/ND-CP on the Management of Waste and Discarded Materials	2015
8	Decree No. 59/2015/ND-CP on Construction Project Management	2015
Circular and Joint Circular		
9	Circular No. 29/1999/QD-BXD on Promulgating the Regulations of Environmental Protection Applied for the Construction Sector	1999
10	Circular No. 10/2000/TT-BXD on Guiding the Elaboration of Reports on the Assessment of Environmental Impacts for Construction Planning Projects	2000
11	Circular No. 01/2011/TT-BXD on Guiding the Strategic Environmental Assessment in Construction and Urban Plans	2011
12	Circular No. 08/2017/TT-BXD on Regulation on Construction Solid Waste Management	2017
13	Joint Circular No. 01/2001/TTLT-BKHCNMTBXD on Guiding the Regulations on Environmental Protection for the Selection of Location for, and the Construction and Operation of Solid Waste Burial Sites	2001
Vietnamese Standard		
14	TCVN 6705: 2009 Ordinary Solid Waste – Classification	2009
15	TCVN 6706: 2009 Hazardous Solid Waste – Classification	2009
16	TCVN 6707: 2009 Prevention and Warning Signs for Hazardous Waste	2009
17	TCVN 6696: 2009 Requirements for Environmental Protection for Sanitary Landfills	2009

they have not even heard of such regulations. To quote interviewee HN-21: "Having practiced in this business for over 15 years, the process of demolition is totally dependent on our experience. Previously, up until the early 2000s, Hanoi did not plan a CDW dumping site; only recently there are several sites designated for this purpose, but they are very quickly filled up. We have to maintain an internal network among ourselves to share information of any lake or any idle construction project site that the owner wants to be backfilled or leveled." However, with a high level of consensus, demolition contractors in both cities highly agreed that regulations must be implemented so that CDW is managed and recycled properly.

3.3 Challenges in implementing compulsory classification of all CDW

When asked about their interests in CDW recycling, all interviewees were enthusiastic, and acknowledged that it is an inevitable trend. Interviewee HN-05 stated: "We hear that Hanoi used to have a project sponsored by Germany to recycle CDW, even a crushing machine was bought to run pilot tests. It is a pity that the project has not yielded any visible results." In HCMC, interviewee HCMC-10 stated: "We have been looking forward to the prospect of CDW recycling, and we will be more than willing to cooperate should such an operation go into action."

Nevertheless, in order to recycle CDW, it must first be sorted into different categories. This extra step may challenge demolition contractors in quite a number of aspects. The authors gave all interviewees a list of nine possible difficulties, both subjective and objective, that they would potentially face when the procedure of classifying all CDW is implemented. Each difficulty was assessed by demolition contractors on five levels, from virtually no challenge to the most challenging. These linguistic variables were then processed with the employment of fuzzy logic with a scale as displayed in Figure 5.

In Table 5, challenges are ranked from 1 (highest numerical value – the most challenging) to 9 (lowest numeri-

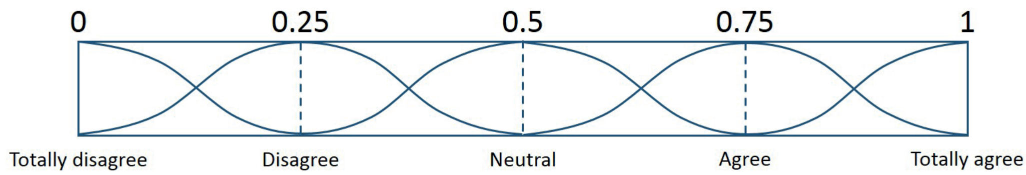

FIGURE 4: Linguistic variables of level of agreement.

TABLE 4: Opinions of demolition contractors on current legislations on CDW in Vietnam.

S/N	Inquiry	Opinions of demolition contractors in Hanoi		Opinions of demolition contractors in HCMC	
		Numerical value	Translation into opinion	Numerical value	Translation into opinion
1	Ease of following	0.47	Neutral	0.47	Neutral
2	Appropriation	0.52	Neutral	0.50	Neutral
3	Economic efficiency	0.35	Neutral, slightly disagree	0.35	Neutral, slightly disagree
4	Necessity	0.78	Agree	0.81	Agree

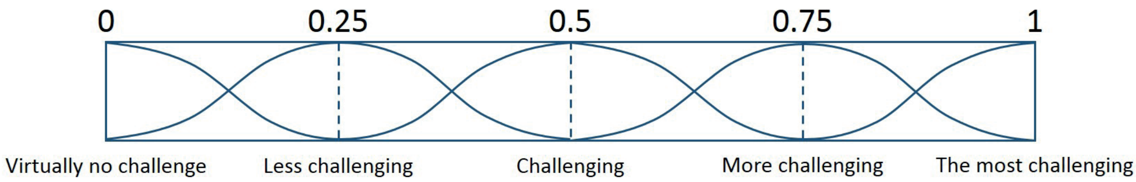

FIGURE 5: Linguistic variables of level of difficulty.

cal value – the least challenging). If there is more than one challenge with the same score, they will share a common rank number. It is observed that the following four challenges are at the top ranking by the assessment of contractors from both cities:

- Cost increase;
- Time consumption;
- Lack of legal guidelines; and
- Lack of post-demolition treatment facilities.

Among these four, the first two are subjective and have a close relationship to each other (cost – time). The other two are objective and need special attention by policy makers as well as researchers to enable such conditions.

4. RESULTS AND DISCUSSION ON GENERATION OF DEMOLITION WASTE, A CASE STUDY IN HANOI, VIETNAM

4.1 General information of surveyed buildings

Applying the survey method described in the previous section, the authors surveyed DW quantities at a total of nine structures being demolished in Hanoi. The demolition works occured from October 2018 to March 2019. Information on nine buildings is presented in Table 6 including the age, function, and site area. It is necessary to distinguish between site area and GFA, since a building may have multiple floors; moreover, the ground floor area may be smaller than the site area. In this current study, all nine buildings had similar structural features: a reinforced concrete (RC) frame with brick walls.

Each building was designated with a unique ID for ease of reference from this point onwards.

4.2 Typical flow of demolition work in Hanoi, Vietnam

Through daily visits to demolition sites as well as through discussions with site managers, the authors are able to summarize a typical procedure for demolishing buildings, as illustrated in Figure 6. It was confirmed by all site managers that this flow of work is typical and is a combination of manual and machine demolition.

The first phase of demolition involves removal of interior mechanical, electrical, and plumbing (MEP) components,

TABLE 5: Ranking of challenges in implementing compulsory classification of all CDW by demolition contractors in two major cities of Vietnam.

S/N	Challenge	Demolition contractors in Hanoi		Demolition contractors in HCMC	
		Numerical value	Ranking	Numerical value	Ranking
1	Cost increase	0.60	3	0.40	3
2	Manpower shortage	0.41	7	0.32	5
3	Time consumption	0.56	4	0.43	2
4	Lack of skill and technology	0.42	6	0.26	7
5	Lack of machinery	0.34	9	0.29	6
6	Limited site area	0.41	7	0.25	9
7	Lack of legal guidelines	0.78	2	0.65	1
8	Lack of post-demolition treatment facility	0.81	1	0.40	3
9	Lack of awareness of workers	0.46	5	0.26	7

TABLE 6: General information on surveyed buildings in Hanoi, Vietnam.

S/N	Name of building (ID)	Year of completion	Age of building (years)	Building function	Type of structure (Number of floors)	Site area (m^2)
1	Thikeco Office (OFC-01)	1967	51	Office	RC frame (04)	846
2	Office at 177 Trung Kinh (OFC-02)	2004	15	Office	RC frame (04)	1490
3	Office of Vietnam Women's Association (OFC-03)	1985	33	Office	RC frame (05)	2583
4	Ngo Sy Lien Secondary School (SCH-01)	1962	56	Institution	RC frame (05)	2728
5	Apartment Building at 93 Lang Ha (APT-01)	1987	33	Apartments	RC frame (05)	5519
6	No. 1 Truong Chinh (PVT-01)	2004	15	Private house	RC frame (02 + ½)	29.76
7	No. 104 Son Tay (PVT-02)	2007	12	Private house	RC frame (04)	58.9
8	No. 154 Tan Trieu (PVT-03)	2001	18	Private house	RC frame (03)	263
9	No. 52-54 Doi Can (PVT-04)	1989	30	Private house	RC frame (01)	318

Source: As-built portfolio, in conjunction with field observation and measurement

interior furniture, doors and windows, and – in some buildings – cladding structures. Most items were salvaged by the previous owner (in case of a private house) or by employees who used to work in that office, as this is a common practice in Vietnam. Cladding may consist of metal components (aluminum or steel) and glass panels that can also be salvaged. Some photos in this phase are presented in Figure 7.

During phase 2, workers use handheld tools to manually demolish slab panels and some structural members (Figure 8). Slab panels are demolished from bottom to top so that materials from upper floors can fall through. Members at the edge are removed first so that this part of the building is separated from adjacent structures, hence minimizing the effects (Figure 9).

Subsequently, heavy machines are deployed on the site to gradually demolish the rest of the building from top to bottom. During this process, water is sprayed continuously at high pressure from a hose to reduce the amount of dust (Figure 10).

Among DW generated from buildings, reinforcing steel has resale value. Thus, it is always separated from RC slabs, beams, and columns and then cut into smaller pieces using oxyacetylene cutting torches (Figure 11). CC-CB exists in a mixed state, sometimes even with other materials such as wood, plastic, gypsum, etc., and is transported out of the demolition site by trucks. DW transportation occurs in the time frame of 21:00–24:00 daily.

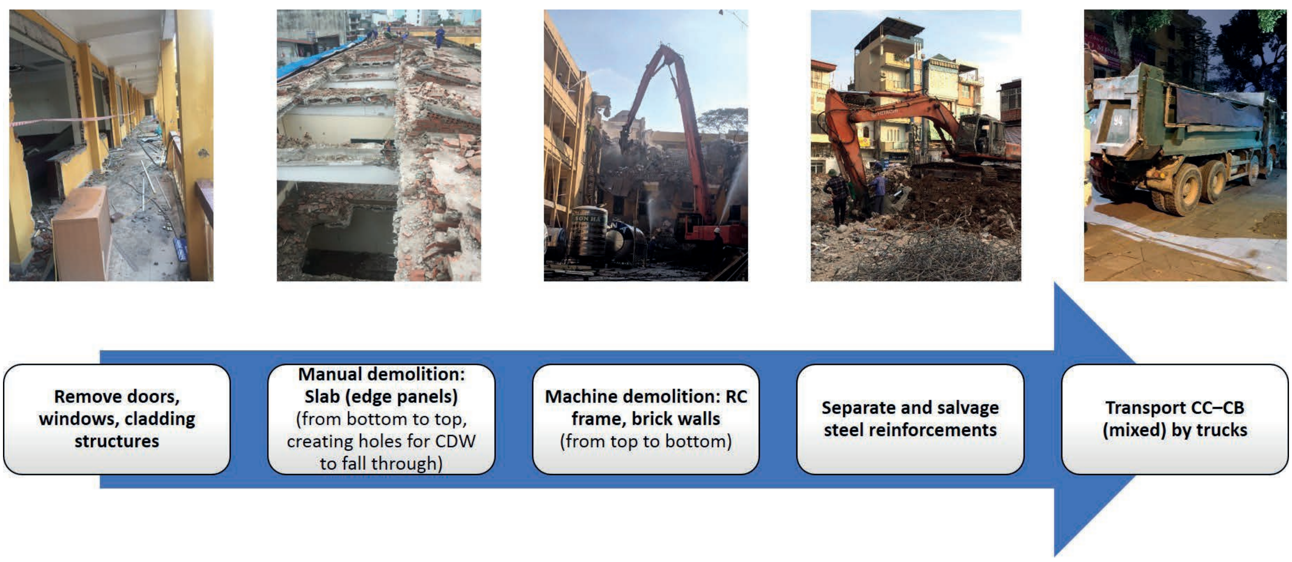

FIGURE 6: Typical flow of demolition work in Hanoi, Vietnam.

FIGURE 7: Removal of (a) sanitary ware; (b) interior furniture; (c) and (d) aluminum façade at OFC-01.

4.3 Summary of raw data and economic value of demolition works

Data of generated DW by category is summarized in Table 7, including CC-CB, steel (reinforcing steel, as well as other sources), aluminum, glass, and wood.

In Table 7, the economic value (EV) is determined as follows:

- The demolition cost in Vietnam is calculated by volume, and the unit price is 250,000 VND/m^3;
- Only the volume of CC–CB is employed for calculating the demolition cost because it makes up the bulk of the DW volume;
- The transportation fee of CC–CB is 40,000 VND/m^3.

The economic value of CC–CB, therefore, is:

$$EV_{CC-CB} = V_{CC-CB} \times (250{,}000 - 40{,}000) \quad [\mathrm{VND}] \tag{4}$$

Steel is sold at 12,000 VND/kg, while aluminum is sold at 30,000 VND/kg. Hence, the economic value of these two types of metal can be calculated by:

$$EV_{metal} = m_S \times 12{,}000 + m_A \times 30{,}000 \quad [\mathrm{VND}] \tag{5}$$

The economic value of the demolition work:

$$EV = EV_{CC-CB} + EV_{metal} \quad [\mathrm{VND}] \tag{6}$$

4.4 Generation rate of DW from surveyed buildings

Applying the formula presented in the previous section,

FIGURE 8: (a) Demolition by handheld tool; and (b) A worker manually pulling down an RC column.

FIGURE 9: (a) Hole for CDW to fall through; and (b) Edge slab panels demolished.

FIGURE 10: Machine demolition at (a) OFC-01; and (b) SCH-01.

FIGURE 11: Reinforcing steel being separated and recollected.

the authors determined the generation rate of two main types of DW from buildings: 1) CC–CB; and 2) steel. Detailed results are displayed in Table 8. Generation rates per unit floor area of CC–CB, and of steel are also visualized in the form of column charts in Figure 12 and Figure 13 (respectively).

TABLE 7: Amount of DW generated from surveyed buildings in Hanoi by category and estimated economic value.

S/N	ID	Days of demolition (days)	CC–CB (m^3)	Steel (ton)	Aluminum (kg)	Glass (m^2)	Wood (m^3)	Economic value (mil. VND)
1	OFC-01	23	900	250	1062	338	-	3220.86
2	OFC-02	37	1260	230	-	-	-	3024.6
3	OFC-03	32	1500	315	-	-	-	4095
4	SCH-01	38	4200	1450	-	-	-	18282
5	APT-01	70	6520	1050	-	-	-	13969.2
6	PVT-01	02	140	0.48	-	-	-	35.16
7	PVT-02	09	435	1.72	72	20	-	117.51
8	PVT-03	09	920	3	238	-	-	236.34
9	PVT-04	04	500	1.1	240	-	20	125.4

Source: Field observation, and interview with informant on-site

For structures that generate aluminum, its generation rate can be calculated by a similar formula:

$$G_A = \frac{m_A}{GFA} \quad \left[kg/m^2\right] \tag{7}$$

Subsequently, the generation rate of DW from surveyed buildings can be obtained by summing the three values of $G_{CC\text{-}CB}$, G_S, and G_A . G_{DW} as displayed in Figure 14.

$$G_{DW} = G_{CC-CB} + G_S + G_A \quad \left[ton/m^2\right] \tag{8}$$

In terms of the scale of the structure, it is clearly seen that large buildings (GFA of 2000 square meters and above) generate much less CC-CB per unit area than private houses (GFA usually less than 1000 square meters). The reason is that large-scale structures have longer spans, thus reducing the density of RC frame members. Furthermore, because these buildings function as an office or school, temporary partition walls are often employed in contrast with the permanent brick walls of small houses. However, the steel generation rate of large-scale structures is significantly higher than that of small-scale structures due to the fact that their significance requires a design with much more reinforcing steel content.

It is also noted that APT-01 possesses a CC-CB generation rate higher than that of other large-scale buildings (almost double) but still much lower than that of small houses. Therefore, it is possible to deduce that building function also affects the DW generation rate.

Considering the effect of building age, one can observe the generation rate of steel from three office buildings. It is obvious that the two recent RC offices were designed with less steel content than the one built in the 1960s (roughly 25% reduction). This reflects a change in the design standard, and even construction materials (old material versus new).

Finally, the chart of the total DW generation rate closely resembles that of CC-CB because it is the dominant category of DW in buildings. For offices, varies slightly at around 0.41 tons/m^2 (standard deviation equals 0.026), whereas for private houses, fluctuates widely with a mean value of 1.39 tons/m^2 (standard deviation equals 0.211).

5. FURTHER DISCUSSIONS ON FINDINGS OF THE INVESTIGATION

Through interview, it is revealed that there is no specialized license for demolition work in Vietnam. All contractors who registered license for construction work are also allowed to dismantle structures. Moreover, Vietnamese demolition contractors are currently relying on experience and

TABLE 8: Generation rate of DW from surveyed buildings in Hanoi.

S/N	ID	GFA (m^2)	CC–CB		G_S (kg/m^2)	G_A (kg/m^2)	G_{DW} (ton/m^2)
			m_{CC-CB} (tons)	G_{CC-CB} (kg/m^2)			
1	OFC-01	2392	747	312.29	104.52	0.44	0.42
2	OFC-02	2960	1045.8	353.31	77.70	0.00	0.43
3	OFC-03	4120	1245	302.18	76.46	0.00	0.38
4	SCH-01	9767.5	3486	356.90	148.45	0.00	0.51
5	APT-01	9030	5411.6	599.29	116.28	0.00	0.72
6	PVT-01	74.4	116.2	1561.83	6.45	0.00	1.57
7	PVT-02	235.6	361.05	1532.47	7.30	0.31	1.54
8	PVT-03	684	763.6	1116.37	4.39	0.35	1.12
9	PVT-04	318	415	1305.03	3.46	0.75	1.31

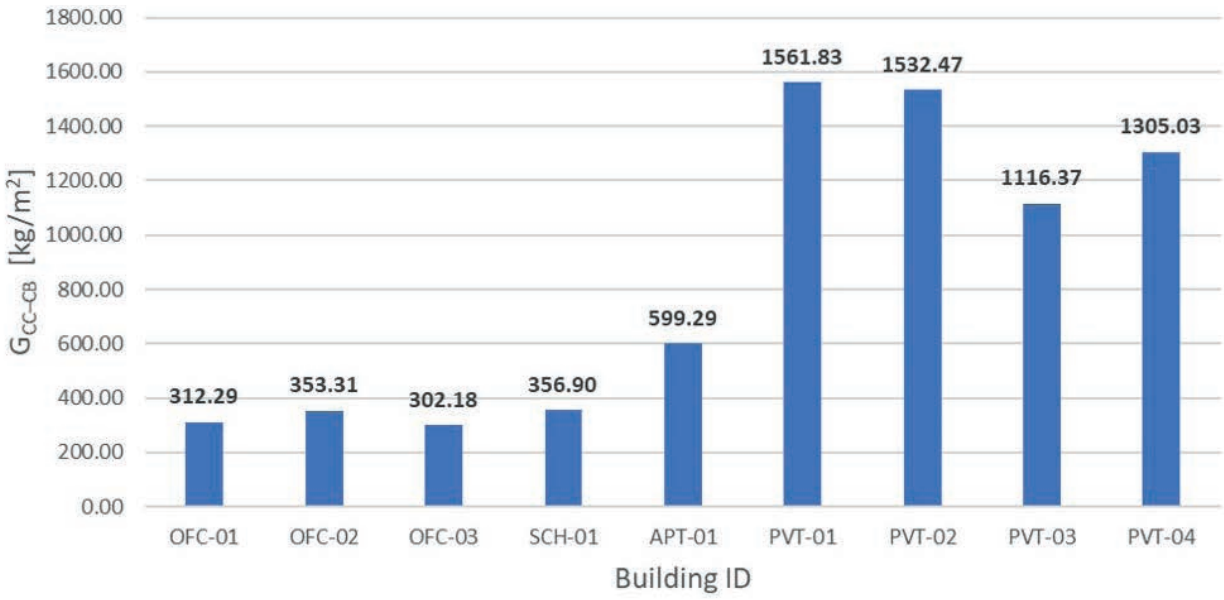

FIGURE 12: Variation of generation rate of CC–CB from surveyed demolition in Hanoi.

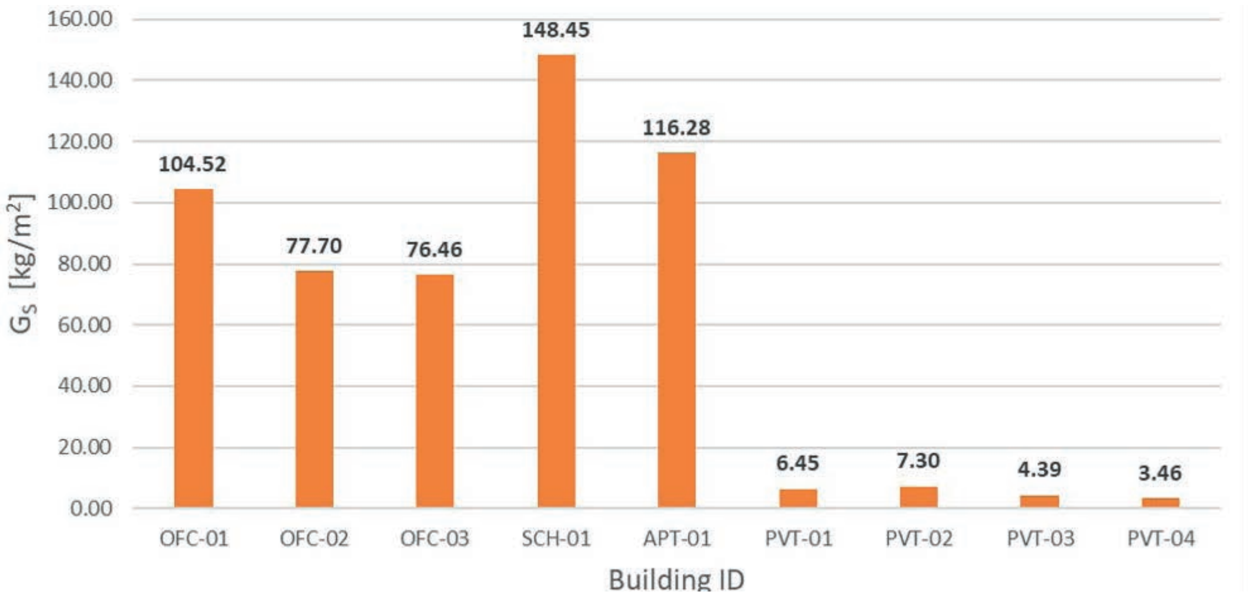

FIGURE 13: Variation of generation rate of steel from surveyed demolition in Hanoi.

personal network to carry out the work rather than proper guidelines and official associations. Being a separate type of work than construction and knowing the practice in countries such as Japan – where demolition contractors need to apply for a specialized license, it is possible to deduce that the management of CDW in Vietnam is unprofessional and lacking. This fact agrees with the low awareness of demolition contractors of current legislations on CDW, as the reason is simple: when operating a business based on experience and personal network, there is no need for legal document.

Many interviewees showed interest in the prospect of reusing and recycling of CDW (mainly CC–CB, as metal components are already making profit) and realized the economic attractiveness of this alternative. Currently, for every cubic meter of CC-CB, contractors have to pay a fee to dump their trucks (Section 4.3 explained in detail the calculation of EV). Furthermore, they are supposed to dispose CDW at official landfills specified by the authority (Hanoi People's Committee) which are limited in number, and are always filled up rather fast. Hence, they have to constantly look for new landfills (private ponds that need backfilling, or abandoned paddy fields…), or have to finally resort to illegal dumping. Should CC-CB is recycled, demolition contractors can sell it to the facility for additional profit while at the same time, they no longer have to worry about dumping sites. Therefore, despite "Cost increase" is ranked first in the list of challenges in implementing classification of CDW, demolition contractors are willing to adopt the ad-

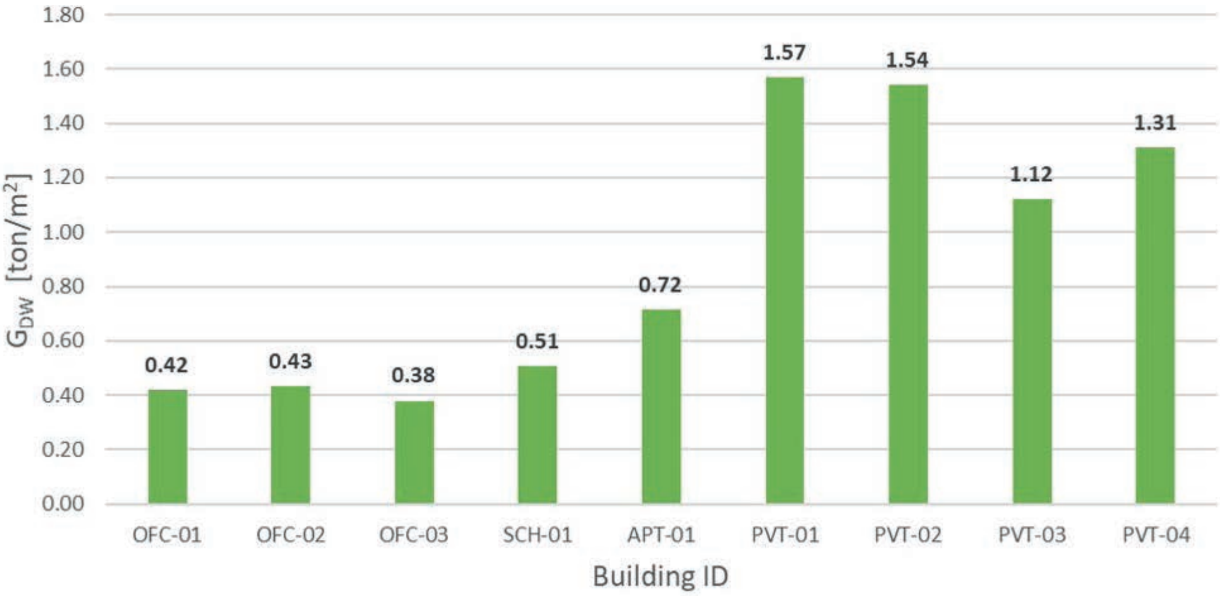

FIGURE 14: Variation of generation rate of DW from surveyed demolition in Hanoi.

ditional process of sorting. This is a concrete evidence to support the opportunites in recycling CDW mentioned in previous studies (Lockrey et al, 2016; Nguyen et al, 2018).

Case study in Hanoi enabled the authors to summarize the typical flow of demolition work, as well as the values of DW generation rate per square meter of GFA for two types of buildings: office and private house. This data could serve as an effective mean to predict the amount of DW generated prior to demolition of buildings of similar types. Combined with the data of new construction available at Departments of Construction of cities (new construction works must be registered with the authority, in which GFA is clearly indicated), it is potentially possible to project a good estimation of DW in the future. This provides an additional approach to the conventional mean of projection by statistical data and growth that are being applied by existing literature.

Compared to a number of neighboring countries, similar studies on the management and generation of CDW, as well as the feasibility of recycling and its economic attractiveness have been conducted much earlier. In China for instance, although dumping CDW at landfills is still the most popular solution due to its simplicity and low cost, it has a potential to cause serious hazards such as the regretable incident in Shenzhen, Southern China in December 2015 where a landslide due to ill-management of dumped CDW resulted in 73 deaths (Yang et al, 2017). Therefore, the central government has been encouraging research on CDW and promoting the recycle of CDW into construction materials (Xiao et al, 2012). It is strongly advisable that Vietnam adopt similar policies and strategies in CDW management in order to minimize the effect of this type of solid waste on the environment.

6. CONCLUSIONS

The paper provides insights into the CDW management situation and practices in Vietnam by conducting interviews with those who are directly involved in such a system – demolition contractors. The investigation first confirmed an alarming increment in the number of demolition works. Secondly, it was revealed that the legislation on CDW in Vietnam is not only lacking, but also ineffective. A feasible procedure to monitor and estimate DW generation was proposed and implemented with nine structures being demolished in Hanoi to quantify the amount of DW. The results yield promising remarks on the relationship between the generation rate and those three factors. Last but not least, data collected from the quantification survey is also crucial in establishing strategies to effectively manage CDW. Future studies may involve strategies for recycling CDW, applications of recycled CDW, feasibility study and business model of producing recycled materials from CDW, expansion of DW generation rate into a thorough study to cover CW and even more types of structures in Vietnam.

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MICRO X-RAY FLUORESCENCE IMAGING COUPLED WITH CHEMOMETRICS TO DETECT AND CLASSIFY ASBESTOS FIBERS IN DEMOLITION WASTE

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ABSTRACT

Asbestos was largely used in the past by several countries all over the world. From 1900 to 1990 asbestos-containing materials (ACMs) were produced in large amounts and mainly utilized to produce insulation, flame retardant materials, as well as to improve the mechanical and the chemical characteristics of construction materials. Its extensive use has therefore led to the presence of fibers in existing buildings and within the construction and demolition waste. A fast, reliable and accurate recognition of ACMs represents an important target to be reached. In this paper the use of micro X-ray fluorescence (micro-XRF) technique coupled with a statistical multivariate approach was applied and discussed with reference to ACMs characterization. Different elemental maps of the ACMs were preliminary acquired in order to evaluate distribution and composition of asbestos fibers, then samples energy spectra where collected and processed using chemometric methods to perform an automatic classification of the different typologies of asbestos fibers. Spectral data were analyzed using PLS-Toolbox™ (Eigenvector Research, Inc.) running into Matlab® (The Mathworks, Inc.) environment. An automatic classification model was then built and applied. Results showed that asbestos fibers were correctly identified and classified according to their chemical composition. The proposed approach, based on micro-XRF analysis combined with an automatic classification of the elemental maps, is not only effective and non-destructive, it is fast, and it does not require the presence of a trained operator. The application of the developed methodology can help to correctly characterize and manage demolition waste where ACMs are present.

1. INTRODUCTION

Asbestos is the common name used for two families of fibrous minerals of different crystallographic and chemical characteristics: serpentine (i.e. chrysotile: $Mg_3(Si_2O_5)(OH)_4$) and amphiboles (i.e. crocidolite: $Na_2(Fe^{2+}{}_3Fe^{3+}2)Si_8O_{22}(OH)_2$ and amosite: $Fe_7Si_8O_{22}(OH)_2$) (Lewis et al., 1996; Paglietti et al., 2016). They can all exist in several different crystalline forms, but only if characterized by a fibrous structure are classified as asbestos. The most used mineral in the industrial sector is chrysotile, as it is contained in almost 95% of all asbestos products and/or artifacts (Virta, R.L., 2005). Among the amphiboles, the most widely used mineral is crocidolite, followed by amosite (Bassani et al., 2007). Asbestos has been widely used in many applications for its technical properties (i.e. resistance to abrasion, heat and chemicals) (Gualtieri, 2017). However, despite its properties, asbestos is recognized as a hazardous material to human health and since 1980 it has been banned in many industrialized countries. The exposition of people to asbestos is quite huge. World Health Organization (WHO) report shows as about 125 million people are exposed to asbestos at the workplace. Every year, asbestos-related-tumors produce the death of about 100,000 people, several thousand related to asbestos exposure at home (Varkey, B., 1983; Olsen et al., 2011). Several solutions were explored to clean up ACMs (Yoshikawa et al., 2015; Zhai et al., 2014; Valouma et a., 2017) but first a preliminary separation of contaminated products from non-hazardous waste is required. Asbestos fibrils are generally very thin and may not all be resolved even by optical magnifications of 400–450×, and so, by eye what is seen as fibers are actually bundles of fibers (Harper et al., 2008). On present evidence fiber counts appear to provide a better index of hazard than respirable

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mass concentration. Considerations of respirability and biological activity together suggest that the counts should be of fibers in the size range. In most legislation a "standard" airborne fiber to be counted is a fiber with a lenght greater than 5 micron and width less than 3 micronwith an aspect ratio l/w greater than 3:1. (Walton, W. H.,1984). Asbestos containing material with fibers exposed on degraded surface, can be a possible sources of respirable asbestos fibers. Additionally, fiber identification techniques are crucial for environmental control in contaminated areas such as the proximity of an asbestos mine (Colangelo et al., 2011). The possibility to adopt fast and reliable analysis methods to detect and identify asbestos fibers in laboratory is of great interest in terms of safety, time and costs. Micro-Xray based systems are widely used in different fields of research and it is frequently used as a technique to identify elements in the laboratory and on portable systems. Imaging X Ray fluorescence is the evolution of point micro-XRF and allows to obtain spatial and compositional information simultaneously (Tsuji et al., 2005). This approach is actually applied in many different research areas, as: biology and medicine (Paunesku et al., 2006), cultural heritage (Rosi et al., 2004; Monico et al. 2011; Pronti et al., 2015; Capobianco et al., 2018), forensic research (Dhara et al., 2010; Nakanishi et al. 2008), natural science (Capobianco et al., 2018). In this work, benchtop micro-XRF was utilized to identify and classify asbestos fibers (Bonifazi et al, 2015; Bonifazi et al, 2018). The proposed strategy, based on the combined use of micro-XRF imaging and chemometric techniques, can be a valid and efficient analytical approach supporting the currently adopted asbestos recognition techniques, such as Fourier transform infrared spectroscopy (FT-IR) (De Stefano et al., 2012), Raman spectroscopy (Petriglieri et al., 2015), polarized light microscopy (PLM) (Lee et al., 2008) and scanning electron microscopy (SEM) (Gandolfi et al., 2016). All these techniques require the preparation of samples and usually allow punctual measurements and/or small areas mapping. The new generation of scanning XRF analytical units, based on confocal XRF method, realizes the best acquisition conditions, both in terms of speed and analytical data set reliability. For every acquired hyper-map, a XRF spectrum is associated to each pixel. Thus, an acquisition consists in a n×m matrix of spectra, where n and m are the number of pixels in the x and y direction, respectively (Figueroa et al., 2014). Aim of this work was to verify the possibility to utilize the confocal micro-XRF imaging-based approach as an analytical technique to perform an automatic detection and mapping of asbestos containing materials (ACMs), without the presence of an operator performing a preliminary identification/selection of the different energy ranges/peaks representative of a specific asbestos fiber. The main aim of this procedure was to provide laboratory scale analytical tests allowing the identification of asbestos fibers bundles, following a not-destructive and not-invasive approach automatically carried out. Following this approach it is thus possible to reduce the quantity of samples to be analyzed by classic analytical techniques, that are more sensitive but require a specific samples preparation, longer analytical time and allow the analysis of smaller samples area. Data where thus analyzed by chemometric techniques (exploration and classification methods) and the results compared with the maps of the elements obtainable by the classical approach (i.e. manual selection of the elements associated to each fiber).

2. MATERIALS AND METHODS

2.1 Experimental set up

Six different ACM samples were investigated, containing 3 different type of asbestos fibers (i.e. chrysotile, crocidolite and amosite) (Figure 1 and Table 1). The ACM fragments were sampled from different contaminated sites by the Department of New Technologies for Occupational Safety of Industrial Plants, Products and Anthropic Settlements, (National Institute for Insurance against Accidents at Work - INAIL). Sample collection was performed following the general guidelines that have to be adopted when asbestos remediation actions have to be performed in superfund sites (INAIL, 2010). XRF analyses according to the rules reported in "Fact Sheet: Recognition and characterization of ACM by micro-XRF" (INAIL, 2020). Selected areas of the sample surfaces were used to acquire the micro-XRF maps. More in details, sample areas of Figures 1a, 1b and 1c were utilized to perform calibration, whereas those shown in Figures 1d, 1e and 1f were utilized to validate the classification model. The micro-XRF based elements mapping was performed at Raw-Ma Lab (Raw materials Laboratory) - Department of Chemical Engineering, Materials & Environment (Sapienza - University of Rome, Italy. A benchtop spectrometer (M4 Tornado, Bruker®), equipped with a Rh X-ray tube with poly-capillary optics as the X-ray convergence technique and XFlash® detector providing an energy resolution better than 145 eV and 5 filters, was utilized (Guerra et al., 2013). The whole spectra comprised 4096 channels with a spot size of approximately 30 µm. Spectrum energy calibration was daily performed before each analysis batch by using zirconium (Zr) metal (Bruker® calibration standard). The sensitivity of µXRF is determined by the excitation probability of the sample and the peak to background ratio. The background intensities were directly computed by the equipment (ESPRIT Bruker® software). The sample chamber can be evacuated to 25 mbar and, therefore, light elements such as sodium can be measured (Nikonow et al., 2016). Constant exciting energies of 50 kV and 500 µA, were adopted for acquisition. The set-up mapping acquisition parameters were a pixel size of 30 µm and an acquisition time, for each pixel, of 10 milliseconds. Through this experimental set-up is possible to map bundles of asbestos fiber inside matrix until a resolution of 30 micron in a wide scanning area (max acquisition area 19x16x12 cm for each sample). Spectral data (i.e. hyper-maps) analysis was carried out adopting chemometric methods, using the PLS_Toolbox (version 8.6, Eigenvector Research, Inc.) running inside MATLAB (version 9.3, The Mathworks, Inc.). Starting from the samples outlined in Figure 1, two mosaic data images were built (Figure 2) in order to define the calibration (Figure 2a) and validation (Figure 2b) data set.

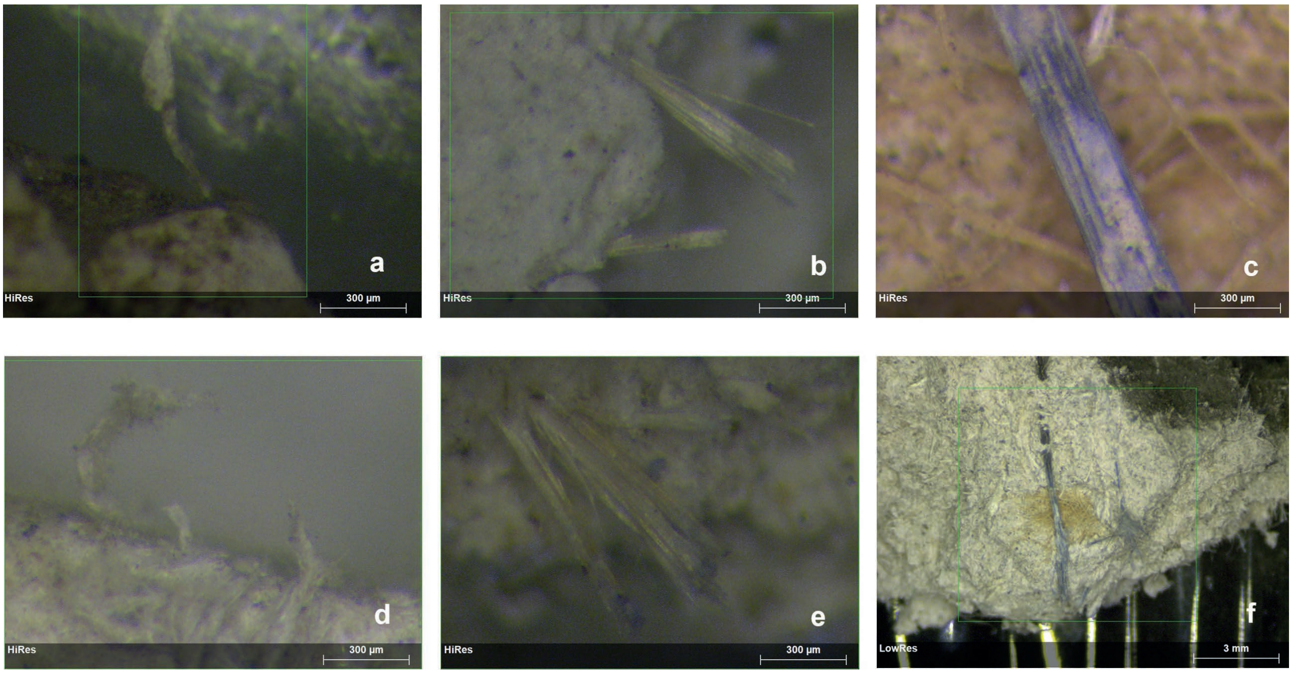

FIGURE 1: Asbestos containing materials (ACM) microscopic images: chrysotile (a and d), amosite (b and e) and crocidolite (c and f). Sample images a, b and c have been utilized for calibration and d, e and f for validation.

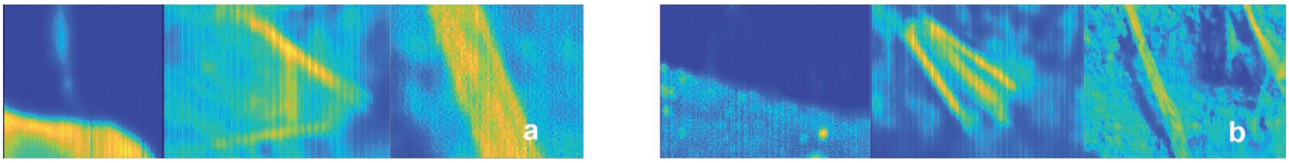

FIGURE 2: Mosaic data image of ACM samples used for calibration (a) and validation (b) dataset, respectively. a: mosaic image obtained by the combination of the images reported in Figure 1a, 1b and 1c. b: mosaic image obtained by the combination of the images reported in Figure 1d, 1e and 1f.

2.2 µXRF: acquisition and data handling

The experimental procedure was defined and implemented in two steps. The 1st step was finalized to the acquisition of the hyper-maps and the further XRF peaks deconvolution in order to identify the different asbestos minerals following the classical expert-based approach. The 2nd step was addressed to energy spectra acquisition and handling to define an "automatic" chemometric based classification model.

2.2.1 Step 1: hyper-maps acquisition and XRF peak deconvolution

ACM samples were acquired by µXRF in order to build, as already mentioned, the element maps. Specific Areas in the Sample (SAS) of the ACM were then analyzed in order to quantify the element concentration and identify asbestos fibers (Figure 3).

TABLE 1: Asbestos samples selected to perform the XRF based recognition/classification procedures.

Samples	Description
Samples containing chrysotile	Fragment of corrugated sheets (Materit)
Samples containing amosite	Fragment of flat slab (Materit)
Samples containing crocidolite	Fragment of a water tank

2.2.2 Step 2: Definition of the calibration dataset and identification of elements by soft independent modelling by class analogy (SIMCA)

Calibration dataset were acquired by micro-XRF to build an automatic classification model able to recognize the different asbestos fibers without any human based investigation. A set of 3 asbestos fibers clearly identified in the ACM samples was used as training dataset to build the classification model (Figure 2a). The classification model was then validated utilizing the ACM samples (Figure 2b). Spectral data analysis was preliminary addressed to explore and to evaluate the quality of the acquired information to be utilized for the further classification model definition, design, implementation and set up. To reach these goals, a preliminary Principal Component Analysis (PCA) and a further Soft Independent Modelling by Class Analogy (SIMCA) was applied. PCA is the most utilized multivariate data analysis method for exploratory data handling, outlier detection, rank (dimensionality) reduction, graphical clustering, classification, regression, etc. (Bro et al., 2014). It

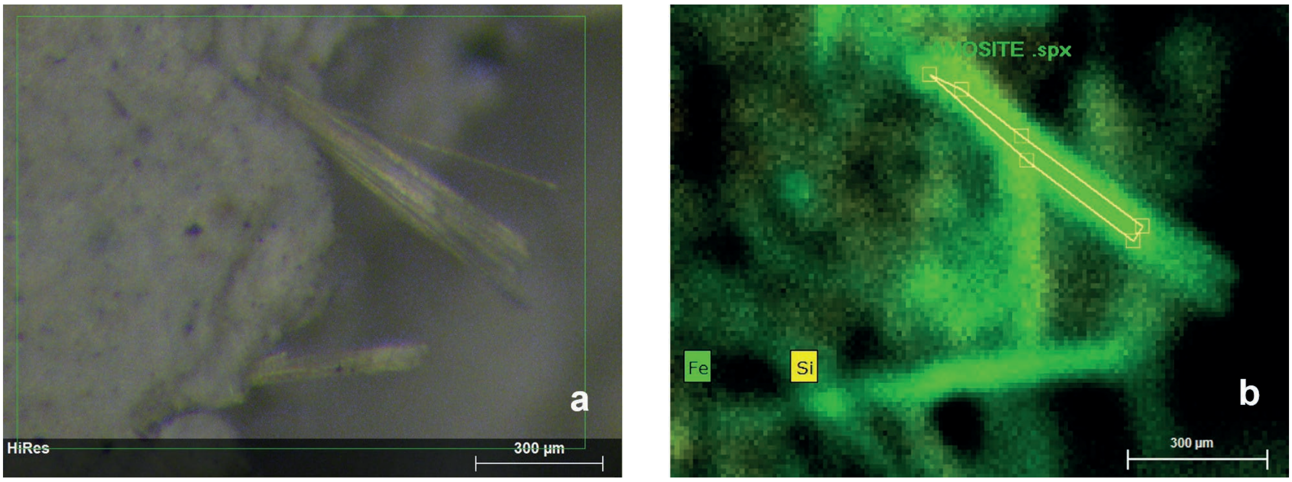

FIGURE 3: Example of procedure adopted to identify asbestos minerals with reference to amosite. a: source image and b: corresponding Fe and Si maps, where a Specific Areas in the Sample (SAS) was selected in order to verify, through the corresponding energy spectra deconvolution, the qualitative and semi-quantitative element presence, characteristic of the target asbestos mineral (i.e. amosite).

was used to decompose the "pre-processed" spectral data into several principal components (PCs) (linear combinations of the original spectral data) embedding the spectral variations of each collected spectral data set. According to this approach, a reduced set of factors is produced and used for discrimination, since it provides an accurate description of the entire dataset. The first few PCs, resulting from PCA, are generally utilized to analyze the common features among samples and their grouping: samples characterized by similar spectral signatures tend to aggregate in the score plot of the first two or three components. Spectra could be thus characterized either by the reflectance at each wavenumber in the wavenumber space, or by their score on each PC in the PC space (Bro et al., 2014). Samples characterized by similar spectra, belonging to the same class of products, are grouped in the same region of the score plot related to the first two or three PCs, whereas samples characterized by different spectral features will be clustered in other parts of this space. Starting from PCA, different energy spectra pre-processing was sequentially applied, that is: Baseline, for spectra background subtraction, Normalize and Probabilistic Quotient Normalization (PQN), for spectra normalization and, finally, Mean centering (MC).

SIMCA is one of the most commonly used class modelling techniques for the classification of spectral data with many applications in different sectors, such as pharmaceutical (Celli et al., 2018), food (Nieuwoudt et al., 2004), biology (Oust et al., 2004) and medicine (Krafft et al., 2006). In SIMCA based modelling unknown samples are compared to the PCA class models and assigned to the class according to their analogy with the calibration samples (Brereton, 2003). SIMCA classifies objects into the category whose principal component model best reproduces the data. Only data points which are members of a given category are used in determining the model functions for that category. The importance of each feature in classification is determined by its contribution to the category covariance matrices. Therefore, once the model is obtained, it can be applied to an entire hypercube and for the classification of new hypercubes. The result of SIMCA, applied to the hyperspectral images, is a "prediction map," where the class of each pixel can be identified using color mapping.

The confusion matrix, reporting the Positive Predictive Value, Negative Predictive Value, Accuracy, False Discovery Rate and False Omission Rate, was also computed in order to evaluate the quality of the model. The possible results in prediction being: True Positive (TP), that is asbestos spectrum correctly identified, False Positive (FP), that is asbestos spectrum incorrectly identified, True Negative (TN), that is asbestos spectrum correctly rejected and False Negative (FN), that is asbestos spectrum incorrectly rejected. The quality of prediction results can be evaluated taking into account the values of Sensitivity, Specificity, Positive and Negative Predictive Value, False Discovery Rate, False Omission Rate and Accuracy parameters obtained for the classification model. Sensitivity measures the proportion of actual positives that are correctly identified as such, while, Specificity measures the proportion of actual negatives that are correctly identified as such (Sharma et al., 2009). Positive and Negative Predictive Values are the proportions of positive and negative results in statistics and diagnostic tests that are true positive and true negative results, respectively (Fletcher et al., 2012). The False Discovery Rate is the proportion of the spectra with a known positive condition for which the test result is negative (Lage-Castellanoset al., 2010). False Omission Rate measures the proportion of false negatives which are incorrectly rejected (Lage-Dresselhaus al., 2002). Accuracy is the degree of correspondence of the theoretical data with the real data (Linnet al., 2012). Other parameters have been also computed, they are reported in the following, that is:

- sensitivity is calculated using the following formula:

  Sensitivity=TP/(TP+FN) (1)

- specificity is calculated using the following formula:

  Specificity = TN/(TN+FP) (2)

- positive predictive value is calculated using the following formula:

  Positive predictive value = TP/(TP+FP) (3)

- negative predictive value is calculated using the following formula:

  Negative predictive value = TN/(TN+FN) (4)

- false discovery rate is calculated using the following formula:

  False discovery rate = FP/(FP+TP) (5)

- false omission rate is calculated using the following formula:

  False omission rate = FN/(FN+TN) (6)

- accuracy is calculated using the following formula:

  Accuracy = (TP+TN)/(P+N) (7)

3. RESULTS AND DISCUSSION

Results and discussion are reported in the following, presenting and comparing the classical human based micro-XRF mapping approach and the proposed automatic one based on SIMCA classification.

3.1 Element maps of asbestos fibers

The results of the semi-quantitative analysis, as resulting from energy spectra deconvolution, are summarized in Table 2, in terms of detected elements inside the different SAS. As shown in Table 2, the SAS analysis of fibers detected on the map allows to identify the elements with the highest concentration and their relative ratios. Light elements with low mean concentration (i.e. sodium) were not detected in map mode probably due to the reduced acquisition pixel per time (i.e. 10 milliseconds).

The chrysotile spectrum was characterized by a Mg concentration greater than those belonging to amosite and crocidolite. The amosite shows a higher Fe concentration than that detected in crocidolite. Crocidolite spectrum is characterized by the presence of Fe and small quantity of Mg. These physical characteristics and their relative values are useful but not sufficient enough to define the presence of asbestos fibers in the complex matrix. The elemental maps of the examined samples shown in Figure 4, summarize the complexity of the analyzed data. For this reason, in order to isolate the spectral signatures of the different fibers and to use them to create a predictive model, a PCA of the identified fibers was carried out. The results shown in Table 2 are not the typical compositions of pure asbestos minerals like chrysotile, amosite or crocidolite. The elements as Calcium (Ca), Aluminium (Al), Sulfur (S), Potassium (K), copper (Cu) and Titanium (Ti), were related to the matrix. The spectral compositions of the asbestos fibers shown in Table 2 was influenced by the surrounded matrix of the demolition waste material. As a consequence, in order to obtain a representative calibration dataset for the classification model, it was necessary to use asbestos fibers coming from fiber of different types of waste product containing ACM.

TABLE 2: Results of the semi-quantitative determination carried out by Energy Dispersive micro-XRF on ACM samples chracterized by a cement matrix.

	Chrysotile (A)		Amosite (B)		Crocidolite (C)		Chrysotile (D)		Amosite (E)		Crocidolite (F)	
Element	Normalize wt.%	wt.% (Sigma)	Normalize wt.%	wt.% (Sigma)	Normalize wt.%	wt.% (Sigma)	Normalize wt.%	wt.% (Sigma)	Normalize wt.%	wt.% (Sigma)	Normalize wt.%	wt.% (Sigma)
Ca	52.79	2.17E-02	30.49	8.79E-02	53.41	1.19E-01	22.28	1.55E-03	10.93	3.57E-03	44.60	7.85E-02
Si	29.84	3.28E-02	21.27	2.04E-01	13.52	3.78E-02	37.50	1.76E-02	7.05	7.72E-03	16.79	5.77E-02
Fe	2.55	7.08E-05	41.54	1.47E-01	27.93	2.92E-02	5.56	1.04E-04	74.90	1.38E-01	32.21	3.79E-02
Mg	9.11	5.84E-03	1.06	2.43E-03	2.65	2.92E-03	15.51	5.81E-03	0.13	1.08E-04	3.38	4.01E-03
Al	3.27	8.39E-04	0.26	2.13E-04	1.02	4.40E-04	2.58	2.73E-04	0.02	6.39E-06	1.10	4.93E-04
S	0.42	1.57E-05	1.18	7.82E-04	0.71	1.32E-04	10.25	1.38E-03	0.11	5.40E-06	1.05	2.44E-04
P	1.29	5.39E-05	0.45	6.77E-05	0.23	9.68E-06	1.51	3.85E-05	0.18	6.46E-06	0.25	9.09E-06
Ti	0.24	2.93E-06	0.12	6.48E-06	0.15	2.74E-06	2.00	2.47E-05	0.02	1.84E-07	0.16	2.40E-06
Mn	0.16	1.21E-06	3.45	1.23E-03	0.08	1.20E-06	0.21	1.18E-06	6.22	1.14E-03	0.08	8.89E-07
Sr	0.02	9.68E-08	0.13	6.57E-06	0.15	2.44E-06	0.05	3.18E-07	0.39	1.49E-05	0.14	1.90E-06
Zn	0.01	5.76E-08	0.00	1.22E-07	0.06	6.15E-07	2.38	2.69E-05	0.03	5.02E-07	0.14	1.49E-06
Cr	0.05	3.31E-07	0.03	1.06E-06	0.04	4.81E-07	0.18	9.41E-07	0.01	5.61E-08	0.03	2.68E-07
P	0.24	1.18E-05	-	-	-	-	-	-	-	-	0.01	3.79E-07
Cu	-	-	-	-	0.04	3.15E-07	-	-	0.03	2.19E-06	0.05	3.75E-07

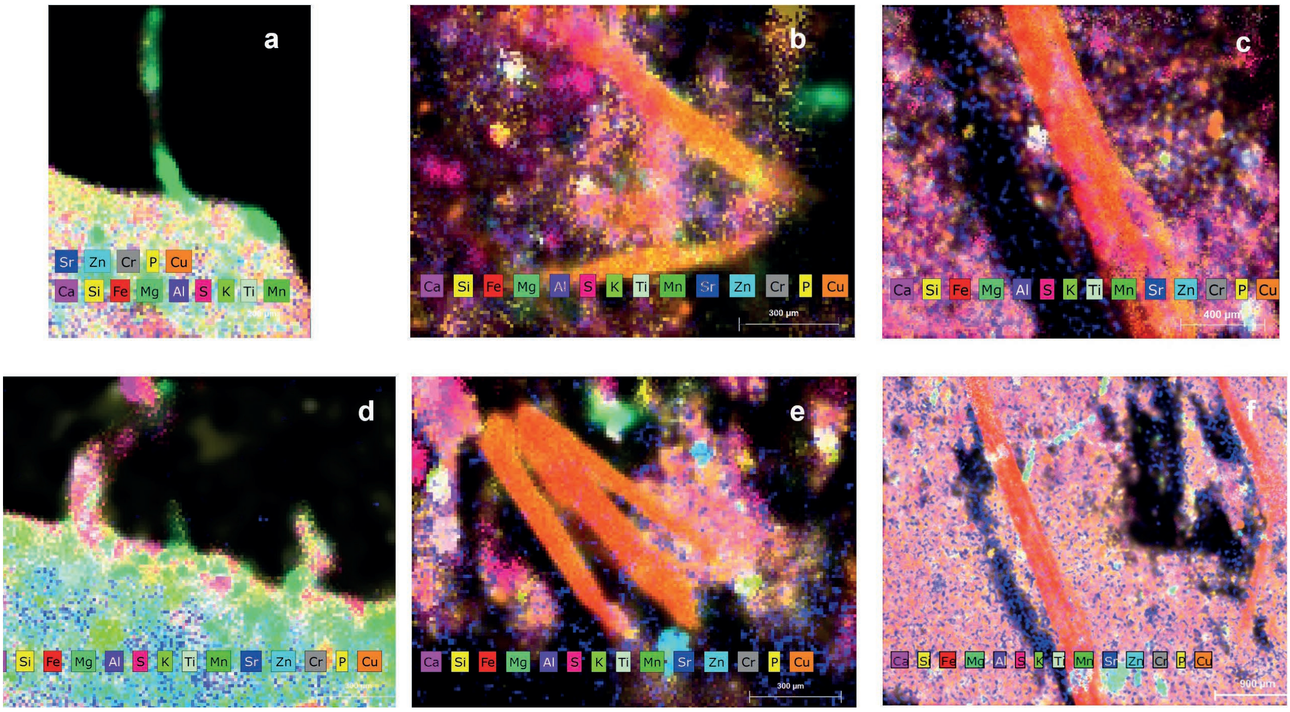

FIGURE 4: Elemental maps as resulting from the micro-XRF analysis carried out on samples. In detail chrysotile (a and d), amosite (b and e) and crocidolite (c and f).

3.2 PCA of calibration samples and identification of fibers by SIMCA

The energy spectra resulting from calibration are characterized by several peaks according to the emission of a photon quantum (fluorescence radiation), related to the energy difference between samples inner and outer shell. To emphasize the spectral characteristics of all the detected elements, "only" the mean spectra between 0 and 10 KeV have been considered, processed and mean centered (Figures 5a and 5), before the application of pre-processing.

A mosaic procedure was applied to obtain a single hypercube for calibration dataset. To emphasize the difference between the various peaks, the following pre-processing were used before the application of PCA: Baseline, Normalize, PQN and MC (Figure 6a and 6b).

The PCA score plot allows identifying three distinct groups according to their spectral signature. The score plot (PC1-PC2) reported in Figure 7a shows a good separation (i.e. distinction) of all the elements, as well as a good uniformity for each class. The loadings of PC1, PC2 (Figure 7b) show, in the region between 1.1 KeV and 7 KeV, the high variance of data, as a consequence 3 principal components are necessary to explain the 93.78% of the calibration dataset variance.

The selected energy spectra have been thus adopted as training dataset and a SIMCA model was built. The obtained values of sensitivity and specificity are shown in Table 3. The Sensitivity estimates the model ability to measure the number of samples of a given type correctly classified as that type. The specificity estimates the model ability to measure the number of samples not of a given type correctly classified as not of that type. Sensitivity and specificity can be assumed as model efficiency indicators: the more the values are close to one, the better the modelling is. In this study, the obtained values for sensitivity and

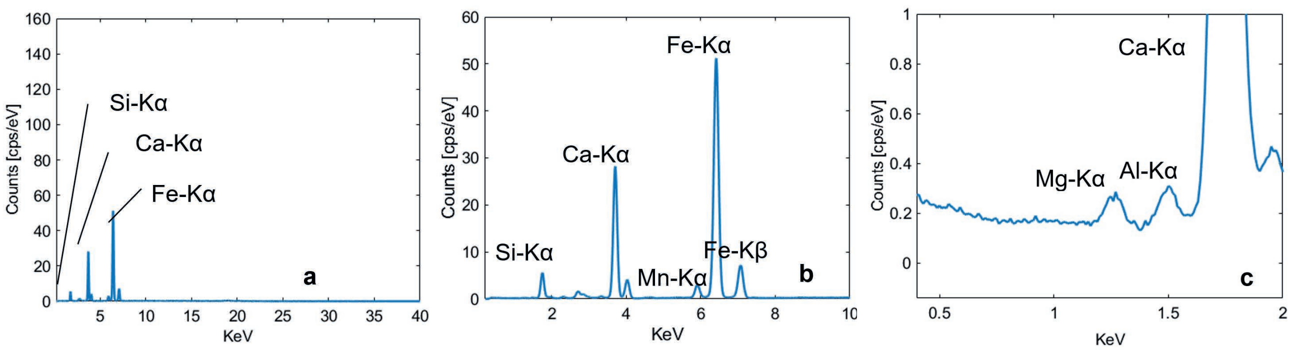

FIGURE 5: Raw energy spectrum of a generic SAS, before (a) and after (b) channels selection (i.e. 0-10 KeV). Detail of Mg Kα spectra area (c).

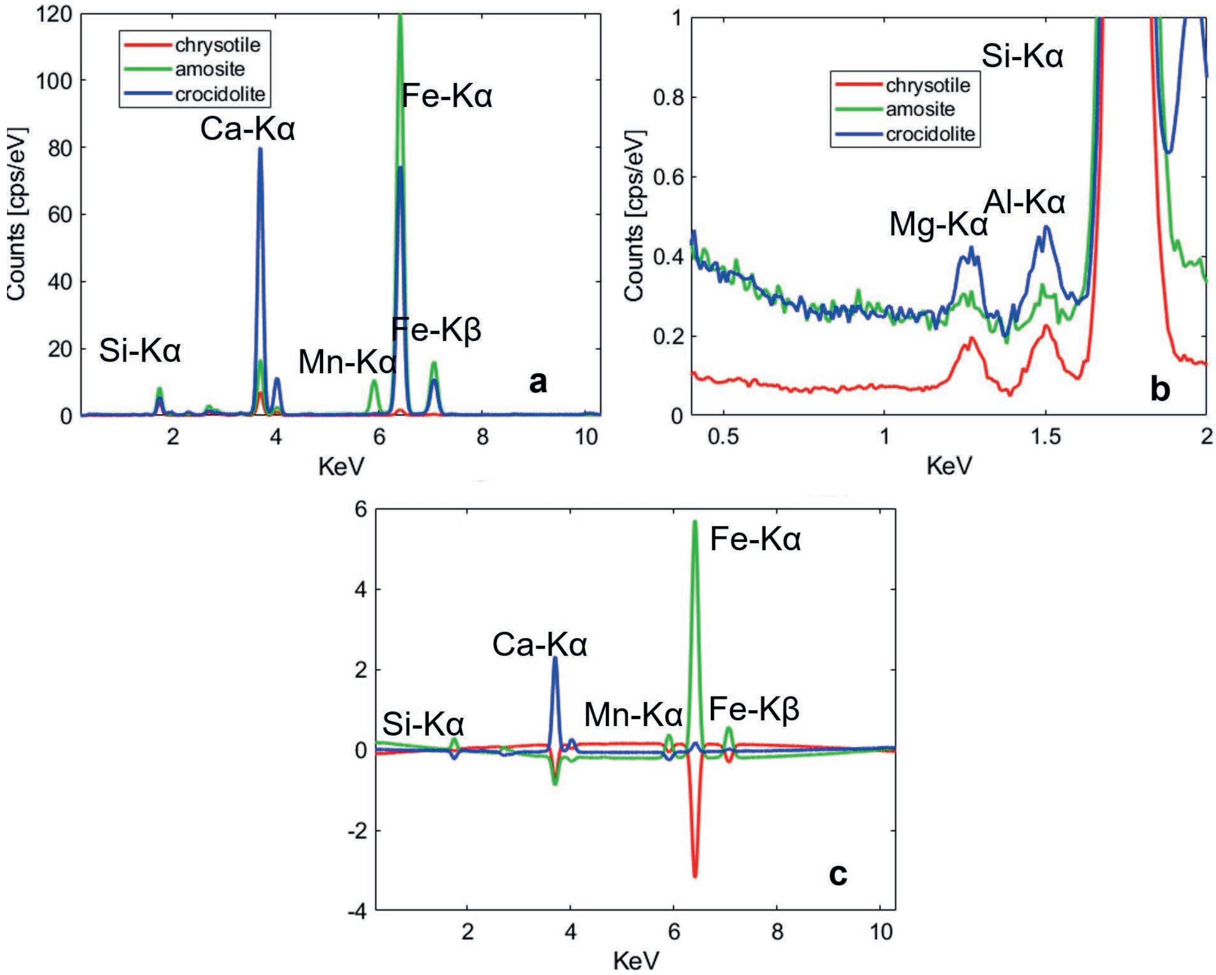

FIGURE 6: Raw (a), detail of Mg Kα spectra area (b) and pre-processed spectra (c), as resulting from the sequential application of different pre-processing algorithms: PQN, Baseline (Automatic Weighted Least Squares), Normalize and Mean Center.

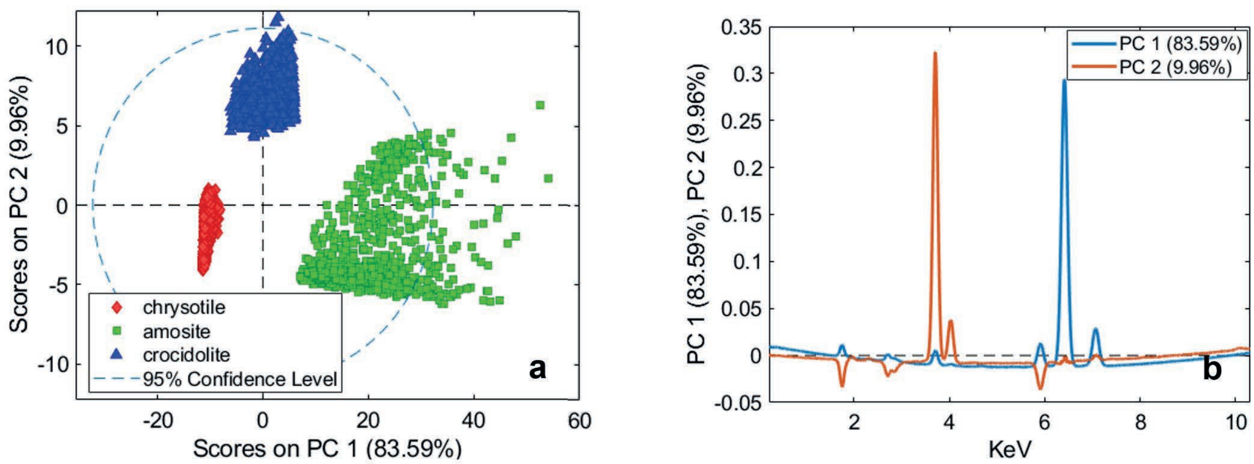

FIGURE 7: PCA score plot (a) e loading plot (b) of calibration dataset.

specificity are very good. To verify its classification ability, the built SIMCA model was applied to the validation of ACM samples data set. Sensitivity and specificity also in validation assume high values (Table 3).

The results in terms of prediction (i.e. "Pred Probability") are shown in Figure 8: the class with the highest probability belongs to the asbestos. The obtained results are very good for all the investigated elements, being comparable with those obtained following the classical "instrument-men-driven" approach. Misclassifications sometimes occur, but they are mainly due to border effect. The prediction of the chrysotile is good. Chrysotile fibers are well iden-

TABLE 3: Sensitivity and specificity for the SIMCA based built model. Cal: calibration; CV: cross-validation and PRED: prediction.

	Chrysotile	Amosite	Crocidolite
Sensitivity (Cal)	1.00	0.98	1.00
Specificity (Cal)	0.99	1.00	1.00
Sensitivity(CV)	0.84	0.85	0.90
Specificity (CV)	1.00	1.00	1.00
Sensitivity (PRED)	0.84	0.99	0.82
Specificity (PRED)	0.99	0.88	1.00

tified, despite the presence of high noise due to the uneven surface characteristics (i.e. de-focusing effect). The identification of amosite and crocidolite is also quite satisfactory. Only few pixels are misclassified.

The results of the confusion matrix in prediction are shown in Table 4. They show good a predictive capability of the model with a value of Positive and Negative Predictive Value closer to one and never under 0.86.

The low value of False Discovery and Omission Rate highlights the low number of spectra which are incorrectly classified with a range value between 0.01 and 0.14. In general, the Accuracy of each asbestos class prediction were over 0.91.

4. CONCLUSIONS

The study was carried out to investigate the utilization of chemometric procedures, based on processing of data-

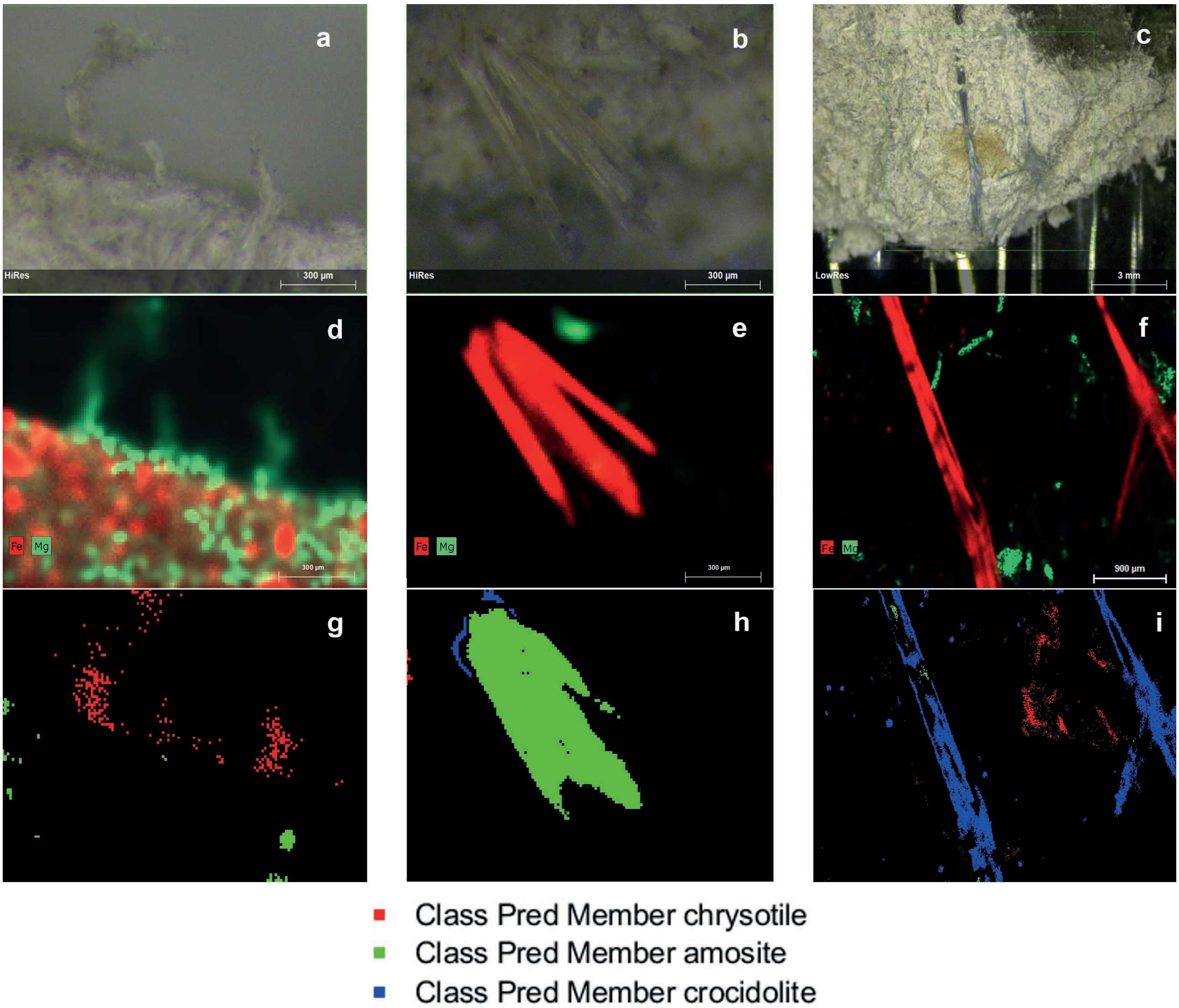

FIGURE 8: Microscope image of the ACM samples: chrysotile (a), amosite (b) and crocidolite (c). Micro-XRF map of iron and magnesium referred to chrysotile (d), amosite (e) and crocidolite (f). SIMCA prediction results obtained for chrysotile (g), amosite (h) and crocidolite (i).

TABLE 4: Positive predictive value, negative predictive value, accuracy, false discovery rate and false omission rate for the SIMCA prediction results.

	Chrysotile	Amosite	Crocidolite
Positive Predictive Value	0.99	0.89	0.99
Negative Predictive Value	0.86	0.99	0.84
Accuracy	0.92	0.94	0.91
False Discovery Rate	0.01	0.11	0.01
False Omission	0.84	0.99	0.82
Rate	0.14	0.01	0.16

set generated by micro-XRF, in order to perform a preliminary laboratory scale automatic check on the presence of asbestos fibers in cement matrix. More in detail, SIMCA, after PCA, was applied to build a model able to recognize/classify asbestos fibers starting from the reference energy spectra representative of the different asbestos minerals. The proposed combined chemometric micro-XRF approach presents many advantages: it is objective, and the classification model does not require an expert user for the interpretation of results. In fact, the final output of the model was a false color map assigning a color for each asbestos type. In the future, using a robust calibration dataset, this procedure could be applied to different anthropogenic ACM waste materials at laboratory scale in order to reduce the quantity of material to be analyzed with consolidated analytical techniques, requiring both sample preparation and longer analysis time. The procedure, after the preliminary model recognition set up, is easy to implement and it is characterized by low operative costs, being the procedure totally software, especially if compared with classical methods usually requiring sample pre-treatment and longer analytical time (i.e. optical microscopy and SEM-EDX). Further studies will be addressed to a systematic application of asbestos recognition in order to perform not only a qualitative control of the different ACM samples, but also to characterize different types of asbestos fibers in different types of matrix. Following this approach, it will be thus possible to design more efficient and specialized strategies for the identification of asbestos fibers using imaging XRF techniques.

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MONITORING OF HEAVY METALS, EOX AND LAS IN SEWAGE SLUDGE FOR AGRICULTURAL USE: A CASE STUDY

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ABSTRACT
Subsequent to the increasing diffusion of wastewater treatment, particularly in high- and middle-income countries, the sewage sludge generated should be treated and valorised in an ecological and economic way, thus contributing to the circular economy. In this study, the monitoring of Heavy Metals (HM), Extractable Organic Halogens (EOX) and Linear Alkylbenzene Sulphonate (LAS) in sewage sludge from 10 different wastewater treatment plants located in Friuli Venezia Giulia (Italy) was reported, and their macronutrient content provided. The obtained results showed, for all tested samples, that HM content in sewage sludge was below the maximum permitted limits provided for by Italian and European regulations for agricultural reuse. Comparison with a similar monitoring campaign carried out in 2006 revealed how, while wastewater treatment plants efficiently resolved water pollution, they accumulated heavy metals and other persistent toxic compounds in sludge, thus restricting their potential reuse. Consequently, consistent and regular sludge monitoring should be undertaken to prevent soil and groundwater contamination. These outcomes could be of particular relevance for the future perspective of agricultural reuse of sewage sludge in waste management practices.

1. INTRODUCTION

From the perspective of sustainable agriculture, reuse of sewage sludge is fundamental due to nutritional and organic matter content (Fijalkowski et al., 2017) and low cost. However, the persistent presence of a series of organic contaminants and toxic elements in sewage sludge may result in environmental and health issues (Anjum et al., 2016). Wastewater treatment plants (WWTPs) are in continuous development, with ongoing construction of new treatment units or upgrading of existing facilities. Sludge management has become one of the most critical environmental issues in the sector. Indeed, nowadays, approximately 50% of total operating costs in WWTPs are incurred in sludge treatment (Quian et al., 2016). In Europe in 2015, approx. 9.5 million tons dry matter of sewage sludge were produced (Eurostat, 2018), thus requiring appropriate disposal.

Several options are available for use in the final disposal of sewage sludge, including energy and resource recovery (Abis et al., 2018; Di Maria et al., 2018; Gherghel et al., 2019; Haarlemmer et al., 2018). Specifically, treated sludge, when applied as fertiliser and soil conditioner are a source of nutrients for the soil, (Yoshida et al., 2018; Ashekuzzaman et al., 2019), although the risks of soil contamination and pathogen transmission should be carefully considered (Singh and Agrawal, 2008; Tsybina and Wuensch, 2018). Council Directive 91/271/EEC encouraged the land application of sewage sludge (European Commission, 1991) due to the related fertilizing and conditioning properties for agricultural soil. However, this practice may also lead to environmental and health risks, due to accumulation of persistent organic contaminants, toxic elements (Valentin et al., 2013) and heavy metals (Chen and Hu, 2019) contained in sewage sludge. When sludge produced by urban WWTPs is used as fertilizer in agriculture, a detailed specification of the properties and quality of sludge is required (USEPA, 1995; American Society of Civil Engineers and American Water Works Association, 1996) to prevent the onset of health and environmental issues. In a study by Laura et al. (2020), decision support framework was used to analyse different strategies for sewage sludge handling in Latin America; a pool of different parameters was considered, including economic, social, environmental and technological

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aspects. Given local peculiar sludge characteristics, such as low HM content and high pathogenic contamination, the application of a composting process before agricultural reuse was recommended as the best solution for managing sludge valorisation (Laura et al., 2020). A study by Collivignarelli et al. (2020), investigated a sewage sludge management chain in the Pavia province, Italy, highlighting the most critical issues for agricultural application, including a need to promote global sludge reduction in WWTPs, the production of sludge with standardized characteristics, together with a better selection of materials in dedicated sludge treatment plants and an increased control of agricultural soil response (Collivignarelli et al., 2020).

A major cause of concern is related to the toxicity of heavy metals even at trace concentrations (order of magnitude of ng/L), in particular Cd, given its high bioavailability (Hu et al., 2017). Heavy metals such as lead, cadmium, mercury, nickel and chromium (Pires and Mattiazzo, 2003; Singh et al., 2004; Hargreaves et al., 2018) in municipal wastewater originate from household sewage, industrial wastewater or urban runoff (Sorme and Lagerkvist, 2002). The majority of these toxic metals accumulate in the sludge, since only a small amount is released with the treated final effluent (Sorme and Lagerkvist, 2002). Once sludge from WWTPs are applied to the soil, the degradation of organic compounds in sewage sludge affects the availability of heavy metals, inducing accumulation in plant biomass, one of the primary elements of the human food chain (Gondek et al., 2014). A recent study by Romanos et al. (2019) highlighted that to reduce metals and pathogenic microorganisms in sludge in line with strict legislative limits, further processing (such as composting) may be required.

Recently, particular focus has been concentrated on organic compounds present in domestic and municipal wastewater due to their accumulation in sludge. The use of some parameters, such as Extractable Organic Halogen (EOX) (Kannan et al., 1999; Niemirycz et al., 2005) and Linear Alkylbenzene Sulphonate (LAS), is particularly meaningful, in view of their good representation of a general wide-ranging organic pollution in sludge.

The importance and usefulness of EOXs in the assessment of environmental quality have been demonstrated in a series of studies (Rodziewicz et al., 2004; Contreras Lopez, 2003; Goi et al., 2006; Rizzardini and Goi, 2014; Braguglia et al., 2014); however, a limited number of investigations have addressed the issue of EOX content in sludge. Sewage sludge application in agriculture represents the main source of LAS entrance to agricultural soil (Jensen, 1999). Sludge concentrations of the latter are strictly dependent on sludge treatment applied, with higher degradation observed under aerobic conditions: commonly found LAS concentrations are 1,000-30,000 mg/kg in anaerobically digested sludge; <1,000 mg/kg in aerobic sludge and <500 mg/kg (dry weight) in aerobically stabilized sludge (Schowanek et al., 2007). Aerobic conditions are also restored during sludge transportation, storage and application on agricultural soil, promoting rapid LAS degradation (Jensen, 1999). The first comprehensive regulation to suggest enhanced monitoring of sludge was the 3rd draft of Directive "Working Document on Sludge" (EU, 2000) published by the European Union in 2000; however, since publication of this document, very few developments on the topic have been fulfilled.

This work provides an important contribution to the concept of introducing EOX and LAS parameters in the monitoring of sludge from WWTPs, as suggested by the "Working Document on Sludge", only partially integrated in some national and regional legislation references (Emilia-Romagna Region, 2005; Italian regulation, 2018).

In this paper, an updated version of a previous study reported by Goi et al. (2006) in the Friuli-Venezia Giulia region (North-East of Italy), the evolution of meaningful characterization parameters in sewage sludge manifested subsequent to upgrading of wastewater treatment processes over an approximately 10-year period was evaluated. In addition, LAS concentration, one of the emerging contaminants in sewage sludge, was also assessed. Due to the increasing stringency of legislation relating to the use of sewage sludge in agriculture, this study may provide a valuable contribution by suggesting the suitability of specific sewage sludges for agricultural reuse and those which, on the contrary, should undergo additional treatment or be forwarded to different final destinations.

2. MATERIALS AND METHODS

2.1 WWTPs and sample collection

Sewage sludge samples were collected from 10 different municipal WWTPs in the Friuli-Venezia Giulia region (North-East of Italy); the analysed plants were reported as samples 1-10 in Table 1, together with a brief description of the main plant characteristics, including treatment capacity (expressed as population equivalent, P.E.), process units and sludge treatment sequences. It should be noted that the majority of analyzed plants were small-scale plants (P.E. in the range of 850-9,000 P.E.) mainly involved in the treatment of domestic wastewater, while two medium-scale plants (n. 1 and 2) were also studied: plant n.1 treated largely municipal wastewater, while plant n. 2 treated a mixture of municipal and industrial wastewater, with the main fraction deriving from chlorine-free pulp and paper industry.

The WWTPs analyzed in this study were localized in the same area as the treatment plants investigated in a similar study in 2006 (Goi et al., 2006). Over the last decade, the selected WWTPs have been transformed and upgraded both in size and process units, thus enabling a comparison of how the improvement of wastewater treatment process could affect sewage sludge quality. For each of the studied WWPTs, a representative sample of 5 Kg was collected manually at the end of sludge treatment from a fresh sludge pile of about 50 kg, by filling polyethylene bags. After collection, each sample was labeled and then placed in a cooler box with ice for transport to the laboratory.

2.2 Sample preparation

Samples were immediately transported to the laboratory and stored at 4°C for subsequent analysis, the samples

TABLE 1: Main characteristics of the plants considered in the present study.

WWTP	Plant size (P.E.)	Wastewater treatment sequence	Sludge treatment sequence
Nr. 1	>100000	Scr. - G.Tr. - O.Rm. - Pr.S.T. - A.S. (N-DN; SBR) - S.Cl. - Disnf.	Thk. - An.Dig. - B.Pr.
Nr. 2	>100000	Scr. - A.S. - S.Cl. - CoTr - Pr.S.T.	Thk. - Aer.Dig. - FP
Nr. 3 *	9000	Scr. - G.Tr. - A.S. (N-DN) - S.Cl. - Disnf.	Thk. - Aer.Dig. – P.D.Bd.
Nr. 4 *	7500	Scr. - G.Tr. - A.S. (N-DN) - S.Cl.	Thk. - P.D.Bd.
Nr. 5	6000	Scr. - G.Tr. - A.S. (N-DN; IFAS) - S.Cl.	Thk. - D.Bd.
Nr. 6	5000	Scr. - G.Tr. - A.S. (N-DN) - S.Cl. - Disnf.	Thk. - Aer.Dig. - D.Bd.
Nr. 7	4000	Scr. - G.Tr. - A.S. (N-DN; MBR)	Thk. - D.Bd.
Nr. 8 *	3500	Scr. - G.Tr. - A.S. (N-DN; MBBR) - S.Cl.	Thk. - P.D.Bd.
Nr. 9	1500	Scr. - G.Tr. - A.S. (N-DN) - S.Cl. - Disnf.	Thk. - D.Bd.
Nr. 10 *	850	Scr. - G.Tr. - A.S. (N-DN; SBR)	P.D.Bd.

*Legend: P.E. = Population equivalent; Scr. = Screening; G.Tr. = Grit Trap; O.Rm. = Oil removal; Pr.S.T. = Primary settling tank; A.S. = Activated sludge; N-DN = Nitrification-Denitrification; SBR = Sequencing Batch Reactor; MBBR = Moving Bed Biofilm Reactor; IFAS = Integrated Fixed-film Activated Sludge; MBR = Membrane BioReactor; SBR = Sequencing Batch Reactor; S.Cl. = Secondary clarifier; CoTr = Coagulation-flocculation treatment; T.F. = Trickling Filter; Disnf. = Disinfection; Thk. = Thickener; B.Pr. = Belting press; FP = Filter Press; Aer.Dig. = Aerobic digestion; An. Dig. = Anaerobic digestion; D.Bd. = Drying bed; P.D.Bd. = Pilot Drying bed; (Dom) = Domestic wastewater; (URB-Dom) = Urban wastewater, mainly domestic; (URB-Ind) = Urban wastewater, mainly industrial * = optimal aeration of the sludge was performed usingy a pilot drying bed (P.D.Bd.) for 6 months.*

were freeze-dried and passed through a 1 mm sieve to obtain well homogenized samples. Sewage sludge samples were frozen at −20°C, then lyophilised using a Coolsafe 55-4 Touch lyophilizer with −50 °C condenser temperature. The ultimate vacuum pressure was 0.4 mbar.

To test potential degradation of LAS over time, a fraction of sludge from the different size WWTPs (samples nr. 3, 4, 8, 10 in Table 1) was placed in a pilot-size aerobic drying bed, where aerobic conditions were maintained by ideal surface venting for 6 months after sludge withdrawal (Table 1).

2.3 Heavy Metal analysis

Heavy metal content was determined using the USEPA 3051 method of Inductively Coupled Plasma Atomic Emission Spectroscopy, ICP-AES, (Varian Vista Pro), as performed in Misson et al. (2020). Calibration was performed using standard solutions (0.5, 1, 5, 10, 30, 50 ppm) prepared from an ICP-standard 23-element solution in 5% HNO3 (Merck solution IV), with yttrium (Y) as internal standard. The method detection limit (MDL) was calculated as 3 s/m (where s is the standard deviation of 10 replicate blanks and m is the slope of the calibration curve) for each element.

2.4 EOX analysis

All samples were freeze-dried, manually sieved through a 1 mm mesh sieve and grinded in a ball-grinder. Subsequently, 1.0 g of pre-treated samples were extracted with 5 mL of solvent (ethyl acetate or n-hexane) by shaking for 24h. Most of the solvent was separated and then evaporated from the extracts under a nitrogen flow, until only 1 mL remained; the resulting sample was then refrigerated until time of analysis. Analyses were performed using Trace Elemental Instrument, Euroglas ECS 1000 upgraded with digital coulometer and control software (TEIS). The apparatus consisted of an injection system, a thermal extraction, a trapping section and a titration cell. 100 µL of residual extract were introduced into the instrument at an injection rate of 20 µL/min, and combustion at 950 °C was carried out, with pyrolysis of organochlorine compounds and release of hydrogen halides. The reaction gases formed during the combustion process, after water removal with sulphuric acid, were introduced into the titration cell where the halogenated acid (formed during the combustion of organic halogens) created a current which could be measured and related to the global quantity of organic halogen compounds in the extract.

2.5 LAS analysis

All measurements were made with a Shimadzu high performance liquid chromatograph LC-20AT (Shimatzu Corporation Kyoto, Japan), fitted with a SIL-20AHT autosampler with a loop 20 µL, equipped with a diode array detector (DAD), a quaternary pump, a vacuum degasser and a thermostatic column compartment. The analytical cartridge column (thermostated at 35°) was a SUPELCOSIL LC-8 (SUPELCO, Bellefonte, PA, USA), 25.0 cm × 4.6 mm ID, 5 µm particle size. Microwave-assisted extraction (MAE) was performed over a Microwave Mars 5 Digestion Oven apparatus (CEM, North Carolina, USA) on 0.5 g of dried sewage sludge samples using methanol (HPLC grade by Merck) as solvent (Mortensen et al., 2001; García et al., 2005; Villar et al., 2007; Braguglia et al., 2014) and the extracts filtered through glass wool and analysed by HPLC.

A good resolution of all LAS peaks was obtained using as mobile phase acetonitrile–water containing 0.1M $NaClO_4$ (55:45) and isocratic elution (acetonitrile was HPLC grade by Merck and sodium perchlorate was analytical grade by Sigma Aldrich). Compounds were eluted isocratically for 6 min at runtime at a flow rate of 0.8 mL/min after 20 µL injection. Instrumental response was preliminarily tested through use of standard LAS solution (standard solutions with C10–C13 chain length were prepared in ultrapure water), highlighting an excellent HPLC cleaning and separation process.

3. RESULTS AND DISCUSSION

3.1 Nutrient content and agricultural reutilization potential

To achieve successful land application, sludge nutrient concentration should be carefully considered. P requirement of most crops is four to ten-fold less than N needs; moreover, P is usually present in more bioavailable forms (inorganic). Consequently, to prevent underestimating crops needs, it is important to consider N requirement when evaluating sludge application rate (Hue, 1995). A comparison with literature data highlighted a wide variation in plant macronutrient concentration in sewage sludge (Table 2). The results obtained on N content in the present work were coherent with other literature studies, while P/N ratio was relatively lower compared to literature studies. (Sommers,1977; Mumma et al., 1988; Mtshali et al., 2014) As shown in Table 2, sewage sludge could be seen as an imbalanced fertilizer due to elutriation of soluble nutrients from sludge during wastewater treatment. For example, K featured a typical range of 0.1 – 0.4 % d.w., consequently, to make the sludge more suitable for agricultural reuse, a K supplement (such as KCl, wood ash and K-rich crop residues) would be required (Hue, 1995; Czerska and Smith, 2008; Pakhnenkoa et al., 2009).

3.2 Heavy Metals

Heavy metal concentration is one of the most crucial factors of concern in the land reutilization of sludge. For monitoring purposes, the heavy metals to be investigated were chosen in line with the suggestions of the "Working Document on Sludge and Biowaste" (European Union, 2004): "heavy metals are intended as cadmium (Cd), chromium (Cr), copper (Cu), mercury (Hg), nickel (Ni), lead (Pb) and zinc (Zn) in metallic form as well as their salts and oxides". The presence of heavy metals is essential for plants and animals, although an excessive concentration of these elements may damage crops and threaten human health by entering the food chain (Usman et al., 2012). Therefore, before land application heavy metal concentrations should meet the limits defined by legislations on sewage sludge management. Table 3 shows the permissible limits for heavy metals suggested by some national legislations and European Council Directive 86/278/EEC (European Commission, 2009), aimed at protecting the environment (in

TABLE 2: Total concentration of selected plant nutrients in sewage sludge

variable	Total Nutrient, % d.w.				
	N	P	K	Ca	Mg
FVG sewage sludge (10 samples)					
Range	3.1-6.8	0.37-1.7	0.17-0.38	1.88-17.13	0.22-4.78
Mean	4.21	0.93	0.27	6.88	1.95
Median	4.15	0.85	0.29	5.47	1.63
Sommers, 1977 (250 samples)					
Range	0.5-7.6	1.1-5.5	0.08-1.1	0.6-13.5	0.03-1.1
Mean	4.9	2.9	0.52	3.3	0.52
Median	4.8	2.7	0.3	3	0.41
Mumma et al., 1988 (15 samples)					
Range	1.19-4.93	0.22-3.13	0.03-0.46	0.32-15.9	0.04-0.81
Mean	2.9	1.2	0.19	3.92	0.35
Median	2.78	0.78	0.15	2.17	0.34
Mtshali et al., 2014 (7 samples)					
Range	0.5-4.5	0.7-2.5	0.04-0.49	0.12-1.59	0.04-0.43
Mean	2.47	1.69	0.15	0.92	0.22

TABLE 3: Selected National and EU permissible limits of heavy metals in sludge for agricultural use (mg/kg d.w.) (EC 2009; Stylianou et al., 2008; Italian regulation, 2018).

Element	Limit 86/278/EEC	Limit Italy	Limit Netherlands	Limit France
Cd	20-40	20	1.25	20
Cr	-	200	75	1000
Cu	1000-1750	1000	75	1000
Hg	16-25	10	0.75	10
Ni	300-400	300	30	200
Pb	750-1200	750	100	800
Zn	2500-4000	2500	300	3000

TABLE 4: Heavy metal concentrations in the analysed sludge (mg/kg d.w.)

Sample	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Italian limits (mg/kg d.w)	20	200	1000	10	300	750	2500
SS no.1	2.12	59.54	339.54	2.12	36.67	92.19	1265.97
SS no.2	0.74	17.74	53.61	0.54	13.06	12.78	109.56
SS no.3	1.17	48.07	584.11	1.36	31.20	63.81	555.47
SS no.4	0.51	32.27	189.89	0.12	20.86	24.59	190.57
SS no.5	1.01	35.17	302.42	0.47	18.86	38.58	436.07
SS no.6	1.65	194.17	593.48	0.36	78.81	81.16	1446.43
SS no.7	0.88	39.71	237.65	0.20	24.76	43.45	309.37
SS no.8	0.94	22.58	146.29	0.07	11.56	24.66	586.92
SS no.9	1.44	41.97	427.55	0.39	27.24	87.21	795.75
SS no.10	1.97	239.63	347.45	6.40	46.61	76.78	5676.40
mean	1.24	73.08	322.20	1.20	30.96	54.52	1137.25
median	1.09	40.84	320.98	0.43	26.00	53.63	571.19

particular soil) when sewage sludge is applied on land for agricultural purposes.

HM concentrations in sludge samples investigated in this study are illustrated in Table 4. As shown in Table 4, heavy metal concentrations in all tested samples were below the maximum permitted limits (Table 3), thus promoting agricultural reuse of the analysed sludge. The only exceptions were Cr and Zn concentrations in sample no. 10 (239.63 and 5,676.4 mg/kg respectively), likely due to the advanced wastewater treatment in plant no. 10. Indeed, plant no. 10 is a small WWTP applying nitrification-denitrification in a sequential batch reactor (SBR); in this kind of process, sludge remains in the tank over extended periods, leading to a significant metal adsorption on the sludge itself.

Generally, the concentration of heavy metals in sludge is affected by the treatment plant potentiality and type of influent wastewater, i.e. domestic or industrial (Spanos et al., 2016; Chanaka Udayanga et al., 2018). Process upgrading in the wastewater treatment circuit may lead to an increase of metal content in the sludge. In practice, comparing actual data with the previous study by Goi et al. (2006) in the same area, a general rise in maximum measured HM concentrations was observed (Figure 1).

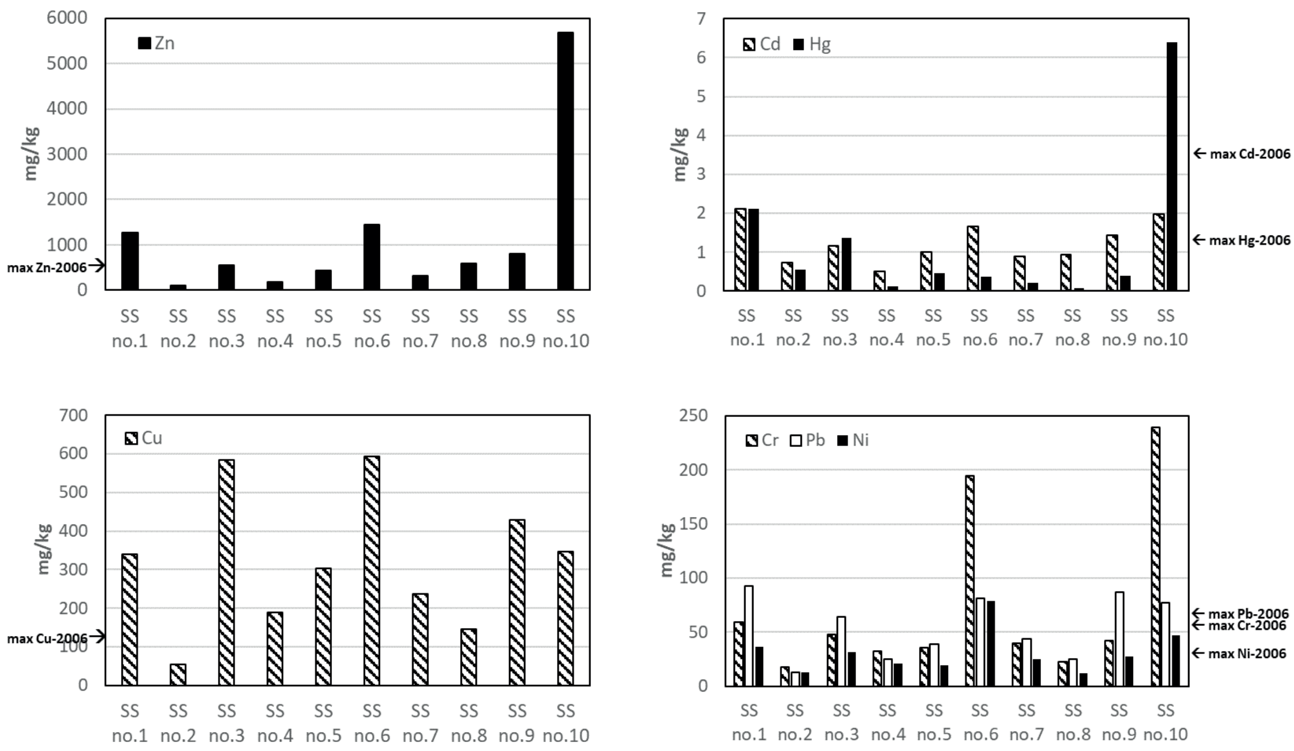

FIGURE 1: Heavy metal concentration in sludge samples and maximum values found in 2006 monitoring campaign.

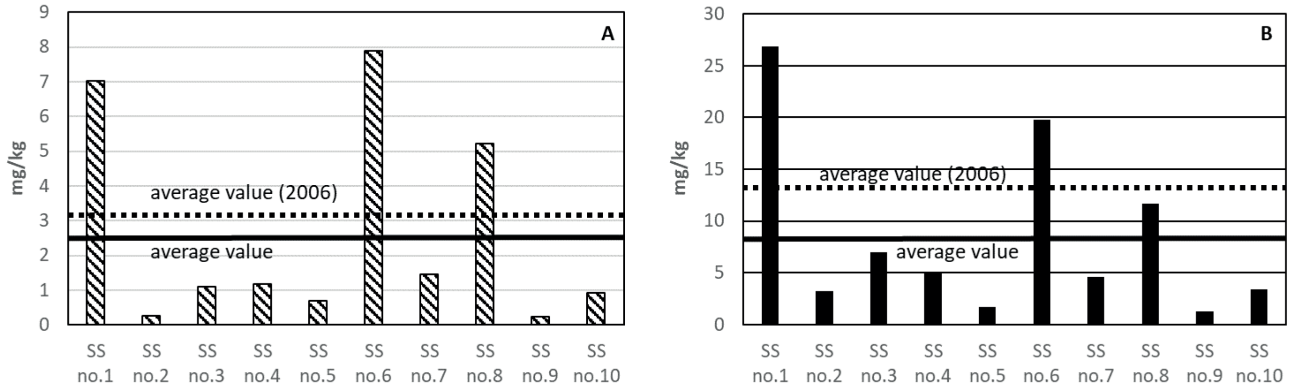

FIGURE 2: EOX concentration measures (A- Hexane extraction; B- ethyl acetate extraction) in sludge samples and average values comparison with 2006 campaign.

This interesting outcome could be explained by the process upgrade and plant revamping undertaken in the studied WWTPs over the last decade. These improvements in wastewater treatment processes have maximized the efficiency of contaminant removal from wastewater, allowing a larger transport of metals into residual solid fraction.

3.3 EOX

Over the last two decades considerable interest has focussed on data relating to the occurrence, behaviour and fate of organohalogen compounds in water, sludge and the environment. The presence of organic halogen compounds in sludge was a key point in the new EU proposed monitoring (European Union, 2004). These compounds, in fact, are highly persistent and do not degrade over time; moreover, they are not absorbable by the soil (especially polar compounds) and consequently directly reach the groundwater, leading to water contamination. Therefore, a sludge with high concentrations of these harmful compounds is generally deemed unsuitable for agricultural reuse (Rizzardini and Goi, 2014).

As well as the singular toxicity of the individual compounds monitored, the utilization of EOX as a sum parameter to obtain global information is a very interesting perspective in monitoring organic halogen content in soils. Moreover, as already observed in a previous case study (Goi et al. 2006), the EOX parameter may be of importance in quality control for the choice of sludge processing routes.

EOX concentrations in the analysed sewage sludge samples, obtained both by hexane and ethyl acetate extraction, are presented in Figure 2 and Table 5. The maximum concentration of EOX found in the present work was 26.86 mg/kg (related to sample no. 1), extracted by ethyl acetate. This EOX content was associated to the largest WWTP with combined municipal and urban sewage sources; EOX concentrations in small municipal WWTPs usually displayed low values in this monitoring campaign, with the exception of samples 6 and 8. At variance with the HM assay, maximum EOX values measured in the actual samples were comparable to those of the previous monitoring campaign.

The measured EOX content in ethyl acetate extractions was 2-6 times higher when compared with hexane extractions, in agreement with the results reported by Reemtsma and Jekel (1996). This suggested that polar halogenated molecules were present in larger quantities than non-polar compounds (i.e. non-polar compounds could only be extracted by ethyl acetate).

TABLE 5: EOX and LAS concentration in sludge samples from analysed WWTPs (mg/kg d.w.)

Sample	LAS	CV%	EOX			
			By hexane	CV%	By ethyl acetate	CV%
SS no.1	574.41	9.6	7.03	5.34	26.86	1.26
SS no.2	136.36	14.8	0.25	45.02	3.26	38.8
SS no.3	180.95	14.1	1.11	20.57	6.95	4.27
SS no.4	53.75	26.3	1.17	28.6	4.98	5.88
SS no.5	523.45	23.6	0.69	55.17	1.74	11.89
SS no.6	220.27	22.7	7.89	16.00	19.81	7.73
SS no.7	301.57	12.4	1.46	15.13	4.56	5.08
SS no.8	55.88	17.3	5.22	7.83	11.7	14.4
SS no.9	427.76	9.2	0.24	31.66	1.24	24.19
SS no.10	138.12	16.3	0.93	11.76	3.38	3.77

EOX monitoring in sludge from WWTPs in the considered area showed a general persistence of halogenic contamination. Upgrading of the treatment plant process apparently produced no visible effect on EOX degradation. However, a general reduction in average EOX concentration was discernible compared to the previous monitoring study (Figure 2).

3.4 LAS

Due to the extensive use of surfactants, specifically LAS, in domestic and industrial applications, their presence in sewage sludge is assured (Granatto et al., 2019). Moreover, these compounds can only be partly degraded in WWTPs, with the extent of degradation largely depending on a) LAS content in raw sewage, b) sludge age after storage and c) process nature (i.e. whether the process is aerobic or anaerobic). Therefore, a measurable portion of LAS invariably accumulates in soils and consequently monitoring of these compounds in sludge is extremely important (Villar et al, 2007).

In Europe LAS concentration in sewage sludge lies between <1 g/kg d.w. and 30 g/kg d.w. (Gawlik and Bidoglio, 2006). Table 5 and Figure 3 show the measured LAS concentration in the analysed sludge samples. The lowest concentrations were found in samples 4 and 8 (55.88 and 53.75 mg/kg, respectively), while the highest amount was highlighted in sample no. 1 (574.41 mg/kg). Stock et al. (2002) analysed more than 150 sludge samples from different WWTPs in a comprehensive study in Westphalia (Germany). They found a correlation between WWTPs size and LAS concentration; it should be underlined that extended aerobic sludge treatment is common in smaller treatment plants and enhances reduction in LAS concentration.

In accordance with the above, LAS concentration in sample 2 (treated by aerobic digestion) was three times lower than sample 1(treated by anaerobic digestion) for equal WWTPs size. The present study demonstrated that LAS content was lower in sludge treated in pilot drying beds, with optimal aeration, or by aerobic digestion process. It is noteworthy that LAS mean concentration was significantly lower than limit values proposed in the "Working Document on Sludge" (EU, 2000).

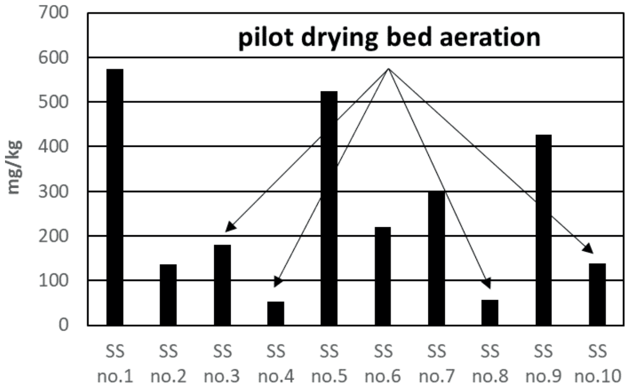

FIGURE 3: LAS concentration in sludge samples from analysed WWTPs.

To summarize, the quality of the investigated sludge (in terms of HM, EOX and LAS) depended on multiple factors, including plant size (PE), type of treated water (Dom., Ind., or Urb.), wastewater treatment process and sludge treatment sequence. It was not possible to directly correlate each parameter to sludge pollution, as all these variables were strictly interconnected and strongly matrix-dependent. Despite the differences and variability present in the analysed parameters, all investigated sludge samples complied with Italian limits suggested by the regulations for agricultural reuse.

Comparison with a previous study (Goi et al., 2006) highlighted a crucial factor: the progressive improvement in wastewater treatment seems to promote a higher concentration of several harmful pollutants in sludge. When HM, EOX and LAS were monitored together, it was found that the more efficient the wastewater treatment line, the higher the inorganic/organic substances transferred to the sludge (Figure 4). This outcome suggests adopting a critical approach to further process development in wastewater treatment lines, considering future perspectives in sludge agricultural reuse together with the need to develop alternative final destination routes, such as thermal processes (incineration, pyrolysis, gasification, hydrothermal treatment) (Chanaka Udayanga et al., 2018). Teoh and Li

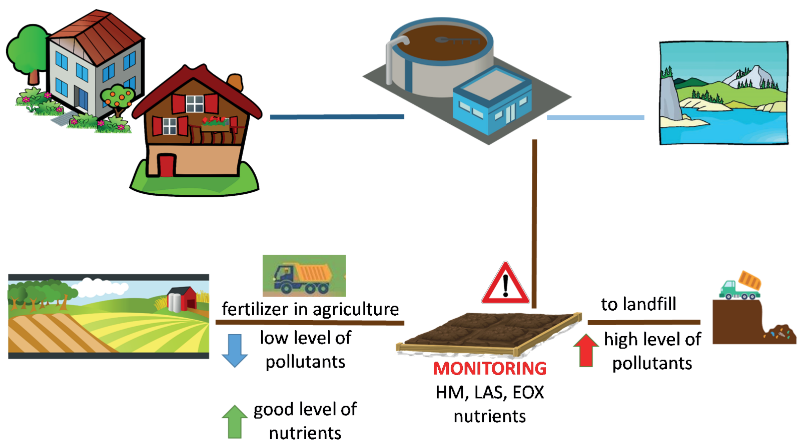

FIGURE 4: Proposed scheme for sludge checking, to prevent the risk of soil and groundwater contamination in the agricultural reuse of contaminated sludge (considering HM, EOX, LAS).

(2020) studied alternative sludge treatment methods applying Life Cycle Assessment (LCA); the investigated aspects were sludge volume, pollutants, global warming and toxicity. It was concluded that anaerobic digestion, pyrolysis and supercritical water oxidation were the best-performing treatment methods (Teoh and Li, 2020), suggesting that a differentiated approach is highly recommended in sludge treatment and valorization, enhancing the recovery of both energy and material, whilst at the same time reducing environmental contamination hazards.

4. CONCLUSIONS

Subsequent to the increasing diffusion of wastewater treatment, particularly in high- and middle-income countries, the sewage sludge generated should be treated and valorised in an ecological and economic way, thus contributing to the circular economy. While wastewater treatment plants are effective in removing pollutants from water, they accumulate heavy metals and other persistent toxic compounds in sludge, thus restricting its potential reuse. In this study, HM, EOX and LAS, three main limiting factors for the land application of sewage sludge, were monitored in sewage sludge samples from 10 different low-middle capacity wastewater treatment plants in the Friuli-Venezia Giulia region (North-East of Italy). The results showed how the concentration of these compounds was much lower than permissible limits suggested by Council Directive 86/278/EEC for the agricultural reuse of sewage sludge, in particular in sludge originating from small municipal plants. Sewage sludge from the studied wastewater treatment plants could be used for sustainable agriculture, exploiting its macro and micronutrient content, without posing a threat for the environment and human health. On the other hand, the present study highlighted a future scenario in which a continuous progress in wastewater treatment, by decreasing pollutant levels released to the receptor body and thus improving water quality, would transfer higher quantities of potentially harmful compounds to the sewage sludge, rendering it unsafe for agricultural reuse. This study, also in view of a similar monitoring campaign performed in 2006, questioned the future possibility of reusing sewage sludge in agricultural applications. Moreover, it raised the need for further constant and regular sludge control, with the specific aim of preventing the agricultural reuse of sludge contaminated with organic and inorganic substances (HM, EOX, LAS), and associated risks of soil and groundwater contamination, as well as potential entrance into the food chain.

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OVERVIEW OF ODOUR MEASUREMENT METHODS: THE ODOUR OBSERVATORY AS AN INFORMATIVE TOOL FOR CITIZEN SCIENCE BASED APPROACHES TO ODOUR MANAGEMENT

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ABSTRACT
Odour pollution is a well-known problem related to a number of different industrial activities. It is also one of the main causes of citizens' complaints to local authorities. Specific programs are needed to manage persistent odour pollution problems within communities in order to avoid possible socio-environmental conflicts. The H2020 project D-NOSES (Distributed Network for Odour Sensing, Empowerment and Sustainability) aims to help citizens co-create local solutions in collaboration with industries, regional & local authorities, and odour experts. The project will develop an innovative bottom-up approach to odour pollution governance by combining citizen science and stakeholder management methods using a quadruple helix model. The first aim of this article is to introduce the D-NOSES project and its methodology. Aside from that, the article presents an overview of the existing odour impact assessment methods currently available to quantify odour pollution. Finally, the different odour measurement methods are compared in terms of their applicability and limitations. This overview will be made available online as a first step towards the development of the International Odour Observatory, a platform to be created within the D-NOSES project to help promote odour pollution management and resolve regulation issues. The platform will become a global resource on odour issues and also include information about odour abatement systems, chemical substances in odour emissions, and odour regulations around the world.

1. INTRODUCTION

Odour pollution is a recognised problem related to a number of different industrial activities, and one of the main causes of citizens' complaints to local authorities (Henshaw et al., 2006). This is particularly true when waste treatment facilities are involved (Lucernoni et al., 2017; Marchand et al., 2013; Sironi et al., 2006). Although odour emissions are generally considered harmless, in some cases they may cause adverse health effects for citizens that go beyond mere inconvenience (Aatamila et al., 2011). In order to avoid socio-environmental conflicts within the impacted communities, specific programs are needed to manage odour pollution problems.

Combining Citizen Science and participatory strategies, the H2020 project D-NOSES (Distributed Network for Odour Sensing, Empowerment and Sustainability) will help citizens to co-create local solutions together with industries, regional & local authorities, as well as odour experts. Ten pilot projects will be conducted in ten European and non-European locations to validate the methodology. Each pilot will involve encouraging local citizens to collect data and use a variety of different techniques for validation. The pilot groups will then employ a stakeholder engagement methodology to improve the management of odour pollution issues within their own local context. D-NOSES will promote the idea that using citizen science to monitor odour pollution is a logical choice since citizens are naturally equipped with the best sensor available to measure odours, i.e. their own noses. A mobile app (OdourCollect, https://odourcollect.eu/) was specifically created for the project, providing a platform that allows citizens to easily gather odour observations and co-create odour maps of affected communities.

In addition, the project will create the International Odour Observatory in order to provide a comprehensive global resource for odour management issues. The ob-

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servatory will gather odour data, raise awareness, and make relevant environmental information available to all interested stakeholders. There will be a section devoted to compiling the scientific and regulatory frameworks currently used to manage odour pollution around the world. This section will also provide other useful and relevant information such as the chemicals that are commonly associated with odour emissions from different activities, the systems that can be used to reduce these odour emissions, and finally an overview of any existing odour regulations in different regions.

The observatory aims to educate stakeholders at all different levels, and in particular ordinary citizens, concerned with odour pollution. One of the most common questions regarding odour pollution from citizens is whether odours can be effectively measured. For this reason, one of the first objectives for the Odour Observatory is to explain how odours can be measured and provide an overview of the methods that can be used for this purpose. Furthermore, the observatory will make clear that it is not just about measuring odours, but rather it is also important to be able to assess odour impacts in a simplified and accessible manner.

This paper has the aim to introduce the D-NOSES project and its methodology, and to present an overview of the existing odour measurement methods that can be applied to quantify odour pollution and impacts. By gathering this information the Odour Observatory made available through the D-NOSES project can be used in the future to compare different methods and help foster a common approach based on best-practices. Some recent review papers are also referenced, provide relevant examples of how such different techniques have been applied in different situations (Conti et al., 2020; Guffanti et al. 2018).

2. THE D-NOSES PROJECT: OVERALL AIMS AND STRATEGY

The overall aim of the D-NOSES project is to develop and validate a methodology for odour pollution management based on a bottom-up approach. This approach focuses on using participatory strategies for citizen involvement, engagement with a broad set of quadruple helix stakeholders, and the co-creation of practical and balanced solutions. The project intends to:

- Raise awareness about odour pollution, and address related environmental and sanitation problems, at global, national and local levels.
- Pave the way for increased sustainability and quality of life through a multi-level engagement strategy to resolve odour issues and related conflict situations;
- Provide easy access to information for all stakeholders through the International Odour Observatory;
- Collect evidence through 10 local case studies in European and non-European countries to validate the methodologies;
- Provide common scientific guidelines for policymaking;
- Produce a Green Paper and a Strategic Roadmap for Governance in odour pollution to advocate and inform the development of common, bottom-up, efficient and coherent regulations.

According to the above listed points, one of the Work Packages (WPs) of the project is devoted to the description of the current scientific and regulatory frameworks for odour pollution across different regions.

This knowledge base will be one of the pillars of the International Odour Observatory, the purpose of which will be to provide a single point of reference for all stakeholders involved with odour pollution issues. Indeed, in order to reach the goal of creating common scientific guidelines for policy making, it is extremely important to define the problem by identifying relevant odour sources, up-to-date techniques for odour impact assessment and abatement, as well as current policies and regulations in European and extra-European countries.

3. THE INTERNATIONAL ODOUR OBSERVATORY

3.1 The Odour Observatory as an informative tool for citizens

Despite being the second most common reason for environmental complaints in Europe (Marchand et al., 2013), information and advice about odour pollution can be hard to find. The level of knowledge about fundamental aspects related to odour pollution tends to be scarce, especially among ordinary citizens. In fact, it is not rare to hear such basic questions as: “What is an odour?” or “Is there a way to measure odours?”. This lack of basic knowledge could severely limit the possible success of any proposed bottom-up methodology for the management of odour pollution problems. In recognition of this limitation, one of the main objectives of the D-NOSES project is the creation of the “International Odour Observatory”. The observatory will help to communicate the required knowledge for all stakeholders to participate effectively in a citizen science based methodology for odour governance. Further, the observatory will also help gather data about specific odour pollution problems and make any relevant and actionable environmental information available to all interested stakeholders.

More specifically, the Odour Observatory will have a “Get informed” section (Figure 1) containing information on which regulations apply in different parts of the world, how odours can be measured and all other relevant information related to odour pollution. The information will be made available at different levels of complexity, targeting the differing interests and requirements of each specific stakeholder. This section will be considered as a first step in educating stakeholders about odour pollution and will provide a natural entry point to get them more actively involved in odour issues.

3.2 Structure of the section describing how odours can be measured

One of the main questions that the Odour Observatory will answer is on how odours can be measured. A preliminary document was created providing a classification scheme for the identified methods for odour measurement.

What does the World Smell Like?

We are excited to introduce to you the International Odour Observatory, one of the main and lasting results of the D-NOSES project that will carry on the work of empowering citizens to tackle odour pollution in their own communities.

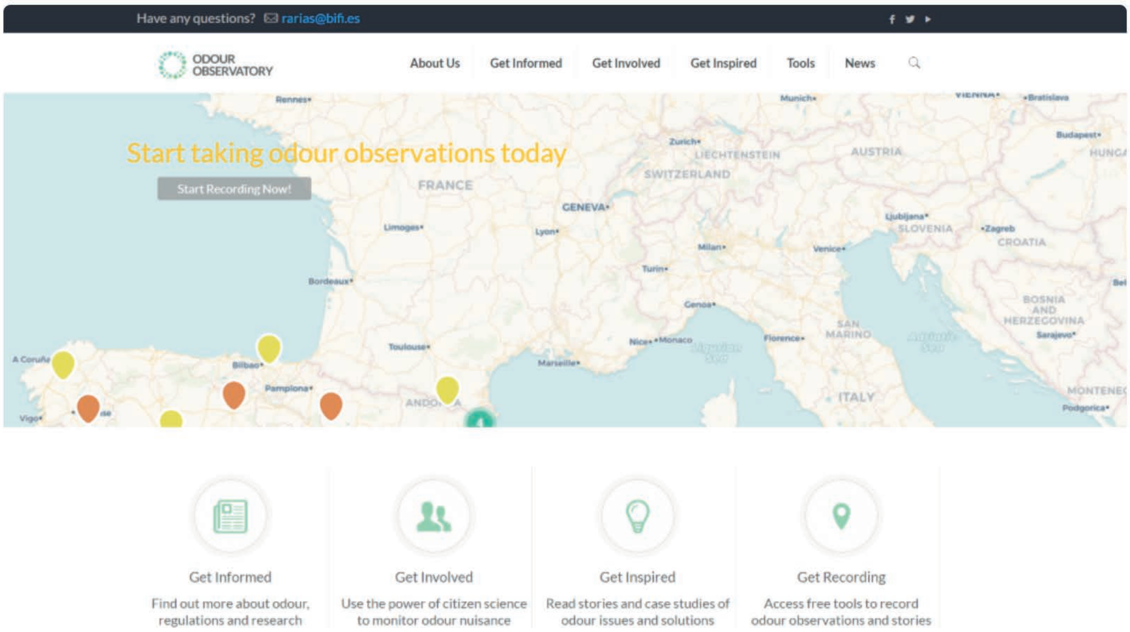

FIGURE 1: Example of homepage of the Odour Observatory (under construction).

Within the Odour Observatory, , each method is described briefly and simply by answering the following questions:

- What is it? (Method description)
- What can it be used for? (Applicability)
- What can it not be used for? (Limitations)

Each entry is further complemented with some examples of relevant applications as well as a list of links to selected reference articles for further detailed information. This way the observatory can serve the needs of both a technical as well as a lay audience.

This paper is limited to the schematization of the methods and to their brief description according to the above-reported list of questions. For more detailed information about relevant applications described in the scientific literature, please refer to Capelli et al., 2019.

4. OVERVIEW OF ODOUR MEASUREMENT METHODS

4.1 General schematization of odour impact assessment methods

As a first step, the existing odour impact assessment methods were identified, and an attempt was made to schematize them according to the type of method (mathematical methods, instrumental measurements and sensorial measurements) and depending on where they can be applied (emissions vs. ambient air). The resulting scheme is shown in Figure 2 (Capelli et al., 2019).

In order to provide an overview of the existing odour impact assessment methods, the following paragraphs present a very short description of each method, then the last paragraph provides a comparison of the different methods in terms of applicability and limitations. Currently this overview only focuses on practical methods for odour measurement, leaving out mathematical methods.

4.2 Dynamic olfactometry

Dynamic olfactometry is a standardized sensorial technique for measuring odour concentrations using the human sense of smell (CEN, 2003). It is related to the sensation caused by a sample directly on a panel of opportunely selected people.

The outcome of this measurement is the odour concentration of the sample, expressed in European odour units per cubic meter (ou_E/m^3). This represents the number of times the sample has been diluted with neutral air to reach its odour detection threshold concentration. Thus, if the sample needs to be diluted 100 times with clean air so that the panel cannot perceive the odour anymore, this means that the sample has a concentration of 100 ou_E/m^3.

Samples of odorous air are collected at the source of the odour in bags. The analysis is carried out by presenting the sample to the panel at increasing concentrations by

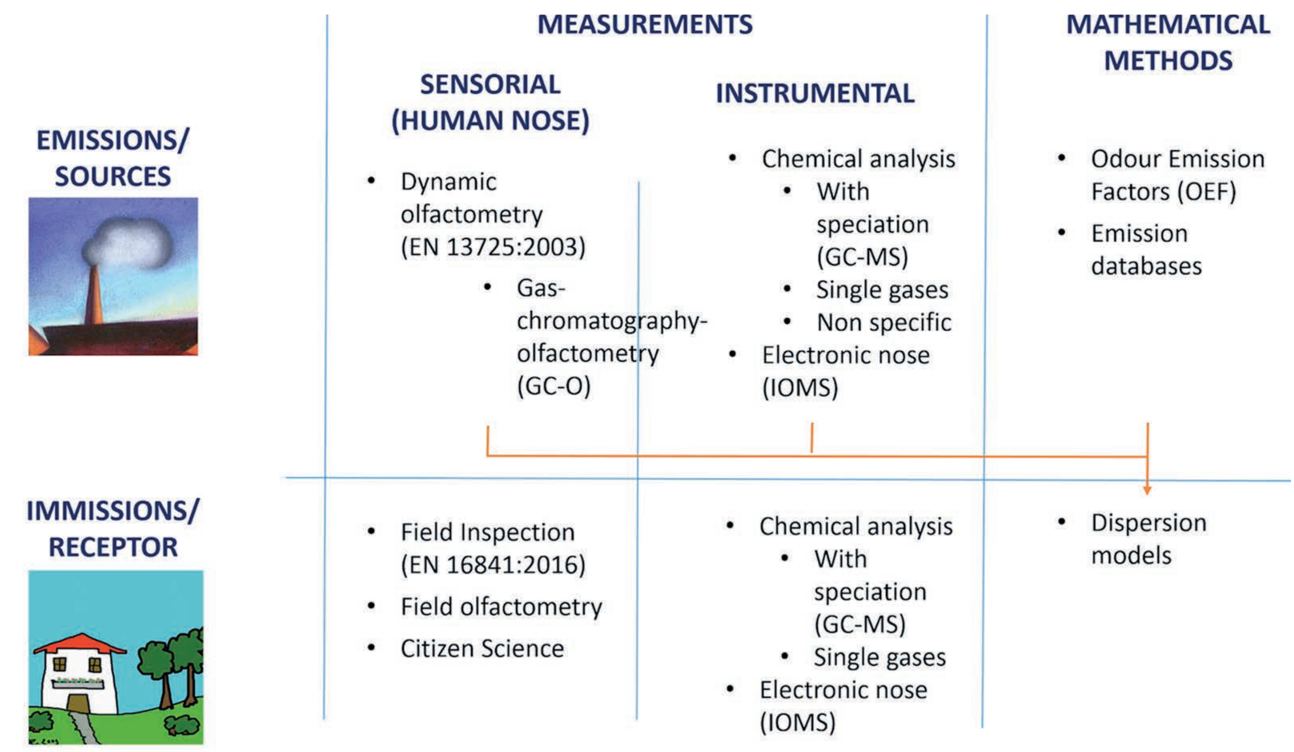

FIGURE 2: Schematization of odour impact assessment methods.

means of a dilution device called an olfactometer, until the panel members can detect an odour that is different from the reference air.

Results of olfactometric analyses can be used as inputs for dispersion models in order to evaluate impacts on receptors (Sironi et al., 2010; Sowka et al., 2016).

4.3 Chemical analysis – with speciation

Chemical analysis (with speciation) of odours is an instrumental analysis for the complete identification and quantification of odorous chemical compounds in an odour sample (e.g., Ávila et al., 2014; Davoli et al., 2003; Rodríguez-Nava et al., 2012; Zhao et al., 2015). It is an "instrumental analysis": the main technique is Gas Chromatography coupled with Mass Spectrometry (GC-MS). GC - MS is a technique that combines the separation capability of gas chromatography (GC) with mass spectrometry (MS) for the identification of the separated compounds.

GC can separate different molecules depending on their chemical-physical properties. After separation, MS breaks each molecule into ionized fragments, obtaining a mass spectrum. A mass spectrum can be considered to be the fingerprint of a molecule. It is characteristic to each molecule and can be used to uniquely identify a substance.

4.4 Gas-chromatography-Olfactometry (GC-O)

This is a method that combines the information provided by chemical characterization and odour perception. GC-O utilises a GC-MS system equipped with an olfactory detection port. At the outlet of the GC is a sniffer mask through which a trained panelist can smell the gas and provide information about the presence of odour in it. GC-MS-O enables simultaneous chemical and sensorial analysis, which can be particularly useful in identifying compounds that are responsible for the overall odour perceived by the human nose (Wright et al., 2005; Zhang et al., 2015).

4.5 Chemical analysis – non-specific

In cases where the odour problem is specifically related to hydrocarbon molecules, such as oil refineries and landfills (Singh et al., 2013; Kormi et al., 2018), a non-specific gas analysis can be applied as a preliminary screening tool to assess the total amount of hydrocarbon compounds. This can be done using easily transportable and inexpensive tools like FID (flame ionization detector) or PID (photo ionization detector) (Samir and Hossain, 2014).

These tools are based on the pyrolysis of the organic compounds using some sort of energy source (hydrogen flame for FID, UV lamp for PID). The pyrolisis produces ions which are detectable by an electric sensor.

4.6 Chemical analysis – single gases

In those rare cases in which the odour pollution problem is due mainly to a single compound, such as NH3 or H_2S, a reliable quantification of odours (in emissions or in ambient air) can be obtained by assessing the concentration of these single gases (Wang et al., 2014). However, it should be highlighted that the situations in which complex environmental odours can be represented by just one tracer compound are rare.

There are specific technical norms that define the sampling and analysis methods for the measurement of single gases in emissions. When the concentrations are quite high (1-10 ppm) it is possible to use electrochemical sensors, which are easy to use and cheap.

When applying the analysis of single gases to ambient air, the compound to be measured should not be ubiquitous; its source must be clearly identifiable. When concentrations are low, more complex and expensive tools are needed, like a chemiluminescence analyzer for NH_3 or gold foil instruments for H_2S.

4.7 Instrumental odour monitoring (E-noses)

An electronic nose is a piece of equipment designed to mimic mammalian olfaction for the detection and characterization of simple or complex odours.

These devices allow the identification of mixtures of organic samples as a whole, providing their olfactory fingerprint, without recognizing the individual odor-generating compounds. Much in the same way that the human nose doesn't need to identify each single odorant molecule in order to distinguish the odour of an apple from the smell of rotten eggs.

To do this, the instrument must be trained: it must be provided with a database of olfactory fingerprints of the odours it may be exposed to during the analysis. That database is put together by analyzing air samples with known olfactory qualities at different odour concentrations and thus defining the olfactory classes (odour types) to be recognized.

Today, electronic noses are increasingly applied in the continuous monitoring of odours from different types of facilities (Deshmukh et al., 2015; Cipriano et al., 2019).

4.8 Field inspection

The main idea behind field inspections is to estimate the degree of annoyance in a determined problematic area by means of the olfactory capacities of a group of people (panel). The panel is specially trained and "calibrated" for this purpose. This method has been recently standardized by a European Norm (CEN, 2016a,b).

Two different approaches for field inspection can be applied:

- Grid method (CEN, 2016a): uses direct assessment of ambient air by panel members to characterize odour exposure in a defined assessment area;
- Plume method (CEN, 2016b): determines the extent of the downwind odour plume of a source (there is no direct relation between the presence of recognizable odours and the occurrence of odour annoyance).

Field inspections are, up to now, the only standardized method that can be used for direct odour exposure assessment. They are also very good methods for the field verification of dispersion model results (Ranzato et al., 2012; Capelli and Sironi, 2018).

4.9 Citizen Science

The Citizen Science approach (Paulos et al., 2009; Bonney et al., 2016) to monitoring odour harnesses the power of crowds and uses one of the most effective odour sensors - the human nose. Communities can record the frequency, intensity and type of odour that they experience. The combination of many individual observations can then build a clear picture of the issue. As more citizens are involved in sharing their findings, or data, the level of subjectivity is reduced. The D-NOSES project will use this technique, with the express aim to develop and validate a citizen science based bottom-up approach for odour pollution management.

4.10 Comparison of methods

The above-mentioned methods are based on different principles and thus can be used to provide very different types of answers. For this reason, it is difficult to make an absolute comparison of these methods. A limited comparison can be provided by stating the limits of applicability of each method, thereby clearly defining what they should, or should not, be used for. This information will be provided in the Odour Observatory, as summarized schematically in Table 1 (Capelli et al., 2019).

5. CONCLUSIONS AND FUTURE WORK

As stated earlier, the aim of the D-NOSES project is to develop a methodology for odour pollution management based on a bottom-up approach. The approach will focus on participatory strategies to involve citizens as well as engage quadruple helix stakeholders in the co-creation of relevant solutions.

To actively involve citizens in the process, it is important that they start with sufficient knowledge to be able to take part effectively. For this reason, the project emphasizes the accessibility of basic information such as the definition of an odour and the basic principles of odour measurement. The main channel for this will be the "International Odour Observatory", which will present simple and relevant environmental information through an easily accessible "get informed" section. The observatory will also address more sophisticated and technical audiences by offering increasingly detailed information to those that are interested.

This paper describes some of the initial information to be included in the Odour Observatory, which will provide answers to questions on how odours can be measured. An overview of the different existing odour measurement methods is given here, including a short description. The methods are distinguished based on the type of method (mathematical, instrumental or sensorial measurements) and on where they should be applied (emissions vs. ambient air). Moreover, a schematic comparison of the different methods is given in terms of their applicability and limitations.

This overview of odour measurement methods is only the first part of the information that will eventually be made available through the Odour Observatory. The next questions that will need to be answered for the development of the Odour Observatory concern: i) the available techniques that can be adopted to reduce odour emissions (odour abatement systems), ii) the presence of – potentially hazardous – chemical substances that can be found in odour

TABLE 1: Schematization of different odour measurement methods in terms of their applicability and limitations.

Measurement method	Applicable to emissions or ambient air	Applicability	Limitations
Dynamic olfactometry	Emissions	• Measure the concentration of odours emitted at the source • Ascertain if regulations are being breached • Provide information that can be used as input data for dispersion modelling in order to evaluate citizens' exposure to odours	• Discontinuous method, cannot be applied to the continuous measurement of odour emissions • No information about odour quality; it cannot identify or discriminate different odours • No information about presence of odours in ambient air: it only provides information about odour emissions
Chemical analysis – with speciation	Emissions and/ or ambient air	• Obtain information about the chemical composition of odours • Identification and quantification of the chemical compounds that are present in an odour emission or in ambient air can be used for the evaluation of the impact on the environment and human health	• Very difficult and not always effective, especially in the characterization of complex odours • Odours are not additive due to synergistic and masking effects between odorants; therefore chemical composition of an odorous sample can not be related to its odour concentration • Less sensitive than human nose for malodorous compounds with low odour threshold
Gas-chromatography-Olfactometry (GC-O)	Emissions	• Gain information about the odour character associated with the different molecules contained in an odour sample, and thus odour quality • High sensitivity: human nose is more sensitive than any instrumental detector; the human nose is sometimes able to detect the presence of odours also where the chromatogram doesn't show any peak	• No information about the odour concentration of the sample. • Because of the separation of the sample in its single components, the olfactory properties of the sample as a whole are not considered • Cannot provide information about the odour impact, and neither can it be used directly as input for dispersion modelling
Chemical analysis – non-specific	Emissions	• Detection of gas leaks, which are potentially associated with diffuse odour emissions • Very useful in the detection of fugitive emissions from equipment or piping in refineries, or leaks in landfill soils	• No information about the odour properties of the analyzed gas
Chemical analysis – single gases	Emissions and/ or ambient air	• Quantify the concentration of single gases in emissions or ambient air • Estimate the odour concentration in emissions, in those rare cases in which the emitted odour is directly correlated to one specific compound (tracer) • Measure the impact of odour in ambient air, in those rare cases in which the odour is directly correlated to one specific compound (tracer), and the source can be univocally identified	• Useless in the case of complex odorous mixtures, whereby odour concentration is not related to the concentration of one single component; this is the most common case with environmental odours, which are mixtures of hundreds of different compounds
Instrumental odour monitoring (E-noses)	Emissions and/ or ambient air	• Continuous and fast results with a limited budget • Continuous measurement of odour concentration at emissions, e.g., for continuous monitoring of the effeiciency of odour abatement systems • Direct determination of the odour impact at receptors and identification of odour provenance	• No information about intensity and pleasantness of the odour • Cannot substitute dynamic olfactometry
Field inspections	Ambient air	• Estimate the degree of annoyance in terms of "odour hours" in a determined area under investigation ("grid method") • Determine the extent of the odour plume from a facility under specific meteorological conditions ("plume method") • With a suitable training, assessors may provide information about odour quality	• No information about odour concentration
Citizen science	Ambient air	• Involve citizens in the process of odour impact assessment • Estimate the degree of annoyance by directly referring to the effect on citizens	• Risk of biased information • Hardly applicable in conflictual situations (e.g., law suits)

emissions, and iii) existing regulations and impact criteria regarding odour impacts in European and non-European countries.

Once collected, this information will form the basis to describe the current scientific and regulatory framework for odour pollution. It will provide a comprehensive knowledge base, considered to be a fundamental step in enabling a methodology that includes a wide range of quadruple helix stakeholders in the entire process of odour pollution management.

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EVALUATION OF ORGANIC AND RECYCLABLE WASTE SEPARATION AT GENERATION SOURCE IN RATNAPURA AND KATARAGAMA LOCAL AUTHORITIES IN SRI LANKA

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ABSTRACT

The Japan International Cooperation Agency implemented a technical cooperation project from 2017 to 2019 in collaboration with Central Environmental Authority (CEA) in Sri Lanka. The project provided technical; and financial assistances in preparing solid waste management plans at selected local authorities, and consequently implemented 3R promotion programs introducing source separation at households and subsequent separate collection by local authorities. Two pilot projects were implemented based on the solid waste management plans which were aimed at reducing the amount of solid waste to be landfilled. This paper described the approaches taken and results obtained through the pilot projects in Ratnapura Municipal Council (RMC) and Kataragama Pradeshiya Sabha Town (KPS) of Sri Lanka. The pilot project in RMC consisted of distribution of household compost bins and the separate collection of five categories of waste: 1) biodegradable waste, 2) plastic and polythene, 3) paper and cardboard, 4) glass, metal and coconut shells, and 5) other residual waste; and provision of advices to households by leaflets, distribution of equipment, and implementation of an awareness program. The pilot project in KPS consisted of procurement of separation bins for biodegradable waste by cost-sharing with dischargers and the separate collection of three categories of waste: 1) biodegradable waste; 2) recyclables including paper, cardboard, plastic, polyethylene, iron and coconut shells, broken glass and glass bottles; and 3) other residual waste. The post evaluation of waste composition and final discharge amounts at disposal sites indicated that pilot projects have contributed to increase 3Rs in both pilot project areas.

1. INTRODUCTION

Sri Lanka, as a lower-middle income country whose GNI per capita is 3,859 USD in 2017 (World Bank, 2019), is still tackling – and has for several decades – many issues of solid waste management. The total municipal solid waste generation in Sri Lanka is approximately 7,210 tons/day (Basnayake, 2018). Most of the collected waste, including biodegradable waste, ends up in 300 or more number of open dump sites or primary landfill sites scatted around the country. The dumpsites are often less than one hectare in area and lack proper landfill components such as leachate, emission and groundwater contamination control measures.

In Sri Lanka, the National Strategy for Solid Waste Management was formulated in 2000 and it recommends solid waste management be conducted as follows:

- Prioritise waste avoidance and reduction over the next stage of waste recycling and other forms of environmentally-sound disposal,
- Reuse unavoidable waste as much as possible,
- Maintain the content of hazardous substances in waste at the lowest possible level, and
- Guarantee an environmentally sound residual waste treatment and disposal as basic prerequisites for human existence.

Furthermore, the National Policy on Solid Waste Management was formulated in 2007 and it defines the environmental accountability and social responsibility of all waste generators, waste managers and service providers and aims to actively involve individuals and all relevant parties in integrated and environmentally-sound solid waste management practices. On the other hand, the Sri Lankan Government prioritises appropriate and sustainable SWM – a Ten-year Horizon Development Framework (2006-

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2016) – and identifies the importance of the promotion of the 3Rs (reduce, reuse, recycle) and the establishment of environmentally-friendly final disposal sites for a sustainable SWM system.

Several engineered landfill sites have been established and are operating since the early 2000s and one sanitary landfill at DOMPE site in Western Province has been operated by the CEA since 2014. The lifetimes of the engineered landfill sites and sanitary landfill site are limited due to shortage of capacity and high waste discharge amounts. In order to improve and upgrade present open dumps, the technical, economic, social and institutional aspects of local authorities need to be fully taken into account in Sri Lanka (Zurbrugg, 2003). Incinerators are not used as a solution for municipal solid waste at the moment. Since the recycling system does not function properly, it is estimated that waste recycling rate is less than 3% in Sri Lanka (JICA, 2019). This means that most recyclable waste ends up merely being disposed of at landfill sites without being processed for recycling. To solve these issues, the Sri Lankan government intends to develop more sanitary landfill sites and new incineration plants. In addition, 3Rs have been promoted by local authorities for a few years to implement proper waste management practices, such as waste reduction at generation source, waste segregation of biodegradable waste and recyclable waste, and intermediate treatment. However, the effectiveness of 3R activities seems limited at some local authorities that mainly concentrate on the segregation of recyclable waste rather than biodegradable waste.

To support the efforts of the Sri Lankan Government, the Japan International Cooperation Agency (JICA) implemented the technical cooperation project, Experts in Pollution Control and Reduction of Environmental Burden in Solid Waste Management (ReEB Waste), from 2017 to 2019 in cooperation with CEA. ReEB Waste assisted preparing solid waste management action plans at local authorities and implemented 3R promotion programs involving solid waste separation and collection pilot projects aimed at reducing the amount of landfilled solid waste generated in target areas in Ratnapura Municipal Council (RMC) in Sabaragamaua Province and Kataragama Pradeshiya Sabha (KPS) town in Uva Province. Moreover, ReEB Waste also implemented pilot projects for rehabilitation of current disposal sites to mitigate environmental issues with locally available, low-cost materials and low-maintenance techniques in Kurunegala Municipal Council in North Western Province and KPS. The selection of cities for strengthening the capacity of local authority to implement source segregated waste collection was based on several criteria such as absence of effective source segregation, level of urbanization, strategic importance for dissemination of good practices, and willingness of local authority for improvement.

The following case studies describe the results and evaluation of biodegradable and recyclable waste separation at generation source in RMC and KPS under the pilot projects of 3R promotion by ReEB Waste.

2. PILOT PROJECT AT RATNAPURA MUNICIPAL COUNCIL (CASE STUDY 1)

2.1 Overview of Ratnapura Municipal Council

2.1.1 Outline of Ratnapura Municipality

Ratnapura Municipality has a population of around 54,000 (in 2016 (JICA, 2019)), 12,931 households and is the capital of Sabaragamuwa Province, Sri Lanka. Ratnapura city centre is 101 km southeast of the Sri Lankan capital of Colombo. Ratnapura is the major commercial and services hub of Ratnapura District, as well as the only local authority with municipal status in Sabaragamuwa Province, Sri Lanka. Ratnapura has a renounced gem industry and education institutions, with many popular schools and tuition centres. Ratnapura City is also a major transit hub in Sri Lanka, connecting Sabaragamuwa, Eastern and Southern provinces.

2.1.2 Baseline study in RMC

In 2017, a baseline study on waste management practices was conducted in RMC area to understand the baseline conditions of in RMC. The baseline information was gathered from series of activities; a structured questionnaire survey from 156 randomly selected households, interview surveys from 55 business and institutes, questionnaire survey in 6 recycling businesses, a waste composition and quantity survey at disposal site, waste material flow inventory survey at resource recovery center (composting and recyclable collection) of RMC. The baseline research revealed that RMC collects two categories of waste,biodegradable and non-biodegradable waste; however, the collected waste was directly dumped at an open dumping site (Kanadola site, owned by RMC).

The baseline study further revealed that waste generation is 53.76 ton per day, while waste collection amount is only 47.6 ton per day in which 47.3 ton of waste disposes at dumpsite. The per capita waste generation rate is 0.537 kg, and the bulk density of collected waste is 0.372 ton/m^3 with moisture content of 43-55%. The waste composition resulted from baseline study is illustrated in Figure 1. The waste composition in 2017 showed that 57% of the total waste is biodegradable (kitchen waste, grass & wood waste), while 32% is inorganic recyclable waste and 11% is residue. In comparison, a study conducted in Kandy city that is the capital of Central Province, showed that 75% of its total municipal waste was biodegradable waste with 13% recyclable waste and 12% other waste (JST-JICA, 2016).

Though waste collected by RMC has greater fraction of recyclable waste, the resource recovery was limited to scavenging on non-biodegradable waste by collection crew at the disposal site. There was a small box composting facility (about 2 ton/day) which was merely enough to handle organic waste collected from vegetable market (JICA, 2017); thus, biodegradable waste from residential and commercial premises disposed at open dumpsite. Eventually, biodegradable and other mixed waste dumped at Kanadola caused serious environmental issues such as leachate generation, vermin and offensive odours. Consequently, SWM action plan was developed in which expansion of composting system was recommended to handle

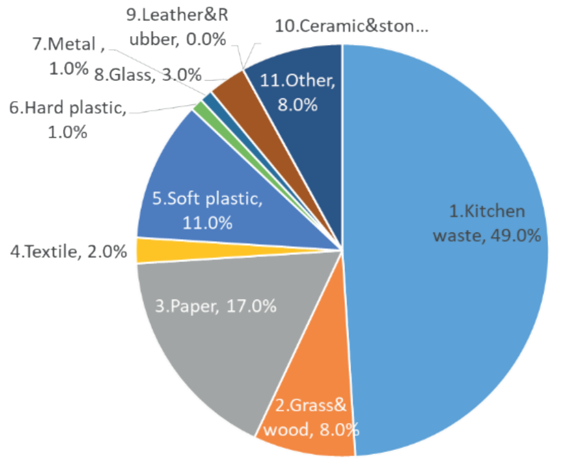

FIGURE 1: Waste composition in RMC (2017).

biodegradable waste from residential and commercial sectors.

2.2 Method of RMC pilot project

2.2.1 Area of RMC pilot project

The baseline study revealed that RMC area is consisted of 16 villages which are further amalgamated to four waste collection zones. Consequently, four villages were selected to implement source segregation pilot programs; Batugedara village (high and middle income households), Town North (commercial centre), Kospalawinna village (multi ethnic) and Muwagama village (middle and low income) based on their unique characteristics. As summarised in Table 1, the pilot project covered approximately a quarter of all households in RMC. According to the SWM action plan of RMC (2018-2022), prepared with the assistance of ReEB

TABLE 1: Numbers of households of the pilot project in RMC.

Area name	Household
Batugedara	423
Town North	698
Kospalawinna	1,158
Muwagama	514
Total	2,793

Waste, experiences in the first year (2018) were planned to be carried over to the second year (2019) to enable the expansion of solid waste separation and collection to the whole city to be completed in 2019, as planned.

2.2.2 RMC pilot project activities

The SWM situation before, the expected situation after the implementation of pilot project in December 2017 and the activities of the project are shown in Table 2. The pilot project increases separation categories from two (biodegradable and non-biodegradable) to five (Biodegradable waste, Plastic and polythene, Paper and cardboard, Glass, metal and coconut shells, other residuals). The project distributed appropriate containers for each waste generators; green colour plastic waste collection bins for biodegradable waste and different colour polysack bags for other non-degradable wastes. PHIs (Public Health Inspectors) and supervisors in RMC explained the new waste separation categories, ways to discharge (i.e. use of the distributed green-coloured bins and bags), and collection dates to residents and commercial premises. Household composting was promoted in pilot project areas expecting to increase the percentage of home composting from 6% to 10%. RMC identified 250 households in four (4) pilot project areas and distributed 250 home compost bins (Batugeda-

TABLE 2: Situation before and after the pilot project, and overview of activities in the project in RMC.

Stage of Solid Waste Management	Before pilot project (before December 2017)	Expected situation (2018)	Overview of activities
Generation	Two separation categories • Biodegradable (Mixed waste) • Non-biodegradable (Recyclable waste)	Five separation categories • Biodegradable waste • Plastic and polythene • Paper and cardboard • Glass, metal and coconut shells • Others	• Creation of leaflet and notice board • Awareness raising programme (door-to-door explanation, community meeting and school programme) • Procurement and distribution of separation bins (3,000 January, 3,200 in June and 1,940 in October 2018), poly sack bags (9,000 January, 9,600 in June and 5,820 in October 2018) • Monitoring
Discharge	No specific announcement by RMC (Shopping bag was being used mainly by residents and commercials)	• Green (residents only) separation bin for biodegradable wastes. • Three different coloured poly sack bags for recyclable wastes (residents only) • Any bag for other waste	
Collection and transportation	• Tractor for biodegradable waste • Three-wheeler and crew cab for non-biodegradable waste	• Tractor and three-wheeler with large collection bins installed for biodegradable waste • Three-wheeler and crew cab for recyclable wastes and other wastes	• Procurement of a three-wheeler, large collection bins and GPSs • Revision of collection and transportation plan • Monitoring
Treatment and Disposal	• Vegetable waste for composting by box compost system • Recovered recyclable wastes for sale • Biodegradable waste (mixed waste) for dumping • Lack of monitoring data	• Biodegradable wastes for composting by expansion of windrow compost yard • Recovered recyclable wastes for sale • Monitoring of treated and disposed waste amount	• Establishment of windrow composting system • Establishment of monitoring system at the site

ra: 48, Town North: 32, Kospalawinna: 90, Muwagama: 80) in March 2018, to those who: were willing to compost at home, had sufficient space, and/or had difficulty receiving waste collection services due to inaccessibility of collection vehicles. Information sessions (small-group meetings) were held at the time of distribution of home compost bins. The awareness focused on disseminating knowledge on home composting techniques such as: where to locate the bin, appropriate types and combinations (carbon: nitrogen ratio) of waste, appropriate waste moisture level, demarcation with Jeewakottu (traditional home composting), as well as troubleshooting for issues such as malodours and insects breeding.

ReEB Waste procured a three-wheeler (with large bins installed) to collect biodegradable waste from the narrow streets that are inaccessible by normal waste collection tractors. The in-house human resource capacity building was conducted by training for staff of RMC including supervisors, collectors, drivers and disposal site workers. In addition to the above mentioned activities in the target areas, the project also expanded the composting facility to a windrow system (15 tons/day) and regularized waste collection inventory recording at the Kanadola site to monitor the amount of waste received and disposed of.

There were no additional staff or workers hired for the pilot project. The following human resources were assigned to solid waste management department of RMC from September 2017 to March 2019: 150-160 collection workers, 20 drivers, one medical officer of health, five public health inspectors, and 20 field supervisors. All the awareness raising programmes, monitoring activities and surveys in the pilot project were conducted solely by these RMC staff.

2.3 Results of RMC pilot project and discussion

2.3.1 Changes in waste amounts resulting from RMC pilot project

Waste collection amount did not considerably change during the pilot project from 1,050.0 ton/month in January 2017 to 1,085.0 ton/month in March 2019. The total amount of waste disposed at open dump declined from 979.6 ton/month in December 2017 to 682.0 ton/month in March 2019 after implementing the pilot project in December 2017 (Figure 2). Contrarily, the collection of biodegradable waste increased from 77.5 ton/month in December 2017 to 325.5 ton/month in March 2019 while the amount of recyclable waste increased from 27.9 ton/month to 77.5 ton/month for the same period. The sequential changes of waste collection amounts in RMC from 2017 to 2019 is illustrated in Figure 2.

2.3.2 Change to waste separation percentage resulting from RMC pilot project

The effectiveness or neatness of biodegradable waste separation at household level was estimated by analysing

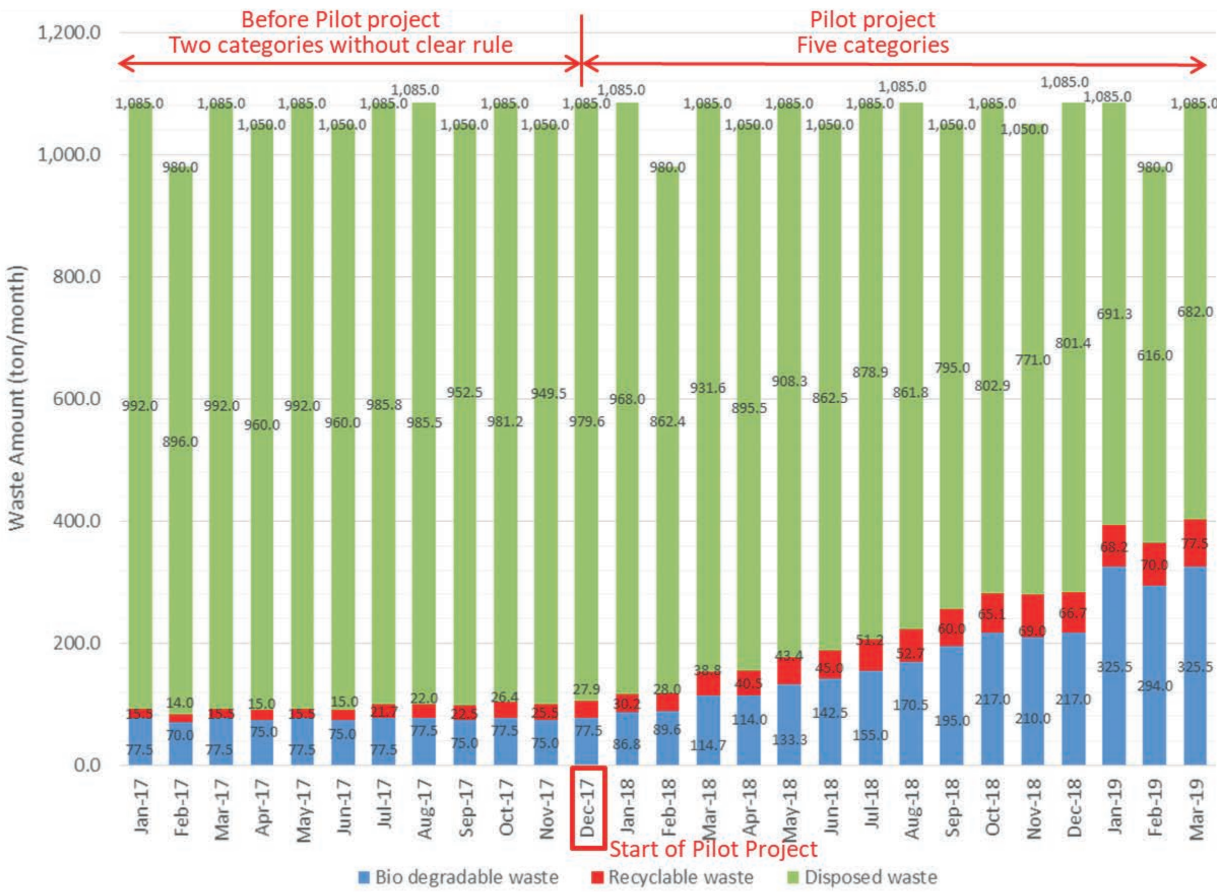

FIGURE 2: Waste amount in RMC from 2017 to 2019.

the composition of biodegradable waste collection before (September 2017) and after the project implementation (July 2018), and is illustrated in Figure 3. The results showed that amount of non-biodegradable waste in biodegradable collection was 39.9% in 2017; however, significantly reduces to 1.7% in July 2018 after implementing the pilot project. Figure 4 shows increasing recovery of recyclable waste fraction from 69.2% in July 2018 to 79.7 % in November 2018, due to sorting of non-degradable waste in to five categories. Thus, the pilot project activities contributed to improve the waste separation percentage within the biodegradable waste category as well as in the five categories of recyclable waste.

2.3.3 Outcome of the pilot project

In RMC, the pilot project was conducted based on the 2018-2022 SWM action plan, which was developed cooperatively by the ReEB Waste and RMC. Table 3 shows the targets of the action plan and the results thereof. The target number of households home composting was achieved by distributing 250 compost bins in the ReEB Waste project. The average amount of separated biodegradable waste received at the Kanadola site dramatically increased from 2.3 ton/day in 2017 to 5.1 and 10.5 ton/day in 2018 and 2019 respectively – which met the targets of the action plan. Though higher amount of recyclables collected in 2017 was higher due to poor quality of source separation, the quality of separated recyclables increased from 2018 to 2019. Increase of capacity of centralised composting facility considerably decreases the amount of waste disposed at final disposal site from 35.8 ton/day in 2017, to 28.7 ton/day and 22.1 ton/day in 2018 and 2019, respectively.

2.3.4 Discussion

The main reason for the reduction of final disposal amount of waste in the RMC pilot project was that biodegradable waste, which accounts for the largest proportion of waste generation, was thoroughly separated at the generation source and intermediately treated by centralized composting. By preferentially separating biodegradable waste at the generation source, the handling of recyclable waste became easier, and as a result, the amount of recyclable waste separated at the generation source also increased. It was found that distribution of separate bins for biodegradable waste and bags for recyclable waste and thorough implementation of separation, discharge and collection rules increases the effectiveness of source separation. RMC had a sufficient financial allocation and human

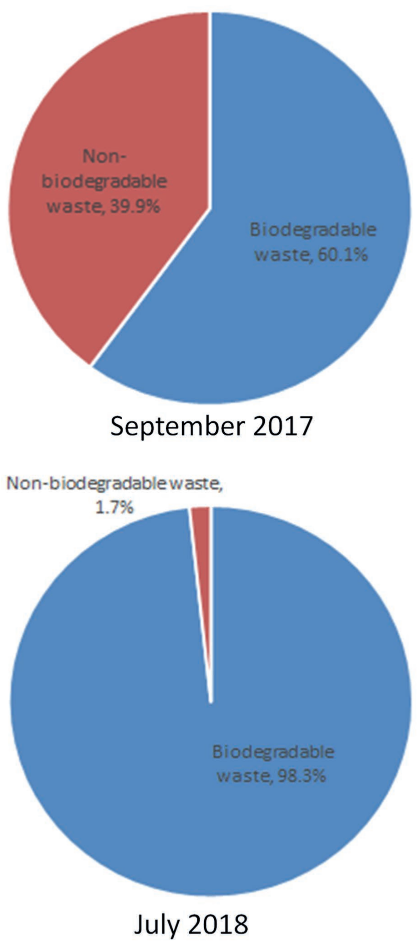

FIGURE 3: Waste separation percentage between biodegradable waste and non-biodegradable wastes of "biodegradable waste" separation category.

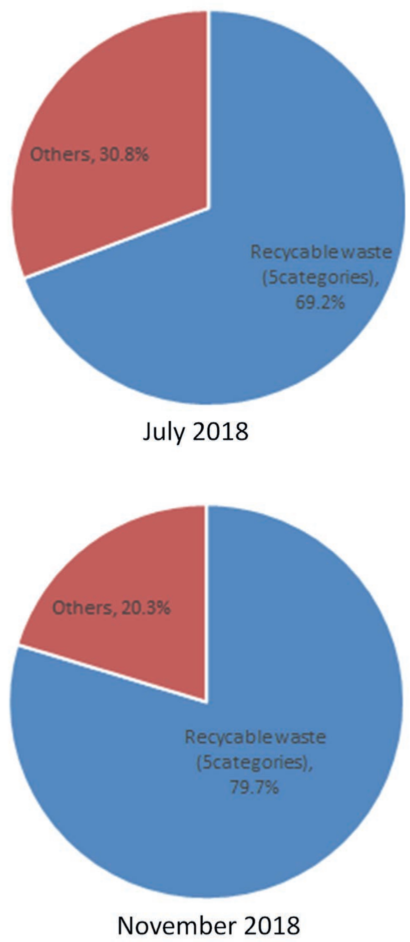

FIGURE 4: Waste separation percentage between recyclable waste and others wastes of "5 categories recyclable waste" separation category.

TABLE 3: Targets of Action Plan for RMC and results.

Items	2017	2018		2019		2022
		Target	Result in November	Target	Result in March	Target
Percentage of households doing home composting (%)	6.0	6.9	6.9	8.0	NA	10
Receiving amount of biodegradable waste at the site (ton/day)	2.3	5.3	5.1	8.6	10.5	15
Compost production at the site (ton/month)	2.3	5.3	2.5	8.6	4.2	15
Receiving amount of recyclable items at the site (ton/day)	2.9	3.7	1.6	4.9	2.9	14.9
Final disposal amount (ton/day)	35.8	35.3	28.7	-	22.1	20.1

resources for solid waste management, so it was possible to simultaneously instruct the source separation of biodegradable waste and recyclable waste into five categories.

3. PILOT PROJECT AT KATARAGAMA PRADESHIYA SABHA (CASE STUDY 2)

3.1 Overview of Kataragama Pradeshiya Sabha

3.1.1 Outline of KPS

Kataragama is a township with a population of 22,640 (in 2017 (JICA, 2019)) located in the southern-most part of Uva Province on the island of Sri Lanka and borders the Yala National Park. The town is home to one of the most important holy sites in Sri Lanka and pilgrims and tourists from all over the country and overseas gather for the annual Perahera festival (religious procession) held over two weeks in July/August.

3.1.2 Baseline study in KPS

In 2017, a baseline study on waste management practices was conducted in KPS area to understand the baseline conditions of in KPS. The baseline information was gathered from series of activities; a structured questionnaire survey from 161 randomly selected households, interview surveys from 62 business and institutes, questionnaire survey from one recycling businesses, a waste composition and quantity survey at disposal site, waste material flow inventory survey at resource recovery centre (composting and recyclable collection) of KPS.

The baseline study revealed that the most serious issue is the management of final disposal site in Galapitiyaya, which, although permitted to be used for landfill site by the government, was operated in an open dumping manner. The site locates within a reserve forest, thus wild animals, especially elephants frequently scavenge at the site. Moreover, waste generation during the festival season increase by two times of daily average and eventually KPS opt to dispose waste in temporally dumpsites along the river. According to a survey conducted by a JICA study, the waste collection amount was approximately 3.5 ton per day in 2015 (JICA, 2016). The study further revealed that the waste generation rate was 0.620 kg per capita, while the bulk density of collected waste was 0.598 ton/m^3 and moisture content was 43-55%. By the time ReEB Waste started, KPS had established a small resource recovery facility with a box composting system and recyclable sorting and bailing machine. However, there were no clear rules or advices given to waste generators on segregation. The survey showed that out of total, 57.3% was kitchen waste; however more than a half (32.7%) of kitchen waste was in non-biodegradable waste collection due to ineffective separation at source. This indicated that the waste separation at source was not effectively practiced by generators. For reference, in the neighbouring municipality of Hambantota, 54% of its total waste was biodegradable waste, 6% was recyclable waste and 40% was other waste in 2016 (JST-JICA, 2016). In another neighbouring municipality, Kalumunai, 71% of its total waste was biodegradable waste, 16% recyclable waste and 17% other waste in 2010 (Ariyawansha et al., 2010).

The waste composition in KPS is illustrated in Figure 5.

3.2 Method of KPS pilot project

3.2.1 Area of KPS pilot project

The baseline study revealed that KPS area is consisted of 5 villages and two main townships, Kataragama and Sella-Kataragama which are 5 km apart. The KPS area had been divided to six waste collection zones. Two of the six waste collection zones in KPS, Zone 3 and Zone 4, were targeted in the pilot project. In Zone 3, 52 guesthouses/hotels and shops are receiving waste collection service. Zone 3 is close to two popular Hindu temples and has small market areas where shops sell souvenirs and offerings (flowers and fruits) to tourists. Waste collection in Zone 4 covers 64 households and 54 guesthouses/hotels and small shops. Zone 4 is close to the city-centre of KPS.

3.2.2 KPS pilot project activities

KPS took a two-step approach to establish an effective source segregation program. The first step was started in November 2017, and the aim was to fully establish a two-category separation system where biodegradable waste and other waste would be separated. This involved disseminating waste separation rules to generators by distributing a leaflet to every household and business, holding community meetings, and organizing field visit for community members to KPS's compost facility. Appropriate waste collection bins, 20-liter bins for households and 45-liter bins for guesthouses/hotels, were distributed to every household and businesses. New waste collection schedule was introduced where collection vehicle collects degradable and non-degradable waste on alternate days. KPS assigned a designated field staff in each zone to follow the collection vehicles to monitor implementation and to correct any malpractice.

After the two-category separation had become well established, second step started in July 2018 to separate

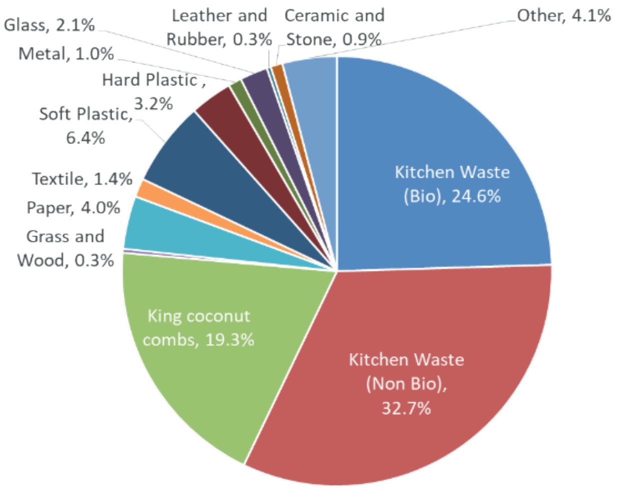

FIGURE 5: Waste composition in RMC (2017).

waste into three categories: biodegradable waste; valuable recyclable that can be sold through identified dealers which include cardboard, metal cans, hard plastics, plastic drink bottles (PET), glass, paper (newspaper, magazines, dry and clean paper) and coconut shells; and other residual waste. The residual waste has is to be disposed at final disposal site.

At the onset of launching the second step, the waste collection schedule was revised. Collection vehicles are sent to each zone on Monday, Wednesday, Thursday, and Sunday for biodegradable waste; Thursday to Zone 4 and on Sunday to Zone 3 for valuable recyclable collection, and other days for residual waste collection. Another leaflet which explains the separation categories and collection schedule was distributed through door-to-door visits. During Step 2 implementation, the newly upgraded landfill opened in September 2018 and representatives of the community were also invited to the opening ceremony to keep them informed of how the waste separated by residents would be treated. All waste collection staff and drivers were trained on waste segregation thus they were instrumental in taking the message to residents on correct source separation at source. Table 4 shows the waste management condition prior to launching the pilot project (2017), project expectations after implementation, and the activities of the project.

During the implementation of the pilot project, ReEB Waste made several inputs towards achieving the project aims amongst residents in target communities, including: organization of awareness activities such as community meetings, house-to-house visits for distributing leaflets and compost site visits; and distribution of 45-liter (to businesses) and 20-liter (to households) bins for separately discharging biodegradable waste. Twenty-five 45-liter bins were distributed in February 2018 to Zone 3 (predominantly guesthouses), and 15 bins were distributed to Zone 4 in June 2018, followed by a further 10 bins in 2019 in response to individual requests in zones 1, 3 and 4. Forty-two 20-liter bins were distributed in March 2018 and a further three in January 2019 to households in Zone 4.

3.3 Results of KPS pilot project and discussion

3.3.1 Changes to waste amounts due to festivals and management issues

Total amount of waste collected by KPS from 2017 to 2019 and changes in waste separation are shown in Figure 6. It should be noted that KPS hosts a large pilgrim/tourist population during the Perahera festival season in July and August every year and the average monthly amount of waste discharged almost doubles during the festive period compared to the first half of the year. The monthly average

TABLE 4: Situation before and after the pilot project, and overview of activities in the project in KPS.

Stage	Before pilot project (before December 2017)	Expected situation	Overview of activities
Generation	• Two separation categories in some areas • Biodegradable • Non-biodegradable (mixed waste) • No separation in some areas • Mixed waste	• Step 1: two-category separation • (November 2017-) • Biodegradable waste • Others • Step 2: three-category separation • (July 2018-) • Biodegradable waste • Recyclables • Other waste (going to disposal site)	• Creation of leaflet and notice board • Awareness raising programme (door-to-door explanation, community meeting and school programme) • Procurement and distribution of biodegradable separation two types of bins: 20-liter for household and 45 litre for guesthouse • Procurement and distribution of compost bins • Monitoring
Discharge	• No specific announcement by KPS	• Both 20 litre and 45 litre bins for biodegradable waste have a lid to mitigate the smell of waste and keep animals away. • Any bag for other waste	
Collection and transportation	• Tractor for biodegradable waste and mixed waste	• Modified tractor with installed partition for separation of categorised waste.	• Training for collection staff and drivers to recognise the separation categories • Revision of collection and transportation plan • Monitoring
Treatment and Disposal	• Vegetable waste for composting by box compost system • Recovered recyclable wastes for sale • Biodegradable waste (mixed waste) for dumping • Lack of monitoring data	• Biodegradable wastes for composting by expansion of windrow compost yard • Recovered recyclable wastes for sale • Monitoring of treated and disposed waste amount	• Establishment of windrow composting system • Installation and operation of weigh bridge at compost plant • Establishment of monitoring system at the site

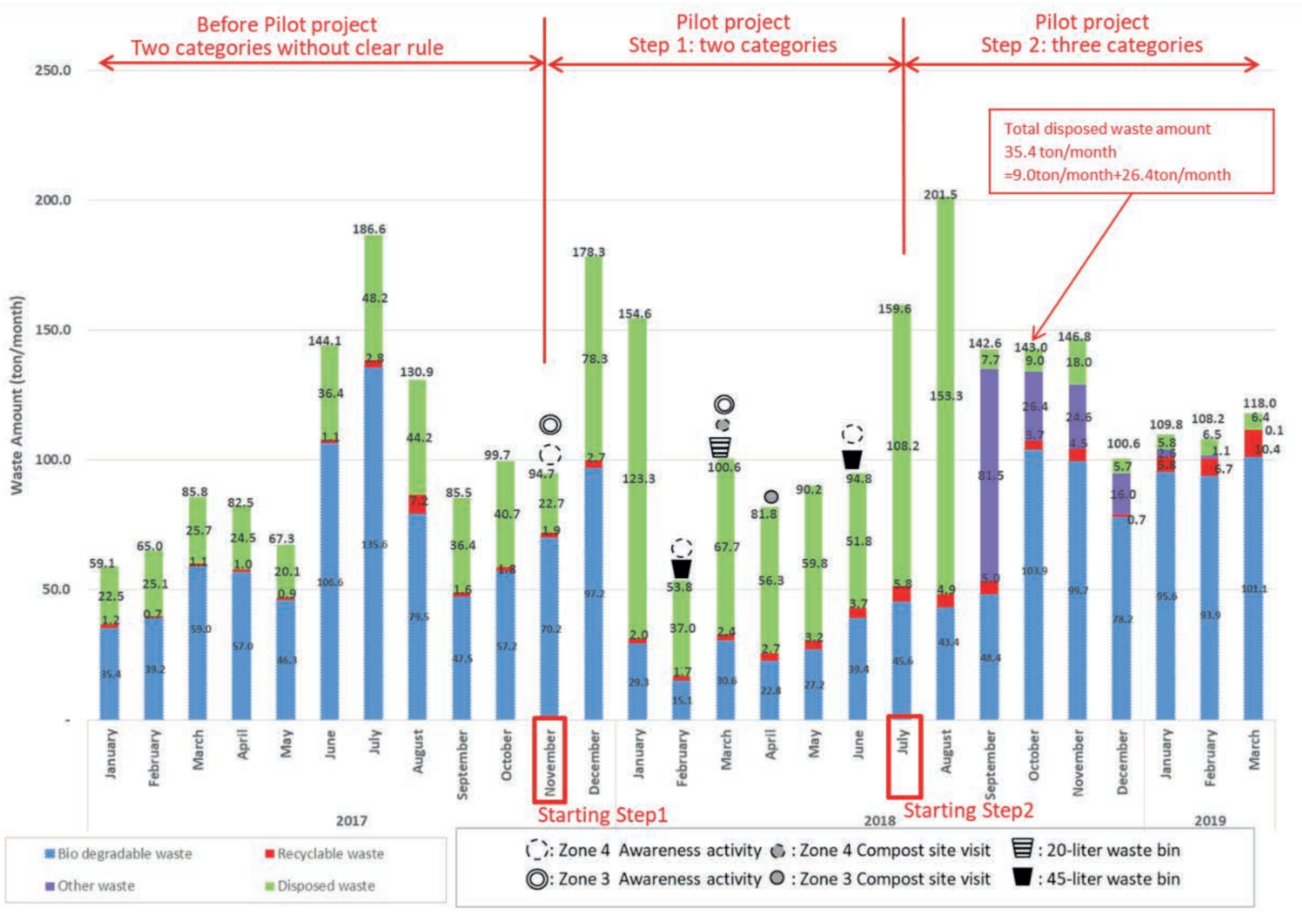

FIGURE 6: Waste amount in KPS from 2017 to 2019.

amount for the first half of 2017 was 84.0 ton/month, while the monthly average for July and August 2017 was 158.8 ton/month. The monthly average amount for the first half of 2018 was 96.0 ton/month, while the monthly average for July and August 2018 was 180.6 ton /month. Moreover, since extra solid waste was taken to KPS from some other local authorities in December 2017 and January 2018, the total waste amount was higher than the normal condition in December and January. On the other hand, number of temporally collection workers decreased by 5 workers (out of 26) from February to June 2018 due to internal management issues at KPS. The labour shortage led to lower the efficiency of overall waste collection in KPS area as well as the efficiency of recycling facility. Consequently, the total waste collection and biodegradable waste collection decreased and the waste disposed to the final disposal site increased, despite the pilot project being implemented. Therefore, in this paper, the impact of the pilot project on waste reduction is analysed excluding the record for July and August each year and for December 2017 to June 2018.

3.3.2 Changes in waste amounts resulting from KPS pilot project

Comparing the amounts of discharged waste during the three months from January to March 2017 (before the pilot project) and the amounts in the same three months in 2019, the amounts of discharged waste increased from 59.1 ton/month to 109.8 ton/month in January, from 65 ton/month to 108.2 ton/month in February and from 85.8 ton/month to 118.0 ton/month in March. KPS did not make any specific changes to expand their waste collection services, thus these increases can be considered as changes in the discharge manner of residents.

Biodegradable waste amount and recyclable waste amount show steady increases during the first half of 2017. In November 2017 (start of the pilot project), the biodegradable waste collection was 70.2 ton/month that increased to 99.7 ton/month in November 2018 at the end of the Step 2. Amount of recyclable waste collection steadily increased from 1.9 ton/month in November 2017 to 4.5 ton/month in November 2018. The increase of biodegradable and recycling waste resulted a decrease in mix waste disposal from 81.5 ton/month at the beginning in September 2018 to 24.6 ton/month in November 2018. Amount of waste disposed at final disposal site decreased from 40.7 ton/month in October 2017 to 35.4ton/month in October 2018,. The further decrease of final disposal amount occurred after December 2018 is due to expansion of three-category separate collection scheme to other collection zones.

3.3.3 Changes to waste separation percentages resulting from KPS pilot project

Before and during the pilot project waste composition surveys were conducted three times: first in October 2017, before the pilot project; second in June 2018, during Step 1; and third in November 2018, during Step 2. These surveys measured the composition of the waste collected separately as "biodegradable" and "non-biodegradable (or other)" wastes from Zone 3 and Zone 4.

Before the implementation of the pilot project, during the collection and loading to vehicle, collection workers separated the unsorted waste as biodegradable and other waste by opening the waste bags. Thus, the waste brought into the compost site was already separated into two categories; however, present in two comportment separated by a wooden flange in tractor trailer. At the first stage of pilot project, new collection method was introduced where only one type of waste is collected during collection, the other type of waste is collected separately in another trip or else another day. On the other hand, slowly degradable organic waste such as hardwood, coconut shells and fibrous banana stems were categorised as other waste that to be disposed at landfill. Furthermore, residents did not allow to dispose garden waste and tree trimmings along with biodegradable waste due to bulkiness, thus only kitchen and food waste were received at composting site. Recognizing the importance of having dry and bulky organics for composting process, at the second stage of project, a small shredding machine was installed at composting facility to reduce the size of garden waste and tree trimmings. Once the shedding machine was installed, KPS started to accept garden waste and tree trimmings, which can be discharged together with kitchen waste. The composition changes in biodegradable waste in Zone 3 and Zone 4 are illustrated in Figure 7 and Figure 8 respectively.

During Step 1, valuable recyclables were not collected separately but mix with the other residual waste that cannot be taken to composting facility. However, since a private recyclable collection business had started to collect valuable recyclables in parallel to first stage pilot project implementation, the composition survey showed that amount of valuable recyclables are low (2018). By the third quarter of 2018, KPS implemented collection of valuable recyclables on a designated day of the week, consequently the composition survey in 2018 showed that the non-biodegradable waste was mainly consisted of plastic films due to effective diversion of garden waste and valuable recyclables for resource recovery.

At the end of Step 1 (at the time of the second waste composition survey), the percentage of kitchen waste was small in other waste, and no kitchen waste or garden waste was found mixed in other waste by the end of Step 2 (at the

FIGURE 7: Changes in biodegradable waste in Zone 03.

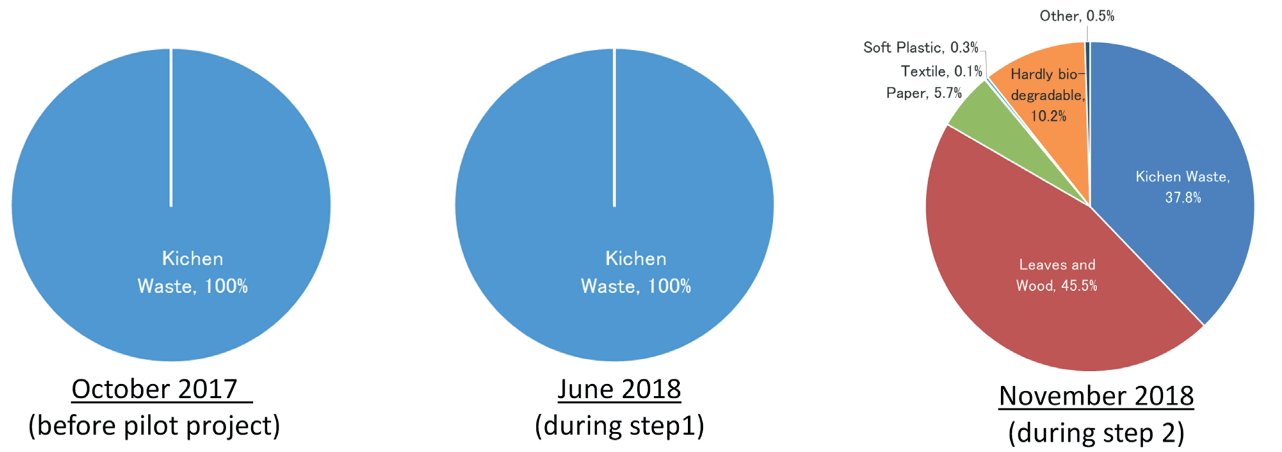

FIGURE 8: Changes in biodegradable waste in Zone 04.

time of third waste composition survey). Moreover, difficult to degrade waste, which is supposed to be "biodegradable waste" but had been instructed to dispose of as "other waste", was also being separated as "biodegradable waste" by the time of the third waste composition survey. These results show the high degree of acceptance of separate collection by the community as a result of the pilot project. Changes in non-biodegradable waste in Zone 3 and Zone 4 are illustrated in Figure 9 and Figure 10 respectively.

3.3.4 Outcome of the pilot project

In KPS, the pilot project was conducted based on the SWM action plan prepared for 2018 to 2022 period, which was developed in cooperation with ReEB Waste and KPS. Table 5 below shows the targets of the action plan and the results thereof. Average amount of separated biodegradable waste being received at the disposal site increased from 2 ton/day in 2017 to 3.4 ton/day in 2018 and 3.5 ton/day in 2019, which met the targets of the action plan. The average amount of separated recyclable wastes also increased from 0.1 ton/day in 2017 to 0.2 ton/day in 2018 and 0.4 ton/day in 2019, although this did not yet meet the target of the action plan. Final disposal amount also decreased from 1.4 ton/day in 2017 to 0.3 ton/day in 2018 and 0.3 ton/day in 2019 due to increase of composting and resource recovery at the disposal site. As an element of waste management plan, in year 2017, the open dumpsite was rehabilitated and upgraded to an engineered landfill by ReEB Waste in project spending 35 million Sri Lankan Rupees (US$ 200,000). Moreover, the lifespan of landfill increased from five years of designed capacity to ten years as a result of reducing the disposal amount through resource recovery activities. The cost of construction per “lifespan year” is thus reduced from approximately 7 million Sri Lankan Rupees (US$ 40,000)/lifespan year to 3.5 million Sri Lankan Rupees (US$ 20,000) /lifespan year.

3.3.5 Discussion

The main reason for the reduction in the final disposal amount of waste in the KPS pilot project was that biodegradable waste, which accounts for the largest proportion of waste generation, was thoroughly separated at the generation source and intermediately treated. Similar to pilot project in RMC, it was found that distribution of separate bins for biodegradable waste and implementation of discharge and collection rules increases the effectiveness of source separation. However, since KPS did not have sufficient budget and human resources for solid waste management, the primary objective was to separate the biodegradable waste at generation source, and the quantity of valuables (recyclables with market value) separated at source and collected was gradually increased. It is im-

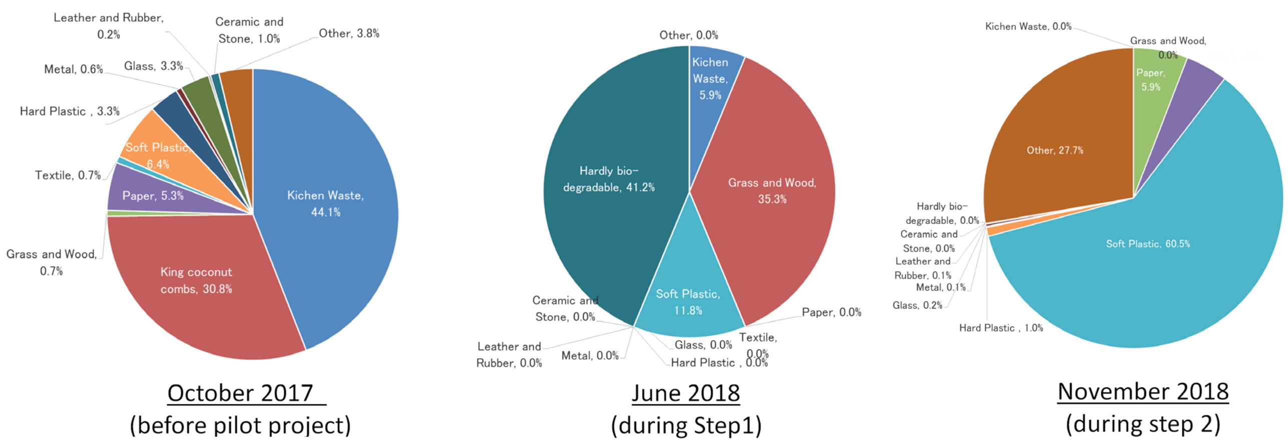

FIGURE 7: Changes in biodegradable waste in Zone 03.

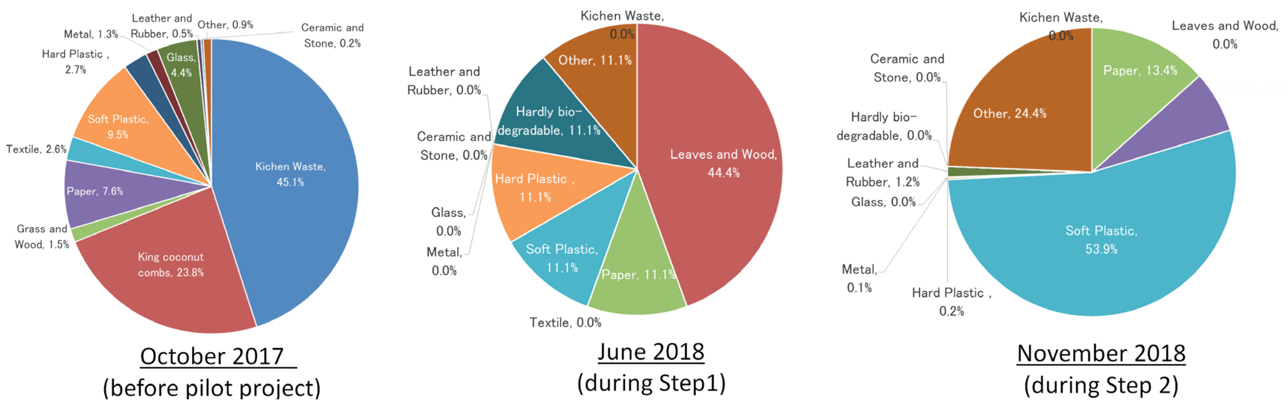

FIGURE 10: Changes in “other waste” in Zone 04.

TABLE 5: Targets of Action Plan for KPS and results of average amount.

Items	2017	2018		2019		2022
		Target	Result in November	Target	Result in March	Target
Collection amount of biodegradable waste (ton/day)	2.0	2.5	3.4	2.9	3.5	3.9
Collection amount of recyclable waste (ton/day)	0.1	0.7	0.2	0.7	0.4	0.7
Final disposal amount (ton/day)	1.4	8.0	0.3	7.8	0.3	7.4

portant to determine the number of categories and types of recyclable waste to be separated according to the local authority's implementation capacity, with priority given to the separation of biodegradable waste.

4. CONCLUSIONS

The changes of waste composition and final discharge amounts at the disposal sites indicate the pilot projects are contributing to promote 3Rs in pilot project areas of RMC and KPS. In addition to pilot project areas, local authorities are expanding the promotion of 3Rs to other areas based on the successful achievement of waste reduction, resources recovery and decrease of final disposal amounts.

It was also found that reduction of waste amount disposed at the final disposal site does not require segregation of waste into many categories, but a remarkable change can be made if waste is separated to two categories; biodegradable waste and non-biodegradable waste. It is also learned that on-site training and immediate instruction and demonstration that combined with distribution of printed instructions by collection workers are more effective way of disseminating the knowledge of waste separation. Also, it was learnt that supply of waste collection bins and bags facilitate rapid establishment of source separation; however, the knowledge dissemination, direct supervision and cooperation of collection workers by accepting only the separated waste during the collection are also important elements to achieve targets of source separated waste collection.

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'BAD' TRASH: PROBLEMATISING WASTE IN BLANTYRE, MALAWI

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ABSTRACT

'Waste' is everywhere, a common aspect of daily life in both the West and the Global South. However, the ways in which we as individuals understand it as a problem is far from universal. It does not exist independently from the people it affects, rather, waste, as a problem, is continually made and remade through human practice. The purpose of this article is to explore how and why certain 'waste' items are and become understood as problems. We adopt Foucault's (1984) notion of 'problematisa-tion', as an analytical lens for conceptualising processes of problem formation through the eyes of two different groups working within and on the margins of Mzedi Dump Site in Blantyre, Malawi: subsistence maize growers and informal waste pickers. Drawing on extensive qualitative and ethnographic fieldwork, our findings suggests that for those working at Mzedi, waste problematisations are shaped by the tangible: the visible, and often painful impacts that Mzedi's hazards have on their lives and livelihoods. However, the ultimate problematisation of waste lies in its utility, i.e. 'good' waste, is internalised based on its value. 'Bad' trash however, is problematised because it has no value, and is therefore considered useless, a problem taking up time and space that could be utilised more profit-ably. Understanding these processes of problem formation, and the degree to which waste problematisations are personal and/or socially constructed, has important ramifications for the adoption of appropriate waste management strategies and should inform a more nuanced and inclusive waste management studies discourse.

1. INTRODUCTION

This place has changed. In the past they used to dump good things here, such as plastic sheeting and paint, but now they have stopped bringing those things... They should dump good trash, not just the waste items they are dumping nowadays, so that when we collect [the items that we want] we will somehow be happy [1].

Waste is a universal facet of human existence. A used paper cup, a polystyrene takeout container, or a discarded plastic shopping bag, these items and many more, make up the collective detritus of our civilisation, which we are constantly forced to confront and manage. However, as Kennedy (2007) notes in his book An Ontology of Trash, most of our refuse, aside from perhaps a scuff or a bit of dirt marring its surface, undergoes little qualitative change in its transition from desirable commodity to something that must be separated from society, hid, or destroyed. As Kennedy (2007, p. xv) wisely asks, "what does the brief act of consumption involve that could cause such powerful ontological effects?" However, as the quote at the top of the page suggests, spoken by an informal waste picker at Mzedi Dump Site (the only municipal-run waste management facility in Blantyre, Malawi), within the Global South, and particularly amongst its poor, ontological distinctions between commodity and 'waste' are less rigid, with objects often living multiple lives of utility; even, as the quote illustrates, for the small percentage of domestic waste that ultimately reaches a dump site. Moreover, as those who dwell within one of Latin America, Asia, or Africa's emerging cities can attest, waste, in the form of litter, is part and parcel of the urban fabric, an element of urban life that must be constantly negotiated, utilised, or, more likely, ignored by city dwellers.

Although Kennedy's work speaks more to why, specifically, a late-stage capitalist West wastes, and the ontological implications of this relationship to Being that this wastage entails, he makes a useful ontological distinction between 'waste' and 'trash' that resonates more broadly. To Kennedy (2007), 'waste' is a subjective notion: beauty is in the eye of the beholder. With 'waste', devaluation is relative and personal: one individual's trash may be another's treasure. 'Waste' has value to the right person who knows how to harness or appreciate it. As Reno (2015, p. 559) articulates, nothing is 'waste' in general, only in particular. 'Trash'

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however, lacks this nuance. If 'waste' results from a relative, subjective devaluation, then 'trash' is the product of unconditional, absolute devaluation (Kennedy, 2007, p. 10). 'Trash' holds no value, and no longer has a place within society. Although the discursive elements of this distinction may not translate neatly from English, to say, Chichewa, the predominant language of Southern Malawi, or one of many other numerous languages spoken throughout the Global South, the divergence allows for more flexible conceptualisations of the journey an item takes from usable commodity to unusable castaway within these under-investigated contexts [2]. However, once an object is understood as 'trash' within Kennedy's conceptualisation of the term, at what point does it become an issue that must be dealt with- either removed, destroyed, or avoided altogether? In other words, for individuals within the Global South, when does 'trash' become a problem, how do these processes of problem formation occur, and how can they be conceptualised?

The purpose of this article is to explore these processes of problem formation through the experiences of two separate, but often interdependent, communities within Blantyre, Malawi: informal waste pickers who salvage saleable waste items for their livelihood within Mzedi dump and local subsistence farmers who grow and harvest maize on and along the margins of the same site. Drawing on extensive qualitative, ethnographic fieldwork within each group, the article explores, how for differently placed people negotiating the same space and confronting similar 'waste' hazards, problem formation remains personal and contextual, informing how each individual navigates and utilises Mzedi for their own purposes. Moreover, responding to a lack of critical social theory to conceptualise these processes of problem formation, this study adapts Michel Foucault's notion of problematisation, first invoked by Foucault as a pathway for critical analysis of the city. Foucault's notion of problematisation is useful a lens through which to observe processes of problem formation because it emphasises the connection between problems and the individuals they affect. Foucault's writings have been applied extensively within the social sciences to understand the process of problem formation at a societal level, particularly as a mode of critique within urban studies. However, this article considers more recent contributions by Bacchi (2015), Barnett and Bridge (2016), and Oberg (2019) which attempt to operationalise the term as an analytical tool to understand processes of problem formation amongst disparate urban contexts and at a personal, rather than solely societal, level. Our findings suggest that Foucault's concept of problematisation is a useful lens through which to interpret the ways in which different individuals 'problematise' the various 'waste' and 'trash' items within Mzedi. Foucault's conception of problematisation, and subsequent elaborations on the notion, allow for a situational analysis of the formation of problems (Barnett & Bridge, 2016). As with individual ontological formations around 'waste' objects, personal conceptions of problems do not exist, as Oberg (2019) identified, independently from the people they affect, rather, these conceptions are continually shaped and reshaped through human agency, need, and livelihood practices. Understanding these processes of problem formation, through which differently placed individuals problematise 'trash', and the degree to which these problematisations are shaped by culture, values, and other contextual factors, has important ramifications for the adoption of appropriate waste management or circular economy strategies. This investigation is particularly important within the context of Malawi, and the Global South more broadly, and should inform a more nuanced, and less normatively Western waste management studies discourse.

2. THINKING PHILOSOPHICALLY ABOUT WASTE

How does an object, once a desirable commodity, become 'waste', and how does this process occur within different contexts, continents or ontological formations? Despite the ubiquity of waste within our modern existence, and since Mary Douglas' (1966) famous characterisation of dirt or waste as "matter out of place," there have been few philosophical deconstructions of the phemonological and ontological links that bind humans and waste, particularly contextualised within Africa or the Global South. Nonetheless, with the recent emergence of Discard Studies [3], a small but growing voice within broader waste management academic discourse, there has been increased interest in the wider role of society and culture in defining and structuring attitudes towards, and behaviours around, waste, however that concept may be construed. This section seeks to provide a brief overview of contemporary philosophical, ontological, and epistemological deconstructions [4] of waste in order to contextualise the recent developments taking place within the discipline and to situate this piece both ontologically and conceptually.

Kennedy's (2007) writings predate the more recent interpretive turn within waste management discourse driven by Discard Studies. Speaking to, yet departing from Heidegger's ontological project, Kennedy interrogates the historical phenomenon of disposable commodities. To Kennedy (2007) trash represents the collapse of the ontological structure of human Being: a failure of humanity at a fundamental level. Waste reflects our own short-comings, our failure to preserve the value originally invested in objects—all we can do as machine-minded, uncaring consumers is purchase, consume, trash, and purchase anew (Kennedy, 2007, pp. 181-182). Both Hird (2012) and Spelman (2011) speak to Kennedy's conceptualisations of wastage in order to provide epistemological commentary on 'knowing' waste, rooted in their respective disciplines, and invite the reader to interrogate their own personal understandings of waste. Likewise, Moore (2012) highlights the ways in which different views of what waste 'is', are productive of many forms of scholarship that have the potential to disturb certain taken-for-granted ideas about values, politics, and socio-spatial relationships. Moore proposes that that waste be thought of as parallax object—not as a binary good or bad, but as something that 'disturbs the smooth running of things'. Reno (2014) speaks directly to Douglas' aphorism, attempting to reframe conceptualisations of 'waste' without dirt as a starting point. To Reno (2014), it is worth considering how waste is interpreted from trans-species perspectives, and how these interpretations serve to mediate transactions between living beings. Focusing on faeces, Reno argues that objects, commonly considered 'waste' are not arbitrarily classified, but purposefully assigned, and thus symptomatic of life's spatio-temporal continuation. In other words, waste is the sign of the living; waste, in this instance, faeces, as the evidence of the lifeform's continued existence. Though Reno does not speak to the ultimate item of waste we leave behind when we exit that existence, our corpses, it is a creative reconceptualisation, nonetheless.

To Kennedy, trash and wastage represent the ontological collapse of our human modern civilisation. However, to Viney (2014) an interest in trash and waste is rather more ageless, representing one of the central aspects of human existence. Drawing on literary, artistic, and socio-cultural tradition from throughout history, Viney (2014) argues that the concept, like the thing, is socially constructed within a given time and space, and that our momentary relations with such items are managed and acknowledged in nuanced ways. Focusing on one specific waste stream, Van Bemmel and Parizeau (2019) examine the materiality of food waste. They point out that often, the lines that delineate edible food from waste are blurred. Moreover, their study suggests that socio-cultural norms, such as aesthetic standards, and systematic factors, such as temperature regimes or best before dates, are central determinants to food making the ontological transition to waste (Van Bemmel & Parizeau, 2019). This transformation from food to waste is not unidirectional however, as Van Bemmel and Parizeau (2019) describe instances of 'rehabilitation', where rejected produce is transformed into higher value processed products or past-date grocery items are scavenged from dumpsters ('dumpster diving'), recalling Kennedy's (2007) more subjective and relational deconstruction of 'waste'. Next, Ablitt and Smith (2019) examine the 'practical objectivity' of objects as they are handled through the everyday work of street cleaners in Gibraltar. Also starting from Douglas' aphorism on 'dirt', Ablitt and Smith (2019) reject her characterisations of the term as a basis for broad theorisation of social structure and culture, instead speaking to the ways in which 'dirt' and 'waste' are locally and practically assembled and negotiated through quotidian interactions. Their findings suggest that on the streets, 'waste' is seeable through a relational contexture of visually available elements within a given street scene (Ablitt & Smith, 2019). More interestingly however, they demonstrate a possibility for certain objects to also be 'seen' as bound to associated categories of people, a conclusion that deserves further investigation within different contexts.

Finally, a number of contemporary reflections on 'waste' have attempted to break free from the traditional western and euro-centric outlook of waste management discourse in order to include valuable perspectives from the Global South, as well as centres of informality. For instance Oloko (2018) examines print media reports in Nigeria to examine the ways in which 'dirt', 'waste', and 'garbage' function as relational and socially constructed concepts within intersecting sanitation and social contexts. Oloko's (2018) analysis reveals that media coverage on issues of waste or sanitation often carries discursive undertones of power, in which emotive terms related to waste are often used to justify the exclusion of marginalised groups or individuals, such as sex workers or the poor, from general society. He concludes that in this context waste' and 'garbage' are not merely relational objects, but rather evocative terminologies adopted by the healthy and powerful to mark out a space of physical, moral and political non-belonging inhabited by the weak or marginalised (Oloko, 2018). Writing about the Eastern Himalayas, Wang (2019) examines how differing interpretations of 'trash' adds to the complexity of relations between Han Chinese and indigenous Tibetans. According to Wang (2019), Tibetan cultural practices include a number of acts construed by Han Chinese as littering, creating disputes between the two groups over what is sacred and what is trash. Wang (2019) argues, however, that these 'littered' objects should not be considered as such, rather they should be conceptualised as people, as their purpose is to mediate the reciprocal relationship between humans and the environment—conclusions, that speak to the need for looser and more contextually appropriate conceptualisations of personhood within waste management practices. Lastly, a number of studies have explored the reconceptulisation of waste as a resource through the eyes and everyday experiences of waste pickers. To these individuals 'waste' is a relational concept, both a hazard and an opportunity. Within the waste picker community, recyclables are symbols of accumulation and individual ability; material testimonials to hard work and business networks (Ka-Ming & Jieying, 2019). Yet, as Gutberlet and Uddin (2017) describe, waste, even household waste, can pose significant risks to those who handle it. Moreover, reflecting Ablitt and Smith's (2019) findings, which suggest that certain objects can be 'seen' as bound to certain categories of people, Ka-Ming and Jieying (2019) show that amongst waste pickers different types of waste can be powerful signifiers to identity and internal status, with poor migrants being associated with general rubbish, but more established and visibly successful individuals monopolising trade in more lucrative waste streams. These previously cited contributions suggest that waste is often a fluid concept within the Global South, and that waste management studies discourses are embracing more flexible constructions of waste and society, however there remains significant scope for further philosophical, ontological, and epistemological deconstructions within these contexts.

3. PROBLEM FORMATION AND WASTE PROBLEMATISATIONS

Once an object is considered trash, what then? Many a city street, in both the Global North and the Global South, is littered with objects that most city dwellers would consider 'trash' in Kennedy's reckoning of the term, yet for the most part, they are ignored, with residents content to pass them by, or at most, kick them out of the way. An occasional conscientious passer-by may pick up an empty bottle and deposit it in the bin, but would they do the same for chewed gum, or for leaf litter? Either these objects, individually or collectively, are not construed by the passer-by as a problem, or as not enough of a problem do anything about. But what if there is broken glass on the sidewalk? Or a used hypodermic needle? Or perhaps there are also children playing nearby? Do these objects now become more than nuisances that can be ignored, transforming into problems that must be dealt with or actively avoided? Similarly, for the individual accustomed to litter on their streets, how much is too much, where a nuisance is transformed into a community issue, which becomes the object of collective or political action? When and how do these processes of problem formation occur, and to what degree are they personal and/or socially constructed. In other words, how do individuals problematise waste, and how do these individual problematisations form?

Despite the richness of recent philosophical reflections, few, including the works mentioned previously, have spoken directly to these processes of problem formation. This observation is not intended as a critique, however, as their contributions are valuable in their own right, and open the door for further interrogation of the phenomenological links between individuals, society, and waste. As Barnett and Bridge (2016) note, the formation of problems is, of course, a longstanding concern within the social sciences. Emphasis has historically been on matching problems with policy, and notable examples would include Bacchi's (2009, 2012, 2016) methodological approaches for analysing how problems are represented in policies and Turnbull's (2006) critique of public policy and problem solving. These pieces however, focus on problem formation and problem soliving (e.g. through public policy approaches) at a societal level, not at the level of the individual going through their quotidian existence. Likewise, regarding waste, Gregson and Crang (2010) argue that much of the work that does address problem formation does so at the level of the categorical (see (Davoudi, 2000, 2009; Hillier, 2009; Petts & Niemeyer, 2004). Rather than opening out into its ontological politics, waste is problematised in ways that can be neatly categorised within policy frameworks rather than through messy depictions of social construction (Gregson & Crang, 2010). These contributions are, of course, deeply contextualised within the writings and social-constructionist traditions of Michel Foucault, specifically his notion of 'problematisation' as a means for understanding processes of problem formation (Barnett & Bridge, 2016). To Foucault, problematisation is the process through which inert, apolitical, and fixed objects (such as waste) transform into sets of fluid, conditional, political relations (Foucault, 1984). According to Oberg (2019) the power of Foucault's view on problem formation, or 'problematisations', is its characterisation of them as results of particular social relations, contingent on human practice, rather than inevitabilities. Until recently, Foucault's notion of problematisation had primarily been utilised as a means for critique within urban studies, at a societal level. However, recent contributions by Bacchi (2015), Barnett and Bridge (2016), and Oberg (2019) have demonstrated how the concept can be utilised as an analytical tool to understand processes of problem formation that occur within the individual, and how these processes may be applied in regards to waste.

Reflecting on the importance of approaching the concepts of urban inquiry problematically, Barnett and Bridge (2016) press the modern relevance of Foucault's conceptions of problematisations as part of thinking problematically about urban issues. Foucault is best known for his theories on power, and specifically, state power. As Barnett and Bridge (2016) describe, Foucault's ideas have often served as frames of analysis for the ways in which we engage critically with the city. However, to Barnett and Bridge (2016), Foucault is not merely a theorist of critique, as he has been most often characterised, but also a theorist of action. This emphasis translates into their interpretation of problematisation, which they, echoing Foucault's (1984) own characterisations, place on action as a 'responsive disposition to difficult situations' (Barnett & Bridge, 2016, p. 1189). Moreover, as Barnett and Bridge (2016) note, Foucault, during his own reflections on the subject, was insistent that problematisations are not simply 'an arrangement of representations', rather, scrutinising problematisations involves two paths by "which one tries to see how the different solutions to a problem have been constructed; but also how these solutions result from a specific form of problematisation" (Foucault, 1984, pp. 389-390). Their view is more democratic than Foucault's however, as Barnett and Bridge (2016) start from a presumption that the act of problematisation is a more broadly lived reality of urban life. To them, it is not a refined academic skill, but a basic feature of engaged action and conscious decision-making across any number of issues or lifestyles (Barnett & Bridge, 2016, p. 1201).

Helpfully, Barnett and Bridge (2016) distinguish between two aspects of use for Foucault's notion of problematisation commonly utilised within the social sciences: its forms as both a noun and a verb. First, problematisation, the noun, characterises the problematisation as an object of analysis. As Barnett and Bridge (2016) describe, it is the label attached to the process by which, for example, various lifestyles, livelihoods, or spatial arrangements are questioned and become the target for motivated adjustments and transformations. The second aspect, problematisation, or problematise, the verb, refers instead to a method of analysis, most commonly utilised by researchers within urban studies. Within this sense of the term, according to Barnett and Bridge's interpretation (2016, p. 1191), the purpose of critical analysis is to question understandings, relationships, and settlements that were previously taken-for-granted. Bacchi (2015) draws a clearer distinction between the usages. To Bacchi (2015), 'problematise' (the verb) tends to be used to describe what individuals or governments do in the face of problems, while the noun (i.e. problematisations), generally refers to the outcomes of problematisations, either in the way in which problems are framed, or governmental problematizing processes. Bacchi (2015) further clarifies that the verb form can be used in two ways. First, to describe a form of critical analysis, such as described by Barnett and Bridge (2016), and commonly utilised within the social sciences when questioning or interrogating a specific issue (Bacchi, 2015, 2016). The second form, however, is more pertinent to the objectives of this study: referring to the ways in which individuals put an issue, object, etc. forward, or designate something, as problematic—"that, is to give a shape to something as a 'problem'" (Bacchi, 2015, p. 3).

Within Barnet and Bridge's (2016) reading of Foucault, problematisations primarily occur at the societal level: inside, in concert with, or in opposition to the state. Bacchi (2015), likewise, considers how governing involves problematizing, shaping issues as 'problems'. However, Bacchi (2015) also extends these processes of problem formation to the individual—how people (policy makers, social scientists, but also regular citizens) internally and phemonologically examine, consider, interpret, and then 'frame' an issue as potentially problematic. Neither speak explicitly to the ways in which individuals might problematise specific objects, or groups of objects, such as waste. Oberg (2019), however, broadens the utility of the term, shifting the scope of analysis from problematisations around a given issue, to understanding how individuals problematise certain experiences, in this instance open defecation in urban India. Her analysis found that nearly all actors, including a broader range of stakeholders within Agra, problematise open defecation for various reasons, viewing it as inconvenient, dangerous, a source of disease, undignified, polluting, and/or primitive. However, some problematisations, such as its inconvenience, were limited to only certain actors, while others emphasise problematisations, such as the risk of disease, that others do not consider as a problem (Oberg, 2019). To Oberg (2019), the value in using the concept of problematisation as an analytical tool is that it allowed for the full range of defecatory experiences in a given context to emerge. As she notes, there is value in simply recognising the existence of all problematisations. Accounting for the full spectrum of experiences "gives voice to subordinated problematisations and the groups enacting them, answering the normative imperative to be more inclusive" (Oberg, 2019, p. 389). A focus on problematisations allows for hidden factors to emerge within decision making and problem formation processes, while considering respondents as rational decision-makers. Most importantly, to Oberg (2019), exploring the full range of problematisations counters dominant problematisations by presenting other, more inclusive, alternatives. In an attempt to build on Oberg, Bacchi, and Barnett and Bridge's broader conceptualisations of Foucault's problematisations as an analytical lens through which to interpret process of problem formation, the purpose of this work is to examine how individuals problematise waste objects within their daily lives. Specifically, we seek to understand, how two groups, who live and work within or around the boundaries of Mzedi Dump Site, in Blantyre, Malawi, problematise the often hazardous waste fractions they encounter daily, and how these problematisations are shaped through their interactions with each other and the broader community, beyond the dump.

4. MZEDI DUMPSITE

This study draws on extensive qualitative, participatory fieldwork in and around Mzedi Dump Site, Blantyre, Malawi's only municipal-run waste management facility. A space of diverse livelihoods and hazards, Mzedi represents both opportunity and hardship for the more than 100 informal waste pickers (IWPs) who work there, as well as for the scattered communities existing on its margins. Cut like a gash from the rolling peri-urban patchwork of homes, maize fields, and shops which hug the main M3 Highway to Zomba, Mzedi is located approximately 12 kilometres northeast of central Blantyre, just beyond the fringes of the sprawling Kachere slum settlement, and symbolically dividing the urban sprawl of the city from the agricultural landscapes of rural Malawi. The site dates back to the mid-1990s, when the Blantyre City Council (BCC) [5], purchased the land on which Mzedi now sits from the Imperial Tobacco Group (ITG), and through minimal excavation transformed a shallow valley into a dump site. Nonetheless, as Kalina et al. (2019), describe, land tenure around Mzedi remains complicated. Although the BCC owns the majority of the land surrounding Mzedi, villagers from nearby Moto Village (Figure 1) have historically maintained the de facto right to use the space for agricultural purposes—a right the city has not actively contested [6]. However, the city has always maintained that this arrangement is temporary, and could be altered, should Mzedi be expanded or developed. As Kalina et al. (2019) describe, the attitude is rooted in the argument that the dumpsite pre-dated the intensive cultivation of the land, a narrative contested by local villagers, who consider the dump site an imposition on agricultural land traditionally considered theirs. This historical connection to the space is amplified by acute land pressure in the area—agricultural plots in Malawi are generational, and Mzedi is set within plots that, according to residents of Moto Village, have been allocated to families for generations. As a result, planting elsewhere is not an option, as alternative land is either not available, or is too distant to be readily accessible (Kalina et al., 2019). Consequently, Mzedi, despite being a dump, remains the site of intensive seasonal agricultural production, with many residents of Moto Village maintaining their traditional plots within and along the blurred edges of the site (Figure 2), to plant maize, as well as an assortment of other crops such as melons, tomatoes, and gourds. During the height of the growing season the site is lost in a sea of maize, as every piece of adjacent, farmable land is utilised.

The dumpsite itself has been minimally engineered. Being set in a natural valley, little excavation was required, and the bottom was not lined prior to commissioning. Construction has been limited to an earthen berm, erected along the southeastern edge of the site, and intended to protect a nearby seasonal watercourse, which passes through Moto Village, from leaching (Figure 1). This berm has evidently proven ineffective, however, as surface water tests, taken from the stream at points adjacent to Moto Village revealed severe biological and physical contamination, as well as unsafe levels of nitrates, copper (Cu), and lead (Pb)[7,8] (SOFET, 2016). Opened with an expected operational life of twenty years (in the mid-1990s), Mzedi is now well past its intended lifespan, and has deteriorated considerably in recent years. The site, once forested, is now exposed, with no fixed boundaries separating dumping grounds from adjacent agricultural land. Furthermore, the access road, once

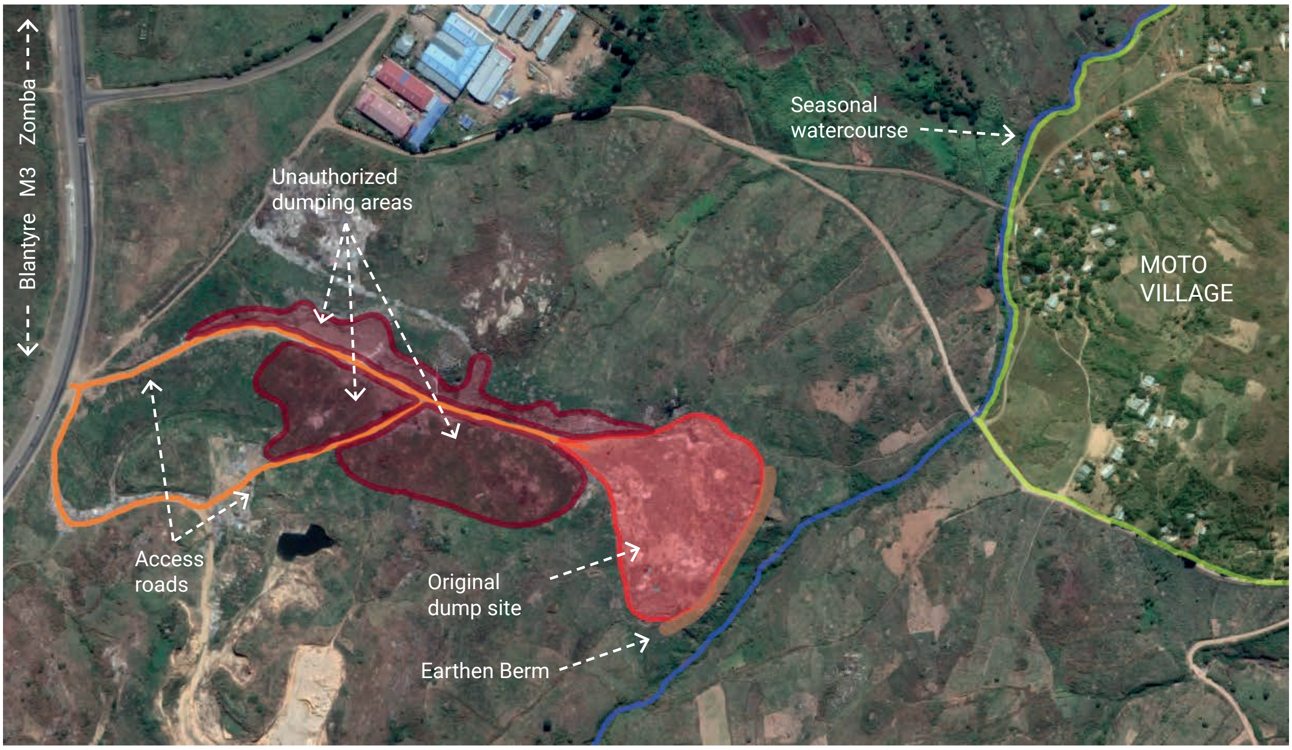

FIGURE 1: Mzedi Dumpsite and Moto Village.

paved, has become deeply rutted, and is often impassable, especially during the rainy season. As a result, unauthorised dumping has proliferated along the margins along the main site, with the access road itself becoming a secondary dumping ground, as trucks illegally unload along it when the main site becomes inaccessible (Figure 2). Unfortunately, this deterioration is expected to continue, as the city lacks both the funds and the heavy equipment to clear the road or conduct necessary upgrades to the site. Looking forward, city officials pin their hopes on the construction of a new

FIGURE 2: Primary access road to Mzedi dumpsite; maize and unauthorized trash dumping are visible along the sides (Kalina, 2019).

site that will improve upon many of Mzedi's failings, however, such an intervention is unlikely to materialise, as waste management services within Blantyre are conducted under extreme financial limitations, necessitating Mzedi's continued use (Kalina et al., 2019).

Prior to Mzedi's construction, dumping within Blantyre was neither formalised nor centralised, with the city utilising a variety of scattered, local, and temporary dumping grounds, typically disused quarries or abandoned construction sites, on an ad-hoc basis when necessary. Nonetheless, despite the BCC centralising waste management disposal at Mzedi, the site still only contains a fraction of the waste produced within Blantyre, as waste collection services remain extremely limited. According to Ndau and Tilley (2018), the City of Blantyre only offers household waste collection within a limited number of 'formal' neighbourhoods, servicing only about 20% of the population. For the rest, the BCC maintains 43 skips (of around $7m^3$) strategically placed at public spaces throughout the city (such as markets, hospitals, etc.), which are rotationally collected and dumped at Mzedi. Furthermore, individuals and private businesses may also purchase permits to dump, and most of Blantyre's manufacturing industries have purchased the right to dump at the site. Unfortunately, the character of the waste currently stored within Mzedi is unknown. The city maintains little control over the types of waste being dumped, as the permit system is predominantly concerned with regulating the quantity, not the quality of deposited waste. The BCC has placed restrictions around the dumping of certain hazardous wastes, such as medical waste, however compliance with these restrictions is largely voluntary, and on-site inspection of deliveries is cursory or absent. Furthermore, unauthorized or unpermitted dumping is common, while hazardous waste is often mixed within general waste [9]. Consequently, even the BCC has a poor understanding of the amount of hazardous waste that may be present at Mzedi, and sightings of hazardous waste, such as chemicals or batteries, and medical waste, including medicines, needles, and even body parts, are common [10].

Despite these hazards, more than 100 informal waste pickers (IWPs) work within Mzedi, gathering specific waste items to resell or reuse within their own households. Both the maize growers and IWPs are interrelated, as many IWPs live in Moto Village and also plant maize within and around Mzedi. The IWPs have suffered from the physical deterioration of the site, with the deforestation depriving them of shade and firewood, while the degradation of the access roads, and the resultant unauthorised dumping, has decentralised waste storage beyond the original dump site (Figure 1), making it more difficult for IWPs to quickly access and pick new deliveries (Figure 3) [11]. Moreover, other infrastructure, such as signage and running water, which the IWPs used to benefit from, is now missing or inoperable [12]. Nonetheless, the BCC maintains an uneasy, but generally indifferent relationship with the IWPs, embodied within the person and function of the dump supervisor, who is primarily responsible for ensuring compliance with the permit system, but also plays an important mediation role between the city and those individuals working and living around Mzedi.

5. METHODOLOGY

This study was conducted in tandem with Kalina et al. (2019), which sought to understand both the real and per-

FIGURE 3: IWPs Access Waste Dumped Alongside the Access Road (Kalina, 2019).

ceived dangers of agricultural production at Mzedi through interviewers with growers as well as laboratory testing for heavy metals within purposively sampled maize, soil, and water samples. Results suggested that agricultural production at Mzedi is reasonably safe, with no samples showing hazardous concentrations of potential toxins (Kalina et al., 2019). Nonetheless, the environmental hazards at the site, such as broken glass, medical waste, and potentially toxic wind blow dust, renders the work risky. Kalina et al. (2019) hint at the quotidian and complex processes of problem formation around waste undertaken by maize growers navigating Mzedi, but without the depth of analysis and theorisation that this article contributes. In contrast to Kalina et al. (2019), which utilised a mixed-methods approach, this study was purely qualitative and interpretative. Data collection consisted of 26 semi-structured interviews with relevant stakeholders associated with Mzedi, including officials from the BCC [13], residents of Moto Village, maize growers, and IWPs. Respondents were chosen using both a purposive and snowballing sampling regimen (Kitchin & Tate, 2000). Interviews [14] were conducted in the local language (Chichewa), with a translator, audio recorded, and transcribed into English. Participation was voluntary, and responses were recorded anonymously [15]. Transcripts were coded and organised using Nvivo, and the analysis was conducted thematically(Denzin et al., 2017).

6. FERTILE MZEDI: GLASS, SYRINGES, SMOKE, AND MAIZE

In Malawi, especially for the poor, summer is characterised by what is known colloquially as 'the hungry months', the gap in between the sowing and reaping of the annual maize harvest, during which long-hoarded maize stores dwindle, food costs spike, and malnutrition rises (Kalina et al., 2019). For subsistence agriculturalists planting at Mzedi, work begins in early November, with a rush to clear the land, till, and plant, before the first of the summer rains arrive. At Mzedi, however, this work is not just considered difficult, but hazardous, as the growers reported frequently encountering sharp objects, primarily shards of metal, nails, and broken glass when clearing the soil—work that is primarily done by hand. These sharp objects were clearly characterised as problems by growers: environmental hazards and occupational risks that needed to be navigated on a daily basis. Indeed, nearly every grower spoke of past injuries and cuts incurred while clearing the soil prior to planting, particularly to the hands and feet, and a cursory glance at the hands of each respondent revealed numerous and often substantial scars (Figure 4) [16]. Moreover, because many growers are also handling waste on a daily basis, and WASH facilities at Mzedi and in Moto Village are basic, wounds often heal poorly or become infected. Despite identifying broken glass as a persistent problem in their plots, growers generally make no effort to remove it, simply shifting it aside when preparing the soil for planting or during harvesting. These objects are clearly problematised by growers, however, most respondents implied in their comments that at Mzedi the glass was nearly as plentiful as the soil, and have essentially accepted it as one of inevitabilities of cultivating the space. Finally, although less common than the ubiquitous glass at the site, one grower reported encountering medical waste, including used syringes and other sharps when preparing the soil within his traditional plot [17]. These items, however, although identified as a problem by the grower, were not problematised

FIGURE 4: An IWP shows a scar caused by broken glass at Mzedi (Kalina, 2019).

in relation to their potential infection risks, which were not mentioned by respondents, but in their ability to cause bodily harm.

One environmental hazard that has been clearly problematised by maize growers, on the basis of its potential health risk, is the persistent cloud of dark smoke which hangs over the site. Caused by the many small fires started by IWPs for cooking, heat, or to burn waste to extract recyclable metals, the smoke caused by these fires is particularly acute during the dry season (May-October), and they can often spread and burn uncontrolled, smouldering for weeks until a rain or until it runs out of fuel. When speaking about the smoke, growers were unable to point to a specific instance in when it made it them ill, however there was a general awareness that the air at the site was polluted and unhealthy. Many reported struggling to breathe and coughing when inhaling the smoke while in the vicinity of the site, while one grower who had been diagnosed with asthma, blamed the smoke for causing and frequently exacerbating his condition [18].

In Malawi, April is the month of harvest, and at this time, in Moto Village, the daylight hours are predominantly occupied with the labours needed to harvest, process, and store the annual maize harvest [19]. Kalina et al. (2019) describe these daily rhythms in detail. Once ripe, the maize is not harvested immediately, but rather is left on the stalk in the field for several weeks in order to partially dry. After picking, the ears are husked and the kernels are stripped from the cob [20]. Bare cobs and husks are dried and burnt for household fires, while the kernels are spread on mats to dry in the sun. Once dried, seed for next year's crop is selected and saved, while the remainder is stored, crushed into maize meal on demand, and consumed throughout the year. Those growing maize in the vicinity of Mzedi dump site largely do so for subsistence purposes, consuming the bulk of what they grow within their own households. However, in good years, some reported small surpluses that they were able to sell [21].

Amongst growers, there was little concern that they maize they were growing came from a dump site, and few problematised the potentially hazardous impacts [22] that the waste in the site could have on their crops, and consequently, their health. One explanation for this is that, according to the majority of growers, the maize that is produced on the margins of Mzedi, and once harvested, is virtually indistinguishable, in terms of size, taste, and colour, from maize grown elsewhere, including what is available in local markets and from the Agricultural Development Marketing Corporation [23] (ADMARC). For the small surpluses that can be sold, kernels from Mzedi are mixed with those from other locations, and respondents reported that their customers either were not aware that the maize they were purchasing came from the dump site or were unable to distinguish it from the other source [24]. One respondent, however, did note that their customers knew that the maize they were buying was grown near the dumpsite, but it did not discourage them from buying, because the product is indistinguishable from other maize [25]. Of the growers interviewed, only two expressed some concern that the waste stored at Mzedi may have a potentially negative impact on the maize grown there. Both expressed a belief that maize grown at the dumpsite had a tendency to rot before it is harvested, while left on the stalk to dry [26]. Moreover, one of the growers described the maize kernels from the dumpsite as being slightly darker in colour and having a less pleasant taste than maize grown closer to the village [27]. Both believed the waste stored at the dumpsite was responsible for these impacts, particularly the rotting, however they were unable to explain what types of waste may be potentially harmful, and were unable to describe what processes or waste types may be responsible for negatively impacting the maize.

To those who cultivate maize on its margins (and to a lesser extent, within) Mzedi is clearly a hazardous space (Figure 5). They have problematised its visible, environmental dangers, such as broken glass and other sharp objects, medical waste, and the persistent pall of noxious smoke, because they are tangible dangers, which have negatively affected their safety and health. For the majority of respondents, problem formation has not occurred around the dump's hidden dangers, specifically, the potential health risks of consuming maize from contaminated soil, because the threat is not visible (potentially harmful concentrations of arsenic, for instance, would be tasteless) or because growers are not aware that potentially negative health impacts are possible. Quite simply, they do not consider dumpsite-grown maize as a problem because it has not yet tangibly affected them, and when broached, the immediacies of survival and food security overshadow such nebulous concerns. However, although most growers have not problematised the relationship between the waste in Mzedi and their maize crop, it is not because they are unable to conceptualise that the former may affect the latter. Rather, and despite its hazards, the plots adjacent to Mzedi are considered attractive places to plant because they are seen as particularly fertile, a fertility that most growers attributed to the waste [28]. However, nobody was able to articulate what processes were responsible for positively affecting the soil or describe which types of waste may have beneficial impacts. Nonetheless, there is some awareness amongst growers that the waste stored at Mzedi may be influencing their maize in some way.

7. 'WASTE', 'TRASH', PROBLEMS, AND OPPORTUNITY

In addition to being a site of agricultural production, Mzedi also supports the livelihood of more than 100 informal waste pickers (IWPs). Many have worked at the site since its opening, live in nearby Moto Village, and have witnessed the steady physical deterioration of the site. Although the official relationship between the IWPs and the city is antagonistic—the BCC Director of the Department of Health and Social Services described the two parties as 'enemies'—on the ground there is less tension, with a mutual toleration between city workers and IWPs, borne out of shared hardship and poor working conditions. For these IWPs, problem formation in regard to waste and Mzedi, has, like with the maize growers, largely centred on the tangible environmental health and safety risks that are a consequence of navigating a dumpsite. Likewise, broken glass was frequently

FIGURE 5: Maize plant growing near the centre of the original dumping area (Kalina, 2019).

cited by IWPs as the most persistent environmental hazard encountered at the site, more so because they are forced to frequently handle or sift through waste without the use of gloves or other personal protective equipment. Most IWPs, displaying scarred hands and feet, described injuries from glass as frequent occurrences that often required trips to the hospital or necessitated them missing periods of work to allow for healing (Figure 4). Another waste item around which problematisations have formed is old food, with some IWPs believing that consuming food waste found at Mzedi could cause illness [29]. However, this problematisation was not universally held, as many reported happily taking old food, either to eat at home or to feed to livestock. Similar problematisations have formed around medical waste, though few IWPs described encountering the needles and other sharps objects described by the maize growers, instead describing uncovering medicines, pills, and other chemicals, which they choose to avoid for the fear that they are poisonous or could otherwise cause sickness [30]. Finally, although not a waste item, the tractors, skip carriers, and dump trucks, either municipal or private, that are responsible for making the daily waste deliveries represented the most serious and visible danger to life and limb for Mzedi's IWPs. This danger stems from the tendency of some IWPs to wait near the entrance of the site for new deliveries, and when one arrives, climb on and ride on the back of the skip to its dumping point in order to have first pick of the waste (Figure 6). This behaviour has had unfortunate consequences however, as a number of IWPs have fallen from vehicles along the extremely rutted and uneven access road. According to the Dump Supervisor [31], these falls most often result in a broken arm or leg, but for

FIGURE 6: IWPs ride a new delivery into Mzedi (Kalina, 2019).

a few who have fallen under the wheels of the tractor, the result has been fatal. Although both sides regret the loss of life, there is a clear distinction in opinion in where the blame lies, with the city problematising the behaviour of the waste pickers as unsafe and reckless, while the IWPs criticise the city and the tractor drivers for not considering their safety or their livelihood needs [32].

Although these dangers shape the ways in which IWPs problematise waste at Mzedi, the single largest factor influencing processes of problem formation within these individuals is their livelihoods—recovering value from what society has discarded. IWPs at Mzedi collect a broad range of materials, with most individuals specialising in a few specific waste streams, while a few prefer to collect more broadly or opportunistically. Commonly collected waste items include: recyclable metals, such as aluminium and copper, which are gathered and resold to scrap dealers [33]; plastic drink bottles which are either reused at a household level or sold to individuals who use them for storing resalable portions of cooking oil or other liquids [34]; scrap paper and wood which is sold as fuel [35]; plastic bags and other scrap plastics which are sold in bulk to commercial buyers or at a smaller scale for use as fire starters [36]; paint cans which are sold for the scrap metal content as well as for any leftover paint [37]; food waste which is used domestically as pig feed [38]; and glass: Carlsberg Beer or Coca-Cola bottles which carry deposits and are collected to be returned [39]. Although the range of items collected are diverse, the motivations driving each individual IWP's collections are consistent: they collect what they are able to sell. If there is a market for it, there is an IWP at Mzedi collecting it. There are a few waste items, however, that IWPs universally avoid; specific examples include most glass waste, chemicals, and some organic waste such as grass and sugar cane chaff. These items are not avoided because they are dangerous, though in some instances they certainly can be, but because, to the IWPs they are worthless. Carlsberg or Coca-Cola bottles can, and do, cause cuts just the same as an empty Heineken bottle or a broken windowpane, however they are not problematised in the same way as these other glass items are because they carry deposits and thus have value to the IWP. Likewise, the recovery of scrap metal can also often be hazardous and the IWPs suffer just as, if not more severely from the fires needed to recover the metals as the local maize growers. However, these items are also not problematised by IWPs at Mzedi in the same way as other environmental hazards because they are among the highest value recyclable items; the kind of waste they would like to see more of in Mzedi, certainly not as a problem that needs to be addressed.

Stemming from this dynamic of only problematising items with no resale value, IWPs had a tendency to speak in terms of a dichotomy of 'good' waste vs 'bad' waste. Akin to Kennedy's (2007) distinction between 'waste' and 'trash', 'good' waste represents items that have value, something that can be sold to support a livelihood, while 'bad' waste was worthless, had no value, and occupied time and space which could be spent more productively. This concept of waste being 'bad' was the principle factor shaping processes of problem formation for IWPs within Mzedi, and although safety considerations were considered within individual problematisations, value and livelihoods remained the overriding concern. Finally, this dichotomy ultimately influenced the way in which they problematise Mzedi itself. Although individuals conveyed concern about the physical deterioration of the site, these concerns were ancillary to a growing expression of alarm that the quality and quantity of recyclable materials reaching the site has decreased in recent years, as the quote at the start of this article indicated. As such, a number of respondents were left questioning the sustainability of their already precarious livelihoods, but, nonetheless, seem resigned to continue a life of quotidian hardships and problems in Mzedi.

8. CONCLUSIONS

The purpose of this article has been to explore how and why certain 'waste' items are and become understood as problems. It adopts Foucault's (1984) notion of 'problematisation' as an analytical lens for conceptualising processes of problem formation, through the eyes of two different groups scraping together livelihoods within and on the margins of Mzedi Dump Site: subsistence maize growers and informal waste pickers. The example of Mzedi, and the challenging lives of the individuals who live and work there, illustrate that problems do not exist independently from the people they affect, rather they are continually made and remade through human practice.

Of the countless pieces of detritus at Mzedi, the refuse of the small minority in Blantyre who have access to waste management services, what 'waste' objects or other environmental hazards have these individuals come to understand as problems, and how and why have those problematisations formed? For both maize growers and IWPs, problematisations are dynamic, driven by daily routine and personal hardship. Problematisations are shaped by the tangible: the visible, and often painful impacts that Mzedi's hazards have on their lives and livelihoods. For instance, both groups have clearly characterised glass and other sharp pieces of waste as problems, because these objects can harm them, and they have evidence: the poorly healed cuts and thick scars. Likewise, maize growers have problematised the persistent smoke that hangs over the site on the basis that it makes it hard to breath, causes coughing, and exacerbates illnesses. These are real hardships that they struggle with on a daily basis, which have crystalised their internalised processes of problem formation. Conversely, other 'waste' hazards such as the potential health risks associated with consuming dumpsite-grown maize have not been problematised because they are more intangible. As we have argued, this is not because growers cannot conceptualised linkages between 'waste' and their maize, but rather because the problem is distant, less visible, and more nebulous, and has not yet manifested in a way that has been seen or understood as a problem by growers. Finally, at Mzedi, the need to earn a living, and the consequences of falling short, is the most tangible reality of daily existence, and this dynamic has in turn shaped how individuals, and the IWPs in particular, problematise the hazards posed by the various waste items found there. 'Good' waste, is internalised based on its value. No matter how sharp or toxic the object may be, it is not problematised, rather it is desired and fought over because someone will buy it. It is not a problem, it is an opportunity. 'Bad' waste however, is not problematised because it is a hazard (it may be dangerous or innocuous), it is problematised because to the IWP it is has no value. Without a market or a willing buyer the item is considered useless, Kennedy's (2007) 'trash'; a problem taking up time and space which could be filled more profitably.

To Barnett and Bridge (2016), the value of Foucault's conceptualisation of problematisations lies in its ability to inform situational analysis of the formation of problems. Problematisations emerge from uncertainty or, occasionally, from distress or hardship. For Foucault (2001, p. 172) the problematisation is a response or 'answer' to something that is real: a concrete situation or a lived reality. Those working in and on the margins of Mzedi have problematised its waste for the tangible impacts it has on their lives. However, once these problematisations have formed, how have they affected how individuals navigate and react to the problems that they have identified? Unfortunately, in regards to Mzedi, clear and universal problematisations of certain waste objects, like broken glass, have not influenced behaviour or contributed to any collective action for change, as most individuals simply struggle to survive, and lack the agency and resources to pursue solutions. Nonetheless as Oberg (2019) reminds us, there remains value in bringing to light the full range of problematisations. Considering the full range of problematisations gives voice to previously disregarded problematisations, as well as the potentially marginalised groups forming them (Oberg, 2019). If the BCC does develop Mzedi in the future, the problematisations of these groups should be considered and acted upon, rather than being subordinated to the dominant problematisations championed by the City or privileged groups.

But once an object has been identified as a problem, what then? Many individuals would describe litter as a 'problem', but are generally happy to step over it on a busy city street, rather than take the time, to engage with it. Yet, at what point does 'trash' become something more than a nuisance that can be ignored, transforming into a problem that must be dealt with or actively avoided? How do these understandings of problems inform choice and individual processes of decision-making, particularly within the African city, many of which have lurched awkwardly into modernity, yet remain spaces of tremendous opportunity and imagination? These questions remain fertile space for further theoretical and empirical investigation.

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[1] Informal Waste Picker #17, 24/04/2019.
[2] Throughout our own analysis we try to maintain Kennedy's distinction between 'waste' and 'trash', though often the distinction does not translate neatly from the original Chichewa, so we occasionally resort to interpreting the intent of the respondents.
[3] Serving as an online hub for scholars, activists, environmentalists, and other communities engaged in waste, Discard Studies provides a platform for critical and informed discussion around the relationships between waste and society, contextualised within broader sociocultural-economic analysis. Their monthly compilation 'The Dirt' seeks to assemble recent articles, job postings, and calls for participation relevant within the discipline.
[4] The purpose has not been to provide a complete survey, rather to highlight pieces that have been particularly useful to the author during the conceptualisation of this study.
[5] Mzedi is owned by the Blantyre City Council (BCC) through the Department of Planning and Estate Management Services, while the site is operated by the Department of Health and Social Services
[6] Dr. Kanjunjunju (04/23/2019).
[7] The Malawi Bureau of Standards specifications for safe drinking water require a maximum Pb concentration of 50 μg/L. According to SOFET (2016), ground water testing in Moto Village revealed Pb concentrations exceeding 715 μg/L. Unfortunately, potentially harmful metals such as As, Co, Fe, Ni, or Zn were not analysed.
[8] This could partially be due to lack of maintenance and the resultant physical deterioration of the berm over the past decade. Across its breadth the berm has severely eroded, failing in a number of spots, spilling solid waste in the water below.
[9] Dump Supervisor (04/24/2019).
[10] Dr. Kanjunjunju (04/23/2019).
[11] Dump Supervisor (04/24/2019),
[12] Dump Supervisor (04/24/2019),
[13] Representatives from the City induced the director of the Department of Health and Social Services and the manager of Mzedi Dumpsite.
[14] Excluding those with City officials, which were conducted in English.
[15] The study was approved by the National Committee on Research in the Social Sciences and Humanities (NCRSH) of Malawi; Protocol NO. P.03/19/356.
[16] Translator (27/05/2019).

[17] Maize Grower #6, (04/24/2019).
[18] Maize Grower #1 (04/24/2019), Translator (27/05/2019).
[19] Many of the interviews conducted with growers for this study were done while the respondent was processing their maize harvest within their homes.
[20] Alternatively, some households dry the maize on the cob, store it, and strip it prior to use.
[21] Maize Grower #2 (04/24/2019), Maize Grower #4 (04/24/2019), Maize Grower #5 (04/24/2019), Maize Grower #6, (04/24/2019).
[22] According to Kalina et al. (2019), the principle danger in consuming maize from contaminated spaces, like a dumpsite, would be its ability to bio-accumulate potentially harmful heavy metals, such as arsenic (As) and copper (Cu). The consumption of potentially toxic quantities of As poses the greatest, and most likely, risk given the context. At a biochemical level, As (arsenic) acts to coagulate proteins, inhibiting the production of adenosin triphosphate (ATP), which is essential in metabolic processes (Okoronkwo et al., 2005). Furthermore, although concentrations may remain low, the potential for bioaccumulation, and the ability for soil contamination to increase over time, suggests that the risk of exposure to heavy metals due to plant uptake renders the use of contaminated soils, such as those present at dumpsites, risky spaces for agricultural production (see (Amusan et al., 2005; Li et al., 2017; Okoronkwo et al., 2005; Opaluwa et al., 2012; Pastor & Hernández, 2012; Sang et al., 2010)).
[23] A parastatal entity, which buys and sells maize in order to regulate prices and relieve shortages.
[24] Maize Grower #1 (04/24/2019), Maize Grower #4 (04/24/2019), Maize Grower #5, (04/24/2019).
[25] Maize Grower #2 (04/24/2019).
[26] Maize Grower #4 (04/24/2019), Maize Grower #6 (04/24/2019).
[27] Maize Grower #4 (04/24/2019).
[28] According to Kalina et al. (2019), at Mzedi, many who used fertilizers sporadically reported not being able to differentiate between fertilised and non-fertilised plots, while those who could not afford fertiliser at all still described positive harvests.
[29] Informal Waster Picker #10 (04/24/2019), Informal Waster Picker #12 (04/24/2019).
[30] Informal Waster Picker #2 (04/24/2019), Informal Waster Picker #6 (04/24/2019), Informal Waster Picker #15 (04/24/2019), Informal Waster Picker #17 (04/24/2019).
[31]Dump Supervisor (04/24/2019),
[32] Dr. Kanjunjunju (04/23/2019), Dump Supervisor (04/24/2019),
[33] Informal Waster Picker #1 (04/24/2019), Informal Waster Picker #5 (04/24/2019), Informal Waster Picker #7 (04/24/2019), Informal Waster Picker #9 (04/24/2019), Informal Waster Picker #16 (04/24/2019).
[34] Informal Waster Picker #10 (04/24/2019), Informal Waster Picker #12 (04/24/2019), Informal Waster Picker #13 (04/24/2019).
[35] Informal Waster Picker #2 (04/24/2019), Informal Waster Picker #3 (04/24/2019), Informal Waster Picker #11 (04/24/2019).
[36] Informal Waster Picker #3 (04/24/2019), Informal Waster Picker #8 (04/24/2019), Informal Waster Picker #11 (04/24/2019).
[37] Informal Waster Picker #17 (04/24/2019).
[38] Informal Waster Picker #6 (04/24/2019).
[39] Informal Waster Picker #5 (04/24/2019), Informal Waster Picker #18 (04/24/2019).

Extra contents

COLUMNS AND SPECIAL CONTENTS

This section comprises columns and special contents not subjected to peer-review

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CISA

ENVIRONMENTAL FORENSICS

ENVIRONMENTAL CRIME SCENE ANALYSIS

Introduction

Although environmental crimes include several typologies (illegal emission or discharge of substances into the environment, illegal trade in wildlife, illegal trade in ozone-depleting substances, illegal transport, shipment or dumping of waste, etc.), the most common is the illegal discharge of pollutants to the environment. Because of this, the most frequent task of an environmental forensic expert is the investigation of pollution crimes. The Pollution Crime Forensic Investigation Manual (INTERPOL, 2014) identifies the following steps for the successful prosecution of an environmental case: (i) collection of the appropriate evidence, (ii) maintenance of legal continuity of the evidence, (iii) organization and documentation of evidence, and (iv) presentation of the evidence to the various audiences (enforcement office management, police, prosecutors, judges and the court). The first step in the process requires a thorough examination of the polluted site or an environmental crime scene analysis.

Procedures to be followed in Environmental Crime Scene Analysis

In the general sense, a crime scene is where a crime has taken place and where the criminals leave crime traces and evidence within a certain range of time and space (Wang et al., 2018). In this sense, an environmental crime scene is the place where the illegal discharge has happened and from where evidence for convicting the polluter can be collected. The general procedures followed in crime scene analysis, such as (i) securing the crime scene, (ii) preliminary crime scene survey, (iii) crime scene documentation (using notes, sketches, photography, and videography), (iv) crime scene searches, and (v) collection, preservation, inventorying and packaging of physical evidence (Miller, 2003), are relevant and significant for pollution crimes. Underpinning all of these steps is the need for a robust and transparent process that can stand up to scrutiny in court. The Pollution Crime Forensic Investigation Manual of INTERPOL (vol.1&2) explains most of these steps in the context of a pollution crime. The manual is designed to 'assist the investigator through the forensic environmental investigation process from initial receipt of information of a potential violation, through planning and implementation of the evidence gathering process, to preparation and presentation of data and evidence in a prosecution brief'. The first volume of the Manual identifies common environmental investigation scenarios and presents a stepwise approach to gathering evidence.

The INTERPOL Manual has guidelines for investigating many pollution crime scenarios normally encountered (Figure 1, for example). However, there are pollution episodes where identifying the crime scene itself can be challenging. Leaking underground storage tanks or undisclosed chemical dumps polluting the groundwater are classic examples (Morrison, 2000; Varghese et al., 2015). In such cases, the effects of the crime are first observed (in the form of pollutants appearing in some environmental media or as impacts of pollutants on human health or ecosystem), and a careful and detailed investigation may lead the forensic expert to the crime scene. Investigations can become all the more challenging when there is a delay in the manifestation of impacts after the polluting incident has occurred. Identifying and deciding on an appropriate 'pollution signature' relevant to the case can be useful in complex situations (Abdul Samad et al., 2020). The pollution signature is the pollutant/combination of pollutants/attributes of the pollution representative of the case under investigation. Each pollution incident may have one or more pollution signatures associated with it that can lead the investigators to the pollution source.

Challenges specific to Environmental Crime Scene Analysis

There are many challenges that are specific to environmental crime scene analysis. Due to the toxic nature of polluting emissions, sometimes going near the crime scene can be detrimental to the health, and even life, of the forensic expert. Ample precautions, including a well thought out risk assessment, are required when there is suspected release of toxic emissions. Another challenge for the environmental forensic expert is the possible presence of several pollutants at the crime scene, some of them caused by legal activities or natural phenomena. Moreover, these pollutants may be present in the different environmental compartments (air, soil, surface water, groundwater, etc.), that too at concentrations varying spatially and temporally, all making crime scene investigations extremely complex.

Pollution crimes, unlike other crimes, may come to light many days, or even years after the crime is committed, making investigation difficult. One reason for this is that the presence of pollutants in the environmental medium can go unnoticed until its effect is felt on human health or on the ecosystem. Very often, it takes a considerable amount of time for the effects to become manifest at environmental doses (Briggs, 2003). Another issue is the way society perceives environmental crimes. It is not a priority in many societies and not even a concern in some of the societies, severely limiting the resource availability for carrying out rigorous environmental forensic investigations.

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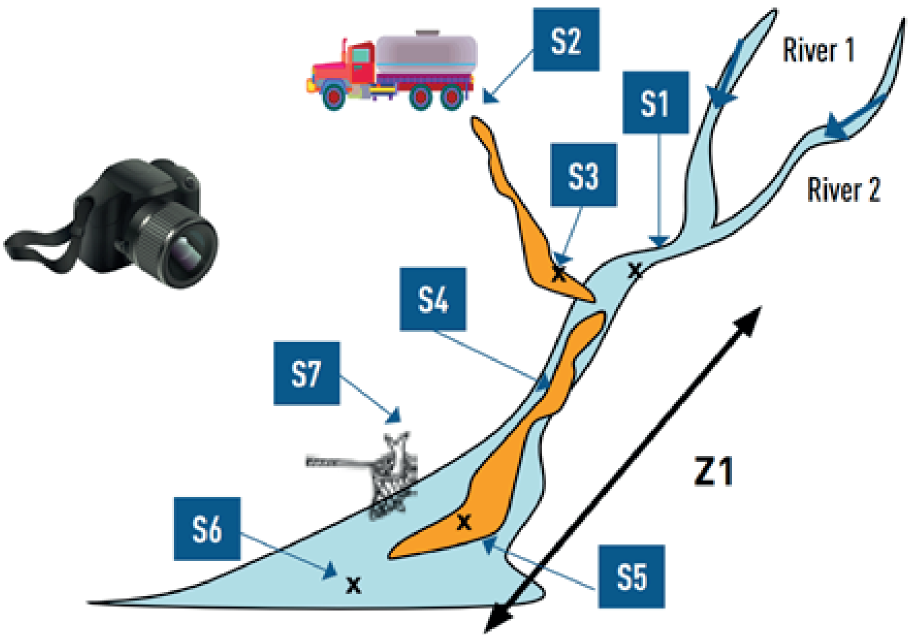

FIGURE 1: Investigative strategy- Improper disposal of sewage through runoff to a water body (INTERPOL, 2014).

Concluding Remarks

Environmental crime scene analysis is the first and often the most critical step in the investigation of environmental crimes, specifically pollution crimes. The general procedures of a normal crime scene analysis can be adopted for the investigation of environmental crime scenes with contextual modifications. The Pollution Crime Forensic Investigation Manual of INTERPOL is a standard reference material for forensic experts for undertaking environmental crime scene analysis. However, there are complex situations where these standard procedures may not fully serve the purpose or are even irrelevant. Improvisation is required to tackle the situation on a case by case basis, which makes the job of an environmental forensic expert challenging.

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DETRITUS & ART / A personal point of view on Environment and Art

by Rainer Stegmann

Artists seldom provide an interpretation of their own work; they leave this to the observer. Each of us will have his/her own individual view of a specific piece of art, seeing different contents and experiencing a range of own feelings and emotions. Bearing this in mind, I created this page where you will find regularly selected masterpieces from different epochs and I express my thoughts on what the work conveys to me personally. My interpretation will refer specifically to the theme "Environment". Any comments or suggestions regarding this column should be addressed to stegmann@tuhh.de.

PABLO PICASSO / Bull´s Head, 1942.

I had the chance to see Picasso´s famous sculpture Bull´s Head in the Picasso Museum of Malaga, Spain. It got my immediate attention -not only because I am a person dealing with waste- but due to its simplicity, expression and beauty.

The Bull´s Head is a typical example of Waste and Art (Stegmann, et.al., 2015)). With materials found by chance or by deliberate scavenging waste is transformed, repurposed, reimagined, given a new life. By these means artists give attention to sustainability and where materials come from. They may have a mission, environmental consciousness or doing it just for pleasure. Using fine arts is in my view an effective "tool" that should be used for education in the widest sense; it is inspiring and initiates creativity. We should see more art in the curricula of Universities.

I guess when Picasso saw the discarded bicycle seat and handlebar he immediately "saw" the bullhead before his eyes. This also has to do with Picasso´s enthusiasm for bullfights and the beauty of bulls. I think often artists have an idea but the final artwork develops by steady inspiration during painting or sculpting; the interpretation of their work lies with the viewer, what he or she feels, thinks about it.

Coming back to Picasso`s Bulls Head: What really amazes me is the simplicity of this installation with this strong expression and aesthetic. Putting together this very simple handlebar and bicycle seat so that everybody immediately recognises a bullhead shows the genius of Picasso. In general I think what makes up a good artist, engineer and other professionals is to find creative solutions in the simplest possible way.

Now, how do I see Picasso´s artwork in relation to the environment? It is obvious that it reminds me of recycling or better up-cycling, use of the value that stays in discarded objects, ironically: in case of Picasso the value probably several Millions of Euros). Picasso often used discarded materials in his collages like newspaper, pieces of wood and cardboard, may be more. The Bull´s Head shows what can be done with objects from waste and it sends the message to the viewer that we should take care of items we want to get rid of, check first if they can be reused or recycled. Another aspect I see is the fun Picasso obviously had putting together seat and handlebar to a bullhead and understand this as a message not loosing the fun in our work even in times of dramatic environmental situation.

Next issue: In the coming issue of DETRITUS I like to present the painting from Rene Magritte "The Lovers II", 1928, Rene Magritte (1898-1967) was a Belgian painter of Surrealism. This art direction wanted to overcome traditional experiences, ways of thinking and viewing by mixing reality with dreams. Magritte called them „Dreams, that do not lull you to sleep but want to wake you up". He created also sculptures and was friend with Andre Breton, Paul Eluard, Joan Miro and later with Savadore Dali.

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(Contents continued from outside back cover)

Columns

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CONTENTS

(Contents continued on inside back cover)

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