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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

ANTARCTICA AS A GLOBAL POLLUTION SENSOR: THE ANTAGPS PROJECT

Pollution knows no borders, it travels the globe and crosses national frontiers in many forms, reaching the most remote areas of the planet. Whilst Antarctica, the only continent that is not permanently anthropized, represents the southernmost part of the planet, this geographic segregation provides no protection against the harmful impact of human activities. This continent therefore, characterized by limited internal sources of pollution but high-burden external routes of contaminants, represents a unique natural laboratory for ecotoxicology studies. The project "Antarctica as a global pollution sensor: aquatic and terrestrial organisms as bio-indicators and meta-analysis of pollutant trends", supported by the Italian National Antarctic Research Program (PNRA), aims to employ Antarctica as a valuable source of information relating to both local and global pollution using its endemic organisms as bio-indicators (Figure 1).

Antarctica, the fifth largest continent after Africa, Asia and the two Americas, is located in the southern hemisphere and features an enormous ice cap covering practically the entire region (59.000 km²), at times exceeding 4.500 m in thickness, and constituting 90% of the fresh water reserve on the planet. The temperature varies from around 0°C during the austral summer on the coast, to approximately -90°C in winter at an altitude of 3000 m in inland areas. Providing an intensely hostile environment for human life, Antarctica constitutes an ideal location for environmental studies as the only region on the planet characterized by an almost total absence of anthropogenic activity and sources of local pollution. Although natural "barriers", such as oceanic and atmospheric circulation, protect Antarctica from lower latitude water and air masses, data relating to the concentration of pollutants detected in several organic and inorganic samples (air, snow, terrestrial and marine organisms) highlight the presence of persistent contaminants derived from other continents in the Antarctic environment (Bargagli, 2008; Roche et al., 2019).

Due to the presence of the Antarctic Polar Front, marine transport of pollutants is modest, whereas long-range atmospheric transport constitutes the main means of their reaching this remote environment. Pollutants such as metals and emerging contaminants, including endocrine-disrupting chemicals, brominated and perfluorinated compounds, pharmaceuticals and personal care products, represent the major pollutants in ecosystems, posing a serious threat to aquatic and terrestrial organisms. Since

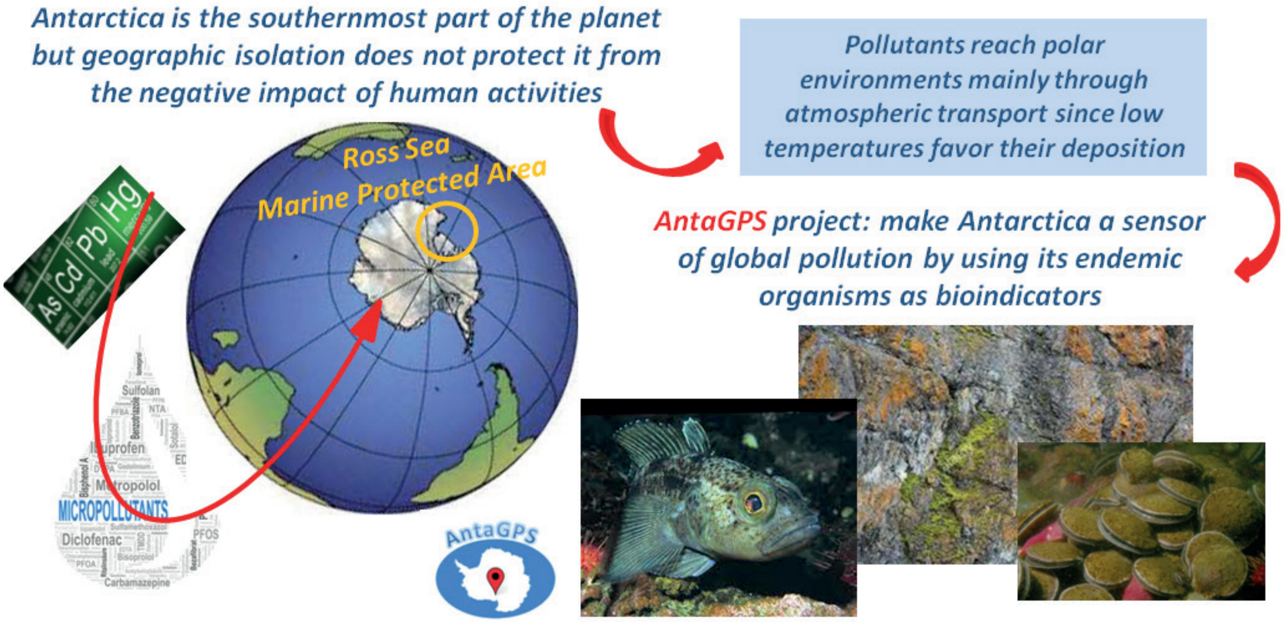

FIGURE 1: Graphical abstract of the AntaGPS project.

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2000, the risks posed by metals have induced numerous countries worldwide to ratify protocols aimed at reducing emissions, while emerging contaminants are currently not included in monitoring programs and their environmental fate and biological impacts are poorly understood. Significant accumulations of metals and emerging contaminants have been identified in Arctic and Antarctic areas despite the considerable distance from pollutant sources (Emnet et al., 2015; González-Alonso et al., 2017). Warm temperatures favor pollutant evaporation (tropical and subtropical land surface), while cool temperatures (high latitudes) favor deposition from the atmosphere on land/water. Climate change may moreover facilitate transport/deposit of contaminants in Polar areas.

Preliminary results obtained from research activities carried out by the AntaGPS project focused on the biomonitoring of metals in the livers of endemic Antarctic fish over a period of 12 years (Marrone et al., 2021). The two fish species used, icefish Chionodraco hamatus and red-blooded Trematomus bernacchii, were collected from the same area in the period 2002 to 2014. Fish are considered to be a particularly apt organism for the monitoring of pollution in aquatic ecosystems, with Antarctic organisms constituting excellent bioindicators for analysis of the trend of metal bioaccumulation over time, due to both endemism and singular adaptation to conditions. The liver was identified as an appropriate target organ for analysis of bioaccumulation and detoxification phenomena of metal pollution. The results obtained showed a significant increase over time in the bioaccumulation of numerous metals, such as Pb, Cd, Sr, Ni, Zn, which displayed a remarkable escalation in bioaccumulation in both species considered. The observed increase in lead bioaccumulation, despite its removal from gasoline at the beginning of the time period considered, is of particular interest (Figure 2). Furthermore, analysis of liver metallothionein-1 expression revealed a significant increase in this biomarker of metal pollution in both species, although with varying degrees, thus indicating a potential species-specific response to the presence of metals in seawater. The results of these studies will contribute to a better-targeted monitoring of the fragile Antarctic ecosystem and to obtaining critical clues indicating the global trend of metal pollution. The data thus obtained will be used to drive future conservation strategies for Antarctic and non-Antarctic ecosystems.

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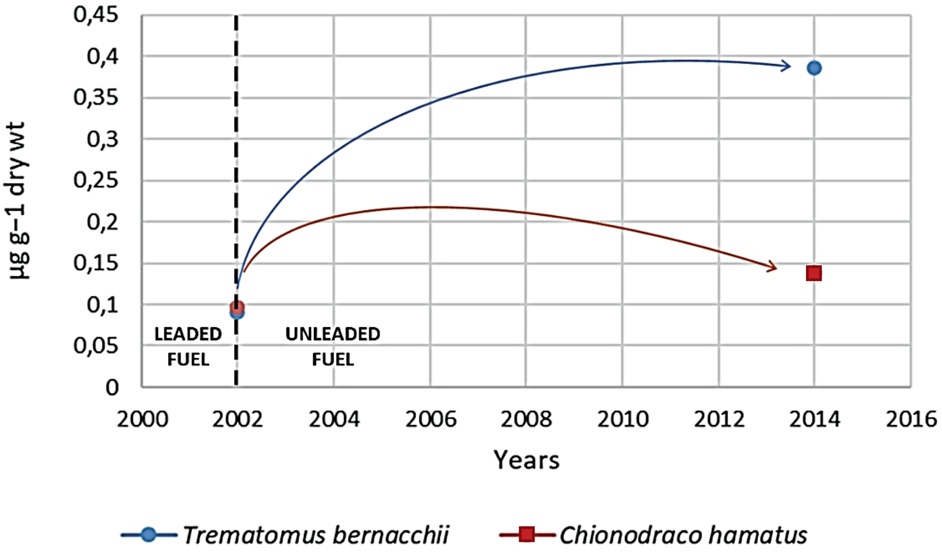

FIGURE 2: Pb bioaccumulation in two Antarctic fish species, from 2002 to 2014.

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A REVIEW OF RECENT TRENDS TO INCREASE THE SHARE OF POST-CONSUMER PACKAGING WASTE TO RECYCLING IN EUROPE

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ABSTRACT

Through the Circular Economy Package, the EU aims to reduce material imports and environmental impacts of waste management. Ambitious recycling targets should help to achieve these objectives. As a response, the waste industry developed technical and organizational solutions that can intervene at different stages of a waste management system in order to increase the recovery of recycling materials, starting with improved separate collection over the use of modern material recovery facilities up to the enhanced treatment of waste incineration ashes. The large question is how each of these options can contribute to increase recycling rates to achieve a circular economy. By reviewing case studies as presented in the most recent literature published since the year 2010 from European countries on the recycling of post-consumer packaging waste of glass, metals, paper, and plastics, this study contributes to answer this question. In the first stage, the review found 644 articles matching with the search terms and published since the year 2010. Of these, 45 remained for an in-depth analysis, since 599 did not present case studies as defined in the scope of this article. The articles reviewed provide a good overview on the state of knowledge on increasing recycling of post-consumer packaging waste by improved separate collection, material recovery facilities, and waste incineration bottom ash treatment. Additional information and case studies, however, are required, particularly large-scale experiments to test new separate collection systems, large-scale tests with different feedstock material at the most-modern material recovery facilities and waste incineration bottom ash treatment plants.

1. INTRODUCTION

The EU largely depends on imports of raw materials and consumer goods (Bruckner et al., 2012). The wastes generated from the consumption of these raw materials and consumer goods are only partly recycled, while the bulk is incinerated or landfilled (Pomberger et al., 2017). This leads to several environmental problems such as global warming and a reduction in ecosystems' quantity and quality. To reduce both, raw material imports as well as environmental impacts due to production of raw materials and consumer goods as well as disposal of wastes, the EU launched the circular economy package (CEP). The CEP, which consists of several directives, ordinances, and strategy documents, defines not only objectives, but also targets and measures to achieve a circular economy. An important target within the CEP is the achievement of recycling rates for selected recyclable materials in construction and demolition waste (Lederer et al., 2020), municipal solid waste (MSW) and particularly post-consumer packaging waste (PCPW) (Fellner et al., 2018; Fellner & Lederer, 2020; Tallentire & Steubing, 2020). For many countries in the EU, the fulfilment of the recycling rates will pose a large challenge, particularly since the definitions for their calculation were tighten (Weißenbach et al., 2020). It is clear that the CEP targets for recycling rates of PCPW can only be achieved if all sectors in the society will contribute to fulfil this task (Korhonen et al., 2018). For instance, the primary production sector (agriculture, forestry, mining) can increase the prices of primary raw materials, which would put secondary raw materials derived from waste recycling into a more competitive position. Also, the production sector can produce PCPW of longer durability and easier to recycle. Furthermore, the service sector can avoid the consumption of raw materials and PCPW generation by business models that promote multi-use instead of single use of packaging materials (Kalmykova et al., 2018). Despite all of these options the different sectors have, however, it is clear that the

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MSW management sector will also play a complementary role for achieving the recycling rates as defined in the CEP. To do so, the MSW management sector has several adjustment screws to turn, starting with the separate collection of PCPW (Mwanza et al., 2018), treatment in terms of sorting of mixed and separately collected PCPW (Cimpan et al., 2015), and finally the recovery of unburnable PCPW from the residues from MSW incineration, particularly bottom ash from grate incinerators and bed ash from fluidized bed incinerators (Bruno et al., 2021; Šyc et al., 2020).

This article reviews case studies from these stages of MSW management that enable the fulfillment of the CEP recycling rate targets. The research questions are:

What is the current status of MSW management in the EU with respect to the achievement of recycling rate targets as defined in the CEP for packaging wastes?

Which options are available at each stage of municipal solid waste management to increase the recycling rates of post-consumer packaging wastes?

How do they contribute to achieve the CEP recycling rate targets in the EU?

To answer these questions, first the scope of this work is defined in section 2, together with the review methodology. After presenting and discussing the results in section 3, a conclusion is presented in section 4.

2. METHODOLOGY

2.1 Scope definition

2.1.1 Product life cycle stages considered

Beside the selection of waste materials to be considered, the life cycle stages for options to achieve the CEP recycling rate targets covered in this article are restricted, namely to the MSW management sector. Other sectors such as primary raw materials extraction and processing (raw materials phase), design and production of products (design & production phase), and the use of these materials as consumer goods (use phase), are not covered, despite their relevance in achieving a circular economy (Kalmykova et al., 2018). Within the MSW management sector, the focus is on 1) the separate collection of recyclable PCPW (collection), 2) the sorting of PCPW containing MSW streams in material recovery facilities (pre-treatment MRFs/sorting), and 3) MSW incineration and the thereof produced bottom ashes (final treatment). Figure 1 shows the considered stages in the life cycle of a product.

2.1.2 Time period considered

Recycling is practiced by humans since the very beginning of their existence on earth. In modern societies, recycling gained much attention not only in the course of industrialization or in times of raw material shortages be-

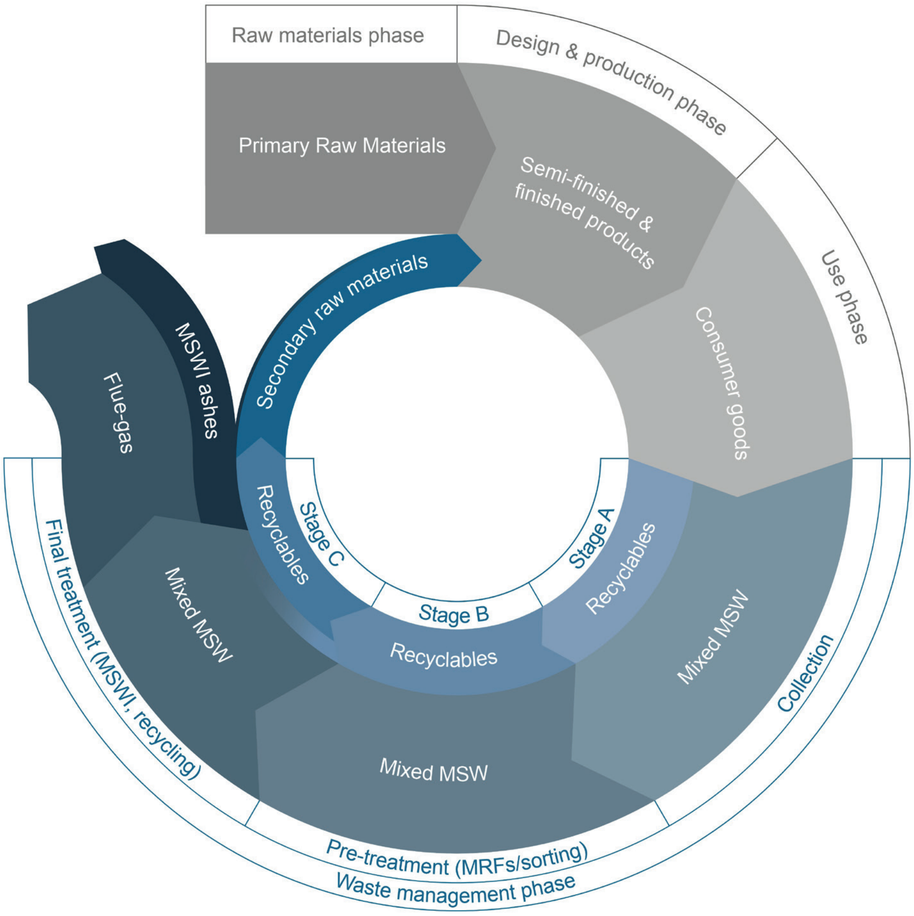

FIGURE 1: Stages in a products life cycle including the waste management phase (blue). Design by solo-ohne© (https://solo-ohne.com/).

tween the two world wars, but also with the emerging environmental discourse in the 1970ies (Keller, 2009; Strasser, 2000; Wilson, 2007). However, since the CEP is a relatively young concept and a lot of progress was made during the last decade, we consider in this article only recent developments. In detail, this means that only studies published between the years 2010 and 2021 were considered.

2.1.3 Geographic area considered

Concepts like the Circular Economy were developed in several world regions or countries at different times. Moreover, the EU's CEP spilled-over to countries and regions outside of the EU. However, since only EU member states are bound by the legal documents contained in the CEP, this article first covers case studies from the EU27 member states. In addition, countries with traditional and legislative bounds in the recent past and future, particularly the longtime EU member UK, and the EFTA states Island, Norway and Switzerland, were included.

2.1.4 Waste materials considered

The main focus of the article is post-consumer packaging waste (PCPW), usually contained in MSW. Other wastes of relevance such as agricultural waste, mining waste, construction and demolition waste, industrial waste, or waste from waste water treatment, are not covered. The waste materials considered are the quantitatively most relevant PCPW wastes for which a recycling rate target in the EU's packaging directive exists, namely aluminum, ferrous metals, glass, paper & cardboard, and plastic (EU, 2018). Wood packaging is not covered, due to its comparatively little quantitative relevance in packaging.

2.1.5 Type of studies considered

A large number of different types of studies on the CEP were published, including original research articles, conceptual works and some excellent review articles, for instance by Assi et al. (2020), Astrup et al. (2016), Blasenbauer et al. (2020); Cimpan et al. (2015), Dou et al. (2017), Rousta et al. (2017), Šyc et al. (2020), Verbinnen et al. (2017), Xevgenos et al. (2015), and Zhu et al. (2021). Since the aim of this article is to highlight original research articles, review articles are not covered. Furthermore, studies that provide economic, environmental or social impact assessments of circular economy scenarios that are not based on primary data on how to increase recycling rates, are not considered in this article. Finally, only original research articles which underwent an internationally considered review process by at least two independent reviewers, are selected for review. Studies presented in other sources that did not undergo such a review process, like student thesis, conference contributions, or working papers, are not selected.

2.2 Data collection and analysis

The data in this article are the studies reviewed, collected by using the online database of Scopus©, since these studies usually comply with the criteria for independent review before publishing. The search is a multi-stage one, in which each stage represents one of the most important steps in traditional MSW management systems. The search terms as well as the stage-wise extraction of relevant articles are presented in the subsequent subsection.

2.2.1 Search terms inserted

Stage 1 – separate collection of PCPW: the backbone of traditional recycling is the separate collection of PCPW (Mwanza et al., 2018). The reason for that is that separate collection of PCPW produces secondary raw materials of higher quality than sorting of mixed MSW (Cimpan et al., 2015). To unveil selected literature on this topic, the search terms "waste" AND "recycling" AND "separate collection" were used.

Stage 2 – waste pre-treatment by material recovery facilities (MRF): when separate collection of PCPW cannot further increase the separate collection rate, or when the costs are too high (D'Onza et al., 2016; Feil et al., 2017; Janz et al., 2011), the extraction of recycling materials by MRFs is suggested as a viable option (Cimpan et al., 2016). Currently an even more important role than by these multi-stream MRFs is played by single- or dual-stream MRFs (Antonopoulos et al., 2021). The case studies published in the literature on these topics are aimed to be found by using the search terms "waste" AND "material recovery facility". The term "recycling" is not used since it is metaphorically included in "material recovery facility".

Stage 3 –MSW incineration bottom ash (IBA) treatment: unburnable waste fractions can also be recovered from MSW IBA. From the considered PCPW fractions, this counts for metals and, in theory, glass (Chimenos et al., 1999; Šyc et al., 2018; Šyc et al., 2020). To consider this option, the search terms "waste" AND "incineration" AND "recycling" AND "bottom ash" are introduced.

2.2.2 Reduction procedure and data analysis

First, all articles were analyzed based on their title, keywords, and abstract. Based on that articles that presented a case study outside of the EU or that focused on a waste type not considered, were eliminated. For the remaining articles, also the full text was analyzed and based on that, articles that did not match with the type of studies defined, were eliminated. The final list that remained was then used in this review and they are presented in this work.

3. RESULTS AND DISCUSSION

3.1 Separate collection

The first search gave in total 219 articles published between the years 2010 and 2021 that contained the search terms "waste" AND "recycling" AND "separate collection". After eliminating these with a geographic focus outside of the one defined, a total number of 163 articles remained. A further elimination of these studies that considered a waste material which was not in the scope of this article, for instance biowaste, waste electronic and electrical equipment, or waste textiles, the search yields 119 articles. From these, 15 articles remained for the in-depth content analysis, as the other articles did not contained case studies as desired by the authors. These 15 articles were analyzed in detail.

Five of the articles used the country or national level for their investigation, and eleven city or municipality level. The Netherlands was the country that contained with six the most case studies (Dijkgraaf & Gradus, 2020; Feil et al., 2017; Picuno et al., 2021; Seyring et al., 2016; Thoden van Velzen et al., 2019; Warrings & Fellner, 2019), followed by Italy with five (Bertanza et al., 2021; Del Cimmuto et al., 2014; Romano et al., 2019; Seyring et al., 2016; Warrings & Fellner, 2019), Germany with four (Feil et al., 2017; Picuno et al., 2021; Seyring et al., 2016; Warrings & Fellner, 2019), Austria (Picuno et al., 2021; Seyring et al., 2016; Warrings & Fellner, 2019), Finland (Dahlbo et al., 2018; Seyring et al., 2016; Warrings & Fellner, 2019), Sweden (Rousta et al., 2016; Seyring et al., 2016; Warrings & Fellner, 2019), Spain (Gallardo, Bovea, Colomer, et al., 2012; Gallardo, Bovea, Mendoza, et al., 2012; Seyring et al., 2016) and United Kingdom (Seyring et al., 2016; Wang et al., 2020; Warrings & Fellner, 2019) with three each; Belgium, Czech Republic, France, Portugal (Seyring et al., 2016; Warrings & Fellner, 2019) and Poland with two case studies (Picuno et al., 2021; Seyring et al., 2016). Switzerland (Haupt et al., 2018) as well as all other member states of the EU28 (Seyring et al., 2016) that have hitherto not been mentioned were covered in one case study. With respect to packaging waste materials, of the ones considered, plastics was contained in all but one case study that solely focused on aluminum. Eleven studies considered metals, seven paper & cardboard as well as glass, while beverage cartons were contained in one case study only. Eight case studies provide a comparison between different municipalities or cities, five a comparison of two years between which a historical development took place, and two case studies provided a cross-country comparison. One case study provided an experiment in terms of analyzing the status quo, carrying out an intervention and then analyzing the result of the experiment (Rousta et al., 2016). Table 1 gives an overview and description on the 16 studies analyzed.

Some of the studies analyzed selected determining factors for separate collection rates. One finding was that the higher the population density and population numbers are, the lower the separate collection rate is. Examples come from Germany and the Netherlands (Feil et al., 2017), Poland (Połomka et al., 2020), but also from analyzing data for the EU-28 (Seyring et al., 2016). In their report for the European Commission, Seyring et al. (2015) showed that the average separate collection rate of packaging materials and biowaste is, with few exceptions, much lower for the capitals of the EU-28 than for the regarding countries. This negative correlation between population density and separate collection is shown for the EU-28 in Figure 2. Contrary to that, Romano et al. (2019) found that in the case of Tuscany region in Italy, the separate collection rate increased with higher population density. For this reason, Romano et al. (2019) suggested to introduce actions that raise awareness and encourage pro-environmental behavior of inhabitants of low-density, rural areas. Since population density can hardly be influenced by waste management, only one of these studies that found a negative relation between separate collection rates and population density, namely Feil et al. (2017), suggested to overcome this gap by introducing post-sorting of recycling materials from mixed MSW by MRFs in areas with high population density and low separate collection rate. Figure 2 shows the separate collection for the EU-28 countries and their capitals, based on data from Seyring et al. (2015).

A more often investigated factor that determines the separate collection rate is the collection system, for instance if it involves a deposit-refund system (DRS) for single use beverage containers or not. All studies that investigate DRS agree that it leads to higher separate collection rates than systems without DRS (Dahlbo et al., 2018; Picuno et al., 2021; Seyring et al., 2016; Warrings & Fellner, 2019). However, the case of aluminum beverage cans shows that a sophisticated separate collection system (e.g. in Italy) or post-sorting of mixed MSW or IBA (e.g. in Netherlands, Belgium, France, Austria) reduces the gap to countries with DRS (Warrings & Fellner, 2019). Also, pay as you throw (PAYT) programs positively influenced the separate collection rate (Seyring et al., 2016; Thoden van Velzen et al., 2019).

Another option to increase separate collection is to reduce the distance for users to the separate collection container by shifting from drop-off to kerbside or door-to-door collection. This had positive effects in many cities and countries (Bertanza et al., 2021; Dahlbo et al., 2018; Dijkgraaf & Gradus, 2020; Gallardo, Bovea, Colomer, et al., 2012; Seyring et al., 2016; Wang et al., 2020). Particularly impressing are the increase in the separate collection rate in Brescia by a factor of 2-3 within three years (Bertanza et al., 2021) and Nottingham with a factor of 1.5 within one decade (Wang et al., 2020). With respect to the quality of the collected material, for instance in terms of extraneous material contents, Bertanza et al. (2021) pointed out that a decrease in the quality was expected. At the paper sorting facility that receives the separate collected materials from Brescia, however, no increase in the rejects was recorded. Information on whether there was an increase in rejects in the paper mill was not available to the authors. The result of Brescia is also displayed in Figure 4, based on data from Bertanza et al. (2021).

In addition, two studies specifically deal with the question of number of drop-off points and distance to households. Gallardo, Bovea, Mendoza, et al. (2012) found a clear negative correlation between distance to drop-off points and separate collection rate for a selection of Spanish municipalities with more than 50,000 inhabitants. Haupt et al. (2018) showed for the example of PET beverage bottle collection in Switzerland that the number of drop-off points increases much faster than the amount of PET beverage bottles collected and recycled. This result which indicates a ceiling for separate collection rates even when the collection system is expanded, is shown in Figure 5 based on data from Haupt et al. (2018).

Finally, some of the studies claim that comparison of different separate collections systems should consider the composition of the collected fractions. Gallardo, Bovea, Colomer, et al. (2012) concluded in their comparison of Spanish municipalities larger than 5,000 inhabitants that a door-to-door collection of commingled recycling materials (metals, paper and cardboard, plastics) is the most

TABLE 1: In-depth analyzed articles on separate collection of waste (n=15).

No.	Authors	Level	Description of area	Materials	Content of study	Comparison
Sc01	Picuno et al. (2021)	Country	AT, GER, NL	plastics	Comparison of collected, sorted, recycled plastics in AT, DE, NL	Cross-country
Sc02	Bertanza et al. (2021)	City	Brescia, IT	glass, metals, plastics, paper	Historical development of separate collection rates in Brescia over 30 years, particularly after changing from bring to kerbside collection	Historical development
Sc03	Dijkgraaf & Gradus (2020)	Country	NL	plastics	Comparison of plastic amounts for recycling from 99 municipalities in NL with different collection systems, including MRFs for mixed MSW	Cross-municipality
Sc04	Połomka et al. (2020)	Municpality	22 Municipalities in Marszów, PL	LWP (metals, plastics)	Comparison of the development of separate collected LWP (metals, plastics) in selected rural and urban municipalities in PL	Cross-municipality
Sc05	Wang et al. (2020)	City	Nottingham, UK	glass, metals, plastics, paper	Separate collection rates in Nottingham between 2006 and 2016, after changing from drop-off points to kerbside collection	Historical development
Sc06	Romano et al. (2019)	Municipality	Municipalities in Tuscany, IT	glass, metals, plastics, paper	Comparison of separate collection rates of municipalities with different organization form (public / private operator)	Cross-municipality
Sc07	Thoden van Velzen et al. (2019)	Municipality	Oosterhout & Waal-wijk Municipality, NL	beverage cartons, metals, plastics	Separate collection rates of beverage cartons, plastics, and metals from 21 households in two municipalities in NL; based on that, re-calculation of separate collection rate in 13 municipalities in NL	Cross-municipality
Sc08	Warrings & Fellner (2019)	Country	AT, BE, CZ, FR, GER, GR, IT, NL, PT, SE, UK	metal (aluminum)	Comparison of separate collection, post-sorting (from incineration bottom ash) and disposal rates of aluminum beverage cans in 11 European countries, including countries with deposit system	Cross-country
Sc09	Haupt et al. (2018)	Country	CH	plastics (PET)	Separately collected and recycled amounts of PET beverage bottles in CH over 25 years, considering extension of collection points (bring system) and PET bottle content in collected amounts	Historical development
Sc10	Dahlbo et al. (2018)	Country	FI	plastics	Separately collected and post-sorted plastics in FI before and after extension of separate collection and installing an MRF for mixed MSW	Historical development
Sc11	Feil et al. (2017)	Municipality	Northrhine-West-phalia, GER; NL	plastics	Separately collected plastics in municipalities of different population densities in GER and NL and estimation of potential of plastics from automatic sorting in a material recovery facility for mixed MSW	Cross-municipality comparison
Sc12	Seyring et al. (2016)	City	28 capital cities in Europe	glass, metals, plastics, paper	Comparison of separate collected glass, metals, paper, plastics in 28 EU-capitals, distinguishing between separate collection system	Cross-municipality
Sc13	Rousta et al. (2016)	Municipality	Borås Municipality, SWE	glass, metals, plastics, paper	Comparison of separate collected glass, metals, paper, plastics in a pilot area in Borås before an after interventions (information campaigns, decreasing distance to drop-off centers)	Experiment
Sc14	Gallardo et al. (2012)	Municipality	115 Municipalities, pop. >5,000, ES	glass, metals, plastics, paper	Comparison of the separate collection rate of glass, metals, paper, plastics of cities with different collection systems (kerbside, drop-off)	Cross-municipality
Sc15	Gallardo et al (2012)	Municipality	45 Municipalities, pop. >50,000, ES	glass, metals, plastics, paper	Comparison of the separate collection rate of glass, metals, paper, plastics of cities with varying distance to drop-off points	Cross-municipality

favorable option, not mentioning the potential losses during sorting in an MRF. These losses, however, can be substantial, depending not only on the MRF technology, but also the quality of the input material (Antonopoulos et al., 2021). Some authors argue that this quality of separately collected PCPW decreases also when extending the separate collection system. Haupt et al. (2018) conclude that the extension of the PET-beverage bottle collection system in Switzerland lead to a higher amount of extraneous materials disposed-off in the collection containers. A similar finding was made by Thoden van Velzen et al. (2019) who showed that higher amounts of recycling materials separately collected does not necessarily mean a better performance, since undesired fractions may increase

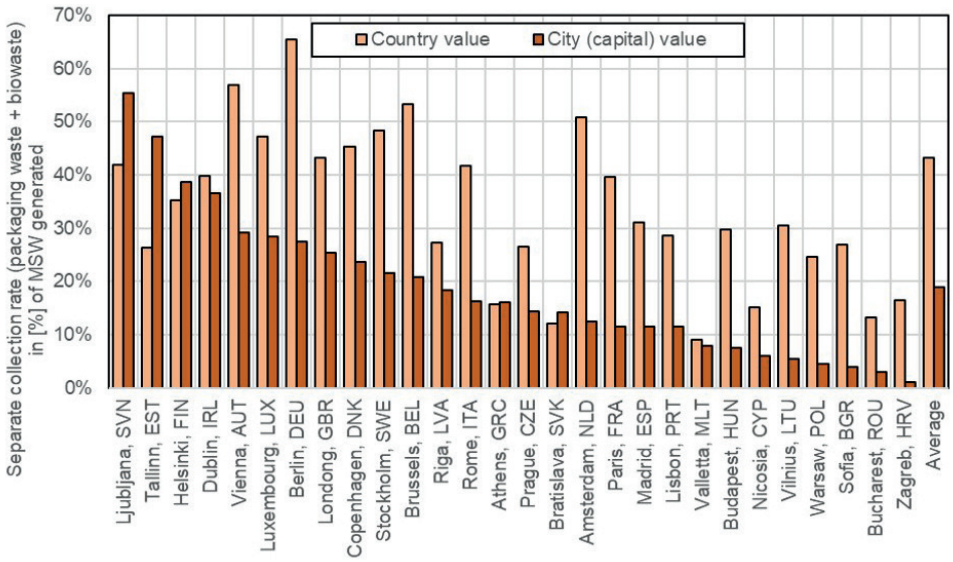

FIGURE 2: Separate collection rates of packaging waste and biowaste in the EU-28 countries and their capitals (cities). Data from Seyring et al. (2015).

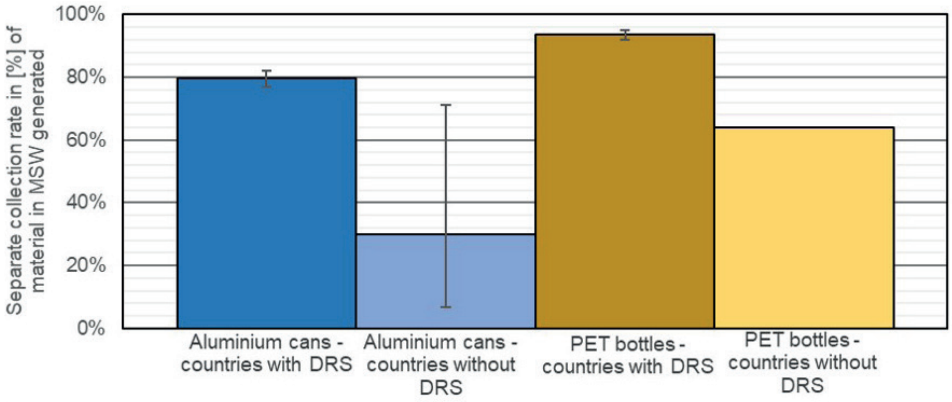

FIGURE 3: Separate collection rates of aluminium cans and PET bottles with and without deposit-refund system (DRS), based on Warrings and Fellner (2019) for aluminium and Picuno et al. (2021) for PET.

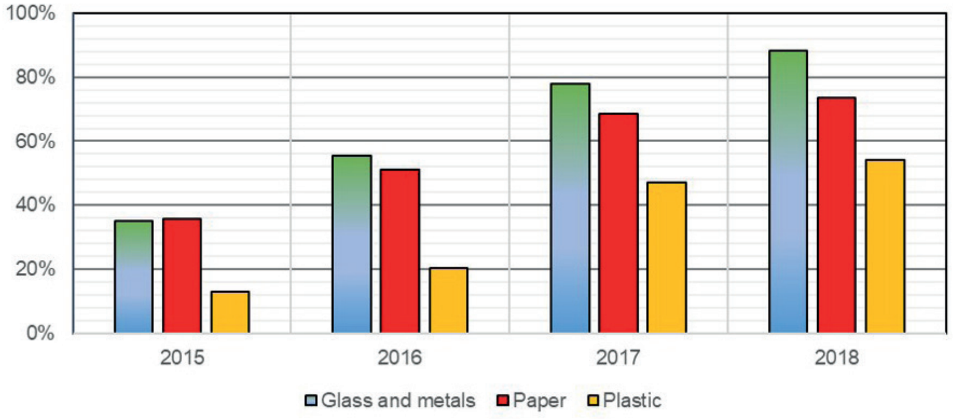

FIGURE 4: Development of separate collection rates of glass & metals, paper & cardboard, and plastics in Brescia after shifting from drop-off to door-to-door collection. Data from Bertanza et al. (2021).

over-proportionally. Also, Połomka et al. (2020) found for their Polish region under investigation that the share of undesired fractions in separately collected lightweight packaging waste increased with extension of the collection service and amounts, while the desired fractions decreased. Furthermore, Wang et al. (2020) showed that in Notting-

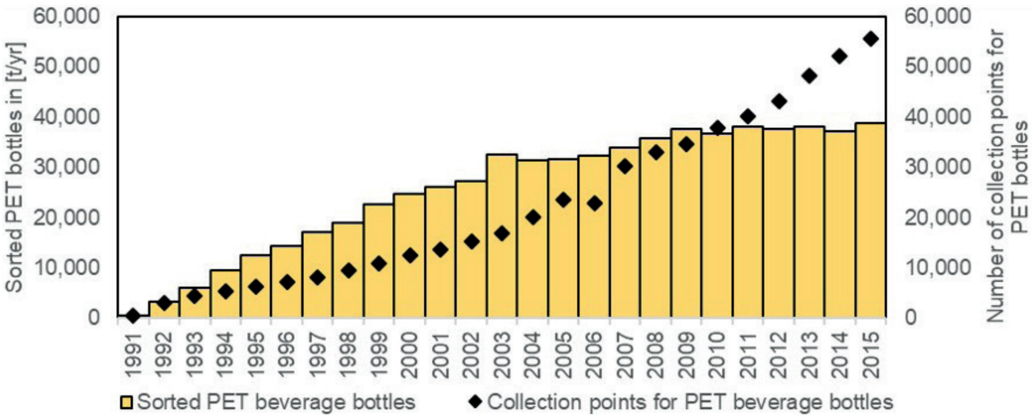

FIGURE 5: Development of the number of drop-off collection points and the separately collected and sorted amounts of PET beverage bottles in Switzerland. Data from Haupt et al., (2018).

ham, the extension of separate collection not only lead to higher amounts of PCPW collected, but also that the share of waste input that was sorted out for recycling declined, from 99.6% in the year 2006/2007 to 81.8% in the year 2016/2017. All of this indicates a decrease in the quality when separate collection systems were extended, and for this reason, data that solely rely on the separately collected amount of recycling material not considering its composition, can hardly serve to assess the efficiency of a separate collection system (Thoden van Velzen et al., 2019).

3.2 Material recovery facilities

The first search yielded 150 articles published in 2010-2021 containing the terms “waste” AND “material recovery facility”. By eliminating 88 articles outside of the defined geographic area, 62 articles remained. Further eliminating these which are out of the scope by either a a focus on a different material than PCPW or from the method used (e.g. review) or data presented (e.g. solely secondary data already published in one of the other articles selected), solely 14 articles, shown in Table 2, remained for the in-depth content analysis.

Of the 14 articles selected, one was at country level, six investigated MRFs as part of the MSW management system of a city or region, six were on plant level only, and one was at the level of a paper factory that received waste paper from different MRFs. Four studies were related to the UK (Kirk & Mokaddam, 2021a, 2021b; Miranda et al., 2013; Wang et al., 2020) and Spain (Ip et al., 2018; Istrate, Galvez-Martos, et al., 2021; Istrate, Medina-Martos, et al., 2021; Miranda et al., 2013), and three to Italy (Ardolino et al., 2017; Gadaleta et al., 2020; Mastellone et al., 2017). One study each was on Portugal, Germany, Austria and without any reference (Cimpan et al., 2016; Dias et al., 2014; Griffiths et al., 2010; Warrings & Fellner, 2021). With respect to input flows, eight studies consider MRFs for mixed MSW, seven for lightweight packaging (LWP) of plastics, metals, and sometimes lightweight beverage cartons (LBCs), and two on commingled packaging waste (CPW) that also include paper or glass. Most studies analyzed MRFs that aim to recover metals (ten), plastics (nine), glass (seven), paper (four), and LBCs (two). However, the most detailed analysis was by most studies carried out on plastics.

Two important indicators for the sorting process in an MRF are the sorting efficiency per material defined as the amount of material in the output flow that is sent to recycling in mass percent of the amount of the material in the input flow, shown in Table 3, and the purity of the output flow defined as the content or grade of the desired material, shown in Figure 6.

The studies agree in the point that sorting efficiencies are generally higher for MRFs feed with commingled (COM) or lightweight packaging PCPW (LWP), as well as for metals, PET and HDPE (Cimpan et al., 2016; Gadaleta et al., 2020; Ip et al., 2018; Istrate, Medina-Martos, et al., 2021). For the latter materials, the values are in the range of other sources (Antonopoulos et al., 2021; Brouwer et al., 2019; Brouwer et al., 2018; Van Eygen et al., 2018). However, the data of Cimpan et al. (2016) suggests that there is a large difference between plants as designed and in their typical operation. This finding by Cimpan et al. (2016) is shown in Table 3.

With respect to the purity of the material for recycling produced, better values were achieved for PET and HDPE than for LDPE and PP. Values were also good for metals and lightweight beverage cartons (LBC), however, there were not many case studies for these materials (see Figure 6). This lack of data was also a problem that Warrings and Fellner (2021) faced in their study. Nevertheless, the input data, i.e. the sorting efficiency on which the authors calculate the increase of the sorted aluminum rates, was not that clearly expressed. Figure 6 shows the purity of the desired material fraction extracted from the MRFs, based on data from Mastellone et al. (2017), Gadaleta et al. (2020), and Ip et al. (2020).

For paper PCPWs, little data was available, particularly with respect to the quality of the output flows in terms of purity. However, the study of Miranda et al. (2013) shows that paper particularly from suppliers from the UK operating small MRFs for commingled PCPW was of low quality. Compared to paper and the other materials, there was even less data on the sorting and the quality of glass, suggesting that it is neither collected with other PCPW, nor sorted from mixed MSW that often, even though there is a large

TABLE 2: In-depth analyzed articles on material recovery facilities (n=14).

No.	Authors	Level	Area	Input	PCPW	Content of study
MRF01	Istrate et al. (2021)	Plant, City	Madrid, ESP	MSW COM	glass metals plastics paper	Material flow analysis (MFA) of MRFs for mixed MSW and commingled PCPW. Both sorted 1% / 23% glass, 44% / 66% iron, 12% / 33% aluminum, 7% / 20% paper, 13% / 20% cardboard, 3% / 43% carton, 5% / 82% PET, 5% / 82% HDPE, 5% / 76% LDPE related to the input (MSW MRF / commingled).
MRF02	Istrate et al. (2021)					
MRF03	Warrings & Fellner (2021)	Country	AUT	MSW LWP	aluminum	Scenarios to increase aluminum recycling concluding that mixed MSW MRFs and improved MSW IBA treatment were the best measures.
MRF04	Kirk & Mokaddam (2021a, b)	Plant, City	London, GBR	MSW	glass metals plastics paper	Sampling input material or mixed MSW MRFs, showing high variation depending on the company delivering the MSW, but also the impact on operation costs and revenues of the MRFs.
MRF05						
MRF06	Gadaleta et al. (2020)	Plant, Region	Bari, ITA	LWP		Determining the sorting efficiency (recovery index RI) and quality (purity index PI) of plastic PCPW from an MRF called ASM. Both indicators were high for PET, HDPE, PP and LDPE >A3, but low for LDPE<A3. All indices were higher than in the MRF Bedonia analyzed earlier by the authors.
MRF07	Wang et al. (2020)	Plant	Notting-ham, GBR	MSW LWP	glass metals plastics paper	MRFs for commingled PCPW and mixed MSW + other waste extracted 82% and 9% of PCPW for recycling, respectively. The commingled MRF contributed to 93% of glass, 95% of paper, 23% of metal and 78% of plastic, and the mixed MSW MRF to 26% of metals and 4% of plastics sorted for recycling. Separate collection contributed 7% glass, 5% paper, 17% metals, and 18% plastics, and MSW IBA treatment 34% metals for recycling.
MRF08	Ip et al. (2018)	Plant	Toledo, ESP	LWP	glass metals plastics	MFA of a commingled PCPW MRF. Grades (purity) of outputs and sorting efficiencies were high for iron, aluminum, PET, HDPE, LBC.
MRF09	Ardolino et al. (2017)	Plant, City	ITA	MSW	metals plastics	An LCA was performed for an mixed MSW MRF, focusing on biowastes. The mixed MSW had 26% plastics and 4% iron, of which 0.9% of plastics and 0.01% of iron were sorted by MRF.
MRF10	Mastellone et al. (2017)	Plant	ITA	LWP	metals plastics	MFA of an MRF for LWP (plastic, ferrous metals, aluminum) was performed by collecting samples of the input and output materials. Based on that, different indicators were calculated representing the efficiency of sorting and the purity of the output material. These were calculated as a time series to show temporal variations.
MRF11	Cimpan et al. (2016)	Plant	DE	LWP	metals plastics LBC	A study that shows great discrepancies between sorting efficiency as designed and in real operation, including an economic analysis.
MRF12	Dias et al. (2014)	Plant	POR	MSW	glass	Amount and content of glass in the heavy output of five mechanical-biological treatment plants in Portugal. Glass dry matter contents 33-83%.
MRF13	Miranda et al. (2013)	Factory	ESP, GBR	Single Com-in-gled	paper	Comparison of the quality of waste paper from old and modern commingled PCPW MRFs, showing better values of the latter. Qualities are worse than in separate collection.
MRF14	Griffiths et al. (2010)	Plant	None	LWP MSW		Engineering principles of MRFs applied to separately collected PCPW (clean MRF) and to mixed MSW (dirty MRF).

potential as well as content in certain output fractions of mixed MSW MRFs (Dias et al., 2014).

3.3 Recovery of recycling materials from waste incineration bottom ash

The first search yielded 275 articles published in 2010-2021 containing the search terms. Then, 121 articles not in the defined geographic area were eliminated and 154 articles remained. A large number of 138 article were eliminated as they focused on other materials or topics. The most important topic therein was the recycling of the mineral fraction of MSW IBA in construction materials like cement, concrete, or as base-layer in road constructions. At the end, 16 articles remained for the in-depth analysis, as shown in Table 4.

All articles dealt with IBA from grate incinerators, but none on fluidized bed incinerators or rotary kilns. Two of the articles investigated recycling of PCPW from MSW IBA at European level (Abis et al., 2020; Bruno et al., 2021), another two at national level (Grosso et al., 2011; Warrings & Fellner, 2019). All other articles investigated the topic at the level of MSWI or MSW IBA treatment plants. Therein, Italy is represented in six articles (Abis et al., 2020; Biganzoli & Grosso, 2013; Biganzoli et al., 2014; Biganzoli et al., 2013; Bruno et al., 2021; Warrings & Fellner, 2019), Germany and Denmark in five (Abis et al., 2020; Allegrini et al., 2014; Bruno et al., 2021; Gökelma et al., 2021; Holm & Simon, 2017; Huber, 2020; Warrings & Fellner, 2019), Austria, the Netherlands and Belgium in four (Abis et al., 2020; Bruno et al., 2021; Hu et al., 2011; Huber, 2020; Van Caneghem et al., 2019; Warrings & Fellner, 2019), Spain and the UK in three (Abis et al., 2020; Bruno et al., 2021; del Valle-Zermeño et al., 2017; Gökelma et al., 2021), Switzerland and all other member countries of the EU in two (Abis et al., 2020; Bruno

TABLE 3: Sorting efficiencies of MRFs (Cimpan et al., 2016; Gadaleta et al., 2020; Ip et al., 2018; Istrate et al., 2021). For PET, the non-weighted mean of different fractions was used.

		Istrate et al. (2021b)	Istrate et al. (2021a)	Cimpan et al. (2016)	Cimpan et al. (2016)	Cimpan et al. (2016)	Cimpan et al. (2016)	Cimpan et al. (2016)	Cimpan et al. (2016)	Gadaleta et al. (2020)	Gadaleta et al. (2020)	Ip et al. (2020)
Input material		MSW	COM	COM	COM	COM	COM	COM	COM	LWP	LWP	LWP
Operation		typical	typical	typical	typical	typical	designed	designed	designed	typical	typical	modelled
Plant type / name		Madrid	Madrid	basic	medium	advanced	basic	medium	advanced	ASM SSC	Bedonia SSC	Toledo
Glass	Glass	1%	23%									
	Aluminium	12%	33%	30%	35%	40%	60%	70%	80%			82%
	Ferrous	44%	66%	40%	44%	48%	80%	88%	95%			92%
Paper	Paper	7%	20%	25%	30%	35%	50%	60%	70%			
	Cardboard	13%	20%	25%	30%	35%	50%	60%	70%			
	Carton	3%	43%	25%	30%	35%	50%	60%	70%			
	LBC			35%	39%	43%	70%	78%	85%			90%
Plastics	PET	5%	82%			40%			80%	94%	57%	98%
	HDPE	5%	82%		27%	27%		54%	54%	97%	97%	88%
	LDPE small	5%	76%	25%	30%	35%	50%	60%	70%	30%	21%	
	LDPE large									85%		
	PP				27%	27%		54%	54%	97%	78%	

et al., 2021) and all other EFTA and EU candidates hitherto not mentioned in one (Bruno et al., 2021). All articles focused on metals, except two that at least mentioned another glass too (Bruno et al., 2021; del Valle-Zermeño et al., 2017).

The guiding research question in almost all articles deals with the recovery of metals from MSW IBA. For packaging metals, which are either iron or aluminum based, the latter received much more attention than the prior. This may have to do with the already high efficiency and thus little losses of iron through magnetic separation, if compared to aluminum (Allegrini et al., 2014; Huber, 2020; Mehr et al., 2021). The latter is basically lost in two ways of oxidation (thermal oxidation during incineration, or oxidation by wheathering in the IBA treatment), or due to low sorting efficiencies. Thermal oxidation takes place twice, during the incineration process itself, but also in the metal smelting. That these losses can be substantial is shown by a number of studies. Biganzoli et al. (2014) estimated a loss of 54-63% during incineration. Allegrini et al. (2014) stated personally communicated losses in metal smelting of between 19-34%, depending on the grain size, while Mehr et al. (2021) reported 14%. Iron is mainly lost in the treatment only, and oxidation seems to be in a lower range (Mehr et al., 2021). The reason for that is that aluminum packaging occurs much more as flexible packaging with thin layers, making it more exposed to temperature (Biganzoli & Grosso, 2013; Biganzoli et al., 2014; Biganzoli et al., 2013; Gökelma et al., 2021). With respect to the sorting efficiency of IBA treatment plant, Figure 7 shows some values from literature. These do not contain oxidation losses during smelting.

The results shown in Figure 7 suggest that there are huge differences in the recovery of PCPW metals from MSW IBA. This also means that there is still a large potential available, not only from packaging, but also from non-packaging metals which are sometimes in practice incorrectly assigned to packaging (Van Caneghem et al., 2019). An even larger potential than for metals is present for glass PCPW, which is only mentioned by two of the reviewed studies (Bruno et al., 2021; del Valle-Zermeño et al., 2017). Neither of these two studies, however, actual-

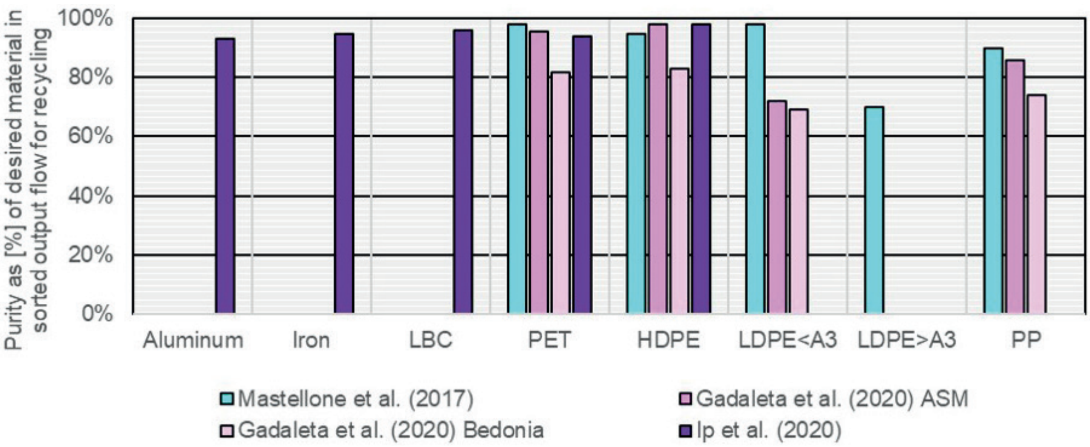

FIGURE 6: Purity of in MRF produced material flow for recycling, defined as the content or grade of the desired material, based on literature (Mastellone et al., 2017; Gadaleta et al., 2020; Ip et al., 2020). ASM and Bedonia stands for two different MRFs.

TABLE 4: In-depth analyzed articles on separate collection of waste (n=16).

No.	Authors	Level	Area	Input	PCPW	Content of study
IBA01	Bruno et al., (2021)	Continent	Europe (EU27, EU candidate countries, EFTA, UK)	MSW	metals	Scenario on the effects of reducing the amount of untreated IBA to 0 and implementing state-of-the-art IBA treatment in 38 European countries on recycling of minerals and metals. Metal reco-very could increase from 1.83 Mt/a to 3.77 Mt/a.
IBA02	Gökelma et al., (2021)	Plant	8 samples from USA, UK, DK	MSW	aluminum	Recyclability of aluminum in IBA, determined by oxide layer thickness, metal yield and coagulation efficiency after re-melting. 76-93% of aluminum was recovered as metal.
IBA03	Mehr et al., (2021)	Plant	CH	MSW	metals	LCA based on updated data of the dry IBA treatment facility in Hinwill. Data for metals including iron, stainless steel, and aluminum >0.3 mm grain size. Expressed in extraction efficiency and recycling efficiency (i.e. substitution of primary raw material).
IBA04	Abis et al., (2020)	Continent	EU 28	MSW	metals	Analysis of the nexus between recycling and MSWI, highlighting the relevance to improve recovery of metals and minerals from IBA.
IBA05	Huber, (2020)	Plant	GER, AT, IT	MSW	metals	Modelling the material flows of metals and minerals based on data from 5 IBA treatment plants from AUT, DEU and ITA. Recovery rates and transfer coefficients are provided.
IBA06	Van Cane-ghem et al., (2019)	Region	Flanders, BEL	MSW	metals	MFA of packaging and non-packaging metals in Flanders, based on IBA sampling. Validation of recycling rates of metal packaging.
IBA07	Warrings and Fellner, (2019)	Country	AT, BE, CZ FR, GER, GR, IT, NL PT, SE, UK	MSW	aluminum	Comparing sorting rates for aluminum packaging waste in 11 EU countries, considering separate collected and IBA recovered metals. The latter are assumed to be higher than reported.
IBA08	Haupt et al., (2017)	Plant	CH	MSW	iron	Investigating the quality of steel scrap from IBA and other scrap, showing lower quality and higher recycling energy demand of IBA scrap.
IBA09	del Valle-Zermeño et al., (2017)	Plant	ES	MSW	glass	Determining the impact of separate collection of glass on the glass content in IBA by sampling inputs and outputs of an IBA treatment plant
IBA10	Holm and Simon, (2017)	Plant	DE	MSW	metals	Comparing two dry and one wet treatment plant for IBA with a focus on use of the mineral fraction for construction, thereby establishing data on recovery rates for metals.
IBA11	Biganzoli et al., (2014)	Plant	IT	MSW	aluminum	MFA of aluminum in two italian MSWI plants with attached IBA treatment. 21-23% of aluminum was recovered. This can be increased by improvement to 28-38%. 54-63% are oxidized in MSWI and thus lost to recovery, but contributing to 1% of energy production.
IBA12	Allegrini et al., (2014)	Plant	DK	MSW	metals	MSW IBA treatment in a plant in DK was analyzed. 85% of iron and 62% of aluminum in IBA was found to be recovered
IBA13	Biganzoli and Grosso, (2013)	Plant	IT	MSW	aluminum	MFA of aluminum in two MSWI plants with attached IBA treatment in Italy, focusing on packaging aluminum. 81% of cans can be recovered, but only 51% of tray, 27% of mix and 47% of paper laminated foils.
IBA14	Biganzoli et al., (2013)	Plant	IT	MSW	aluminum	Testing of improved aluminum recovery from IBA fine fraction <4mm by H2 production. 15% was metallic and of this, 21% were recovered.
IBA15	Hu et al., (2011)	Plant	NL	MSW	aluminum	Comparing the recovery rates of different aluminum packaging. These were around 90-95% for cans, 85% for foil containers and 77% for thin foils, expressed in metallic aluminum.
IBA16	Grosso et al., (2011)	Country	IT	MSW	aluminum	Modelling aluminum recovery for scenarios in Italy for the MSW management system, including IBA treatment.

ly focused on recovering glass from IBA, even though del Valle-Zermeño et al. (2017) showed that there are large amounts of glass in IBA. One reason for that might be that past experiences with glass recovery from MSW IBA produced in a grate incinerator with wet discharge were not that successful (Šyc et al., 2020).

3.4 Towards a recycling-based circular economy from a systems perspective

The studies reviewed showed not only that there is already a lot of literature and thus knowledge available to increase the amount of PCPW available for recycling. They also suggest some important future directions of research.

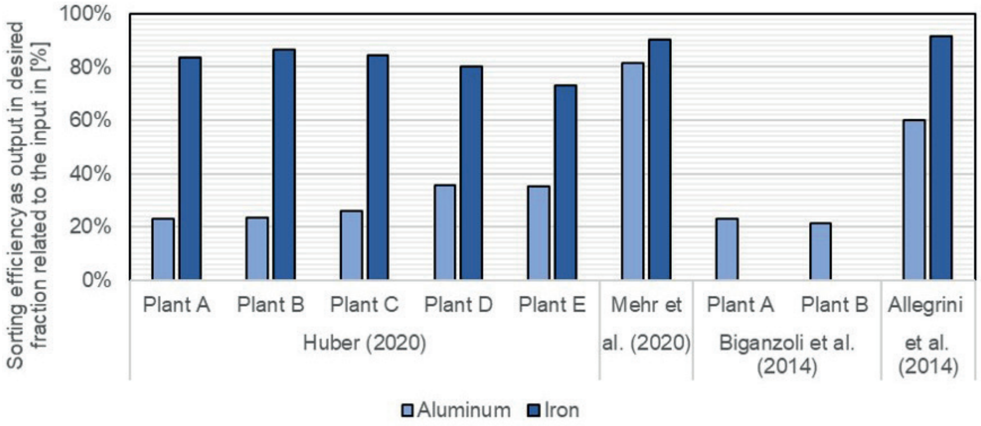

FIGURE 7: Sorting efficiency of IBA treatment plants. In Huber (2020), Plant C refers to the plant described by Allegrini et al. (2014) and Plants D and E to the plants in Holm and Simon (2017), all adapted to the Austrian situation.

For separate collection, which will very likely be the backbone of any recycling system also in future, a number of different systems are available, from single stream over combined light weight packaging (lightweight beverage cartons, metals, plastics) or mixed packaging (glass, metals) to commingled systems collecting all PCPW in one container (Cimpan et al., 2015). Yet, the question is which system fits best under certain circumstances and contexts. Such a context can be the population density or the size of a municipality. For instance, in dense populated areas where average housing area per capita is too scarce to reserve a lot of space for a large number of different bins for recyclable PCPW, commingled systems might be one alternative which should be tested before implementation, for instance in large-scale experiments that also involve the most important stakeholders of separate PCPW collection (Lederer et al., 2015; Pedersen & Manhice, 2020). This would mean that the decision on PCPW separation is shifted from the consumer to central sorting facilities, i.e. MRFs (Gundupalli et al., 2017). The recent progress in automatic sorting allows this option, even up to a level that mixed MSW can undergo such a recovery process (Feil et al., 2017). Event though the quality of the recycling material produced in these mixed MSW MRFs is expected to be lower for materials like paper and plastics, they provide a cost-effective measure to recover materials that otherwise would be incinerated (Janz et al., 2011). Nevertheless, additional post-treatment steps might be required to improve the quality of PCPW sorted from mixed MSW, but considering the size of the problem current societies are facing, it can be expected that the technological progress to deal with these quality issues will be available to recycling systems in the near future. For unburnable materials, MSW IBA treatment is the last option before the material is lost for recycling, and a lot of progress for recovering metals from IBA was made in the last years, as the increase of recovery rates in Figure 7 as well as literature shows (Šyc et al., 2020). Nevertheless, it is hardly imaginable that even the most sophisticated IBA treatment can replace separate collection or MRFs to recover PCPW metals (Warrings & Fellner, 2019). This counts particularly for aluminum and its high oxidation rates (Biganzoli et al., 2014). Moreover, some unburnable materials, and therein particularly glass, is often not even attempted to be recovered. This is interesting since glass makes a considerable portion of IBA (Chimenos et al., 1999; del Valle-Zermeño et al., 2017; Huber et al., 2020). However, probably, it is solely the firing technology that avoids the recovery of glass even from IBA. Considering the high contents of glass in the heavy fraction from mechanical biological treatment plants in Portugal (Dias et al., 2014) and that in some countries like Austria, this fraction is often burned in fluidized bed incinerators (Bösenhofer et al., 2015; Purgar et al., 2016), it is likely that IBA from fluidized bed incineration is more suitable for glass extraction and recycling. Beside the glass-rich feedstock, these incinerators usually burn with lower temperatures (Lecker & Lind, 2020) and furthermore, they discharge the IBA in dry form. Dry discharge also means that valuable materials like glass or metals are not present in clusters by sticky, wet fine particles of ash, making it easier to extract these materials by automatic sorting processes (Šyc et al., 2020). This makes it inevitable to investigate and consider not only the recovery potential of glass, but also of easily oxidizing metals (aluminum) and even the mineral fraction of different incineration technologies. The example of fluidized bed incinerators also shows that circular-based MSW management system need to be designed from a systems perspective. One argument against fluidized bed incineration were the high costs associated to the provision a mixed MSW splitting plant upstream of MSWI plant (Leckner & Lind, 2020). If an MRF for mixed MSW is needed anyway in order to comply with the recycling targets for plastics or paper, as presented and discussed in Section 3.2, however, it can also serve as such a splitting plant. Thus, from a systems perspective, solutions which were unfeasible in the past, might be beneficial in future, in order to establish a recycling-based circular economy, as shown in Figure 8.

4. CONCLUSIONS

The negative consequences of the life style of modern societies requires a mammoth project in order to provide a sustainable development for present and future generations, and the circular economy package of the EU is exactly the kind of project that might bring societies a step further into the right direction. Waste management can contribute a lot to achieve the objective of the circular economy package, but only if all potentials at each step of a waste management system are fully exploited. Both, societal and technological innovations to do so are available. These must be tested in different situations in order to implement them successfully, for the achievement of circular and sustainable society.

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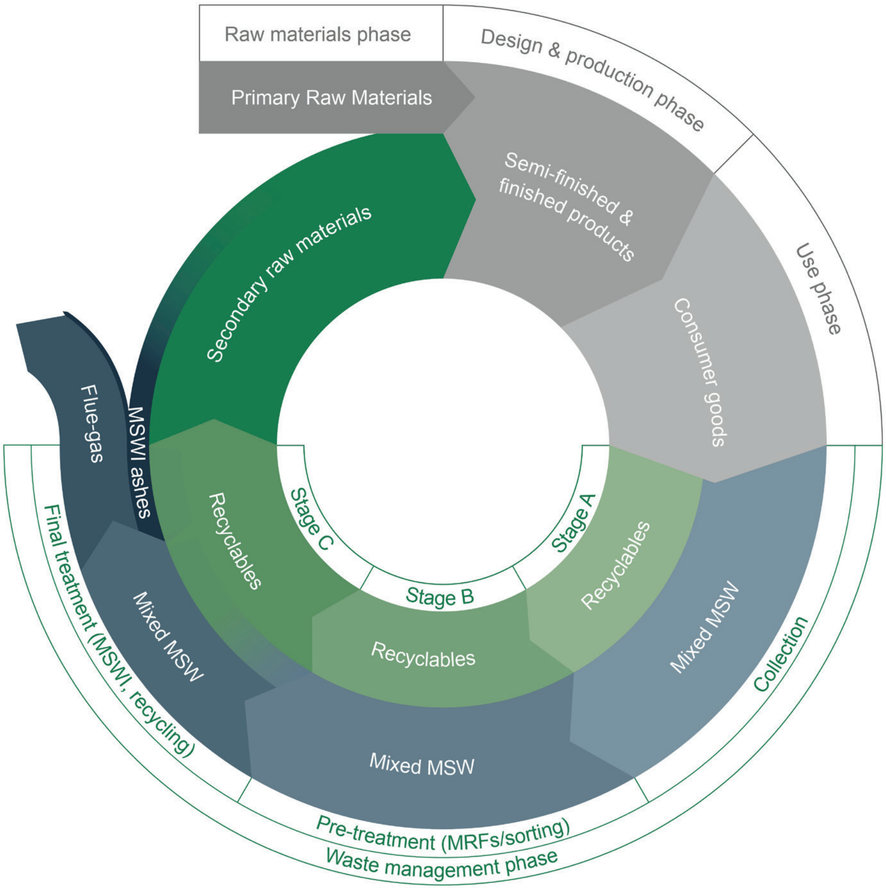

FIGURE 8: The life-cycle of a product in a more circular economy, based on enhanced municipal solid waste recycling. Design by solo-ohne© (https://solo-ohne.com/).

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EXTENDED PRODUCER RESPONSIBILITY (EPR) AND PACKAGING REGULATIONS IN ARGENTINA: REFLECTIONS ON THE ASPECTS ASSOCIATED WITH THE BLOCKING OF THE DRAFT LEGISLATION INITIATIVES

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ABSTRACT

For more than two decades there has been an on-going debate in Argentina about the draft bills presented to congress for the management of packaging waste and the notion of extended producer responsibility. This article analyses the points of agreement and controversies among the stakeholders with respect to their approach to the debate. The analysis essentially covers the discussions around three key issues related to a proposed packaging waste management system: the obliged subjects liable to make contributions, the state or private nature of the entity responsible for managing such system, and the role of the waste pickers and their organizations. A number of secondary sources were consulted (legislative files, dossiers, public records and bibliography), and several interviews were conducted with a number of key actors (specialists in this field, legislative advisers, waste picker's referents, lawmakers, public servants and technical experts) for the writing of this paper.

1. INTRODUCTION

1.1 Who is financing the cost of waste management?

In Argentina, the services of waste collection and street sweeping and cleaning (categorized as "urban hygiene") represent a large share of local government's resources. Studies carried out in different localities at different times show variations of up to 30% of municipal budgets (Gutiérrez, 2015; Pírez y Gamallo, 1994; Schejtman e Irurita, 2012). To partially fund these expenses, citizens are charged with a municipal fee commonly known as ABL (Lighting, Sweeping and Cleaning; "ABL" is the acronym in Spanish) . The calculation of the fee is based on the valuation of the buildings, which mainly takes into account the property's built up and land areas in square metres. Some authors question the criteria used for the calculation of the ABL fee, arguing that it should have a closer relationship with the amount of waste generated rather than the fiscal valuation of the household or commercial property. For example, Herrero contends that "the payment of the service concerning the size of the buildings must be adjusted by taking into account other elements that reflect more accurately the real generation of waste, especially some activities such as retail and other activities, that may simply appear to generate domestic solid waste" (Herrero, 2003). Following this rationale, several jurisdictions across the country have recently introduced legislation that established a distinction between taxpayers according to the volume of waste generated by them (1 tonne of waste per month is the threshold to surpass to become a "big generator") and their legal status or activity type. Those taxpayers who qualify as "big generators" are excluded from the regular waste collection service funded by the ABL fee, and are required to hire specific and certified waste collection and treatment contractors (Sarandón y Schamber, 2019).

Even if the criteria to establish fees is changed, from the valuation of buildings to the volume of waste produced by each generator, the issue is still stuck on the idea that the generators are liable to pay a fee for waste management, in other words, the consumers that generate leftovers after consuming products are the ones charged.

This way, the key actors that produce and supply the packaged goods to the market, typically known as producers or packagers (P/P), continue to not assume any re-

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sponsibility for waste or recycling management. The waste management costs of the products that P/P put on the market are beyond their scope. Their responsibility ends at the retailers' shelves, with the quality reassurance that the goods delivered are in good condition and before the expiring date. Once the consumer takes a product from the shelf and pays for it, the responsibility of the Producer/Packager (P/P) ceases, being relieved of what happens with the packaging waste after products are consumed.

The idea that evolves from this position is known as Extended Producer Responsibility (EPR), which understands that the liability of the P/P doesn't expire after the commercial transaction, but is extended after the product is consumed. The EPR perspective "sustains that the producer or importer must take charge of the environmental impact of the product throughout its life cycle, from the extraction of raw materials, distribution, usage, treatment and final disposition. It is argued that a producer generates an environmental risk when choosing materials or processes that generate pollution instead of using other options that could have a lower environmental impact" (Hernández Vidal, 2015). Precisely, EPR implies the acknowledgement of an economic, social and environmental externality, and justifies the establishment of regulatory mechanisms to assume the costs. As said by other authors "extending the environmental responsibility of the products to the post-consumed stage, works as a market-driven instrument that provides incentives to the companies that produce goods with 'deferred' impact, to take initiatives to mitigate environmental externalities, internalizing costs" (Matteri y Nassi n/d).

The alternative approach that engages P/P with the funding of packaging waste management (PWM) has been implemented for many years in different parts of the world, and in fact, in Argentina it has been the subject of debate for almost two decades. For instance, in 2004, in the Centro Cultural San Martín (City of Buenos Aires), a minimum standard law project for the management of packaging waste was presented, like no other time, with important support from the public and private sectors. The project was drafted by an ad hoc commission formed within the "National Packaging Waste Workshop" organized by the Secretariat of Tourism, Production and Sustainable Development (City of Buenos Aires), Secretariat of Environmental Policy (Province of Buenos Aires) and the Secretariat of Environment and Sustainable Development (National) which took place on September 2003. The workshop had the explicit support from the Coordinating committee of Industrial Food Producers (COPAL), Industrial Association of Personal Hygiene and Household Products (ALPHA), Argentine Chamber of Cosmetic and Perfumery Industry (CAPA), (Argentine Chamber of Aerosol Containers (CADEA), (Industrial Brands Association (ADIM) and Inter American Sanitary Engineering Association (AIDIS) in representation of various NGOs. In its rationale, the project acknowledged as a main objective, the complementation of the recently sanctioned Domestic Solid Waste bill (Congreso Nacional Ley Nº 25.916/04), and subscribed to the EPR principle through new instruments of private management and public control and approval. Specifically, the proposed law bill introduced two of the three key ideas discussed throughout this article: a definition of the obliged subjects and their responsibilities, and the implementation of a management system for packaging waste.

1.2 Packaging waste law projects

In fact draft legislation aiming at transferring the responsibility from the consumers to the producers/packagers (P/P) are referred to as Packaging Waste Laws. These regulations are based on environmental foundations, since they take into account the internalized costs of the materials used to produce each pack or container. In other words, the proposed laws promote what is recognized as eco-design, lessening the burden for P/P that include environmental features in the design of their products, defining lower fee rates for packaging materials that are lighter, long-lasting and recyclable, or if they contain post-consumer recycled materials. Looking beyond the environmental issue, those projects can also be classified as financial initiatives, since they propose a substantial change to the subjects who will be responsible for funding the PWM systems. The responsibility of the local authorities for financing these systems (through the charging the ABL fee to consumers), in the proposed legislation will move upstream to the P/Ps, the originary generators of the environmental risks, who have effective control of their supply and distribution chains. P/Ps define their product's design, materials and prices, therefore they have the responsibility for the environmental impact caused by the products they produce along their lifecycle.

In addition to the environmental and financial dimensions of the proposed packaging waste law bills, a social dimension must be included, as it is a key aspect of the local discussion in Argentina. The proposed law bills would have an impact on the role of the waste pickers (commonly known as "cartoneros", which describes a person collecting discarded cardboard) in the formal or informal circuits, to a greater or lesser extent, whether by act or omission.

In fact, in Argentina, the debates around EPR started in the Autonomous City of Buenos Aires (CABA) in 2002 with the recognition of the waste pickers following the approval of law 992/02 (enacted on 12/12/2002. B.O. Nº 1619). This bill acknowledged waste pickers as key players in the domestic solid waste management, and promoted their inclusion in the official differentiated collection schemes, bolstering training and health programs, aiming at the improvement of working conditions, as well as their relationship with the local community. Discussing here, the massive impact waste pickers had since the start of 21st century in the main cities of Argentina, with regards to the discussions about the characteristics that waste management should have, would imply shifting the focus of this article, and deserves a separate work. Nevertheless, in order to have an idea about its importance, it is worth highlighting a number of academic papers that focused on the "waste pickers phenomena". The following postgraduate theses (some of them published as books) worth to mention are: Álvarez, 2011; Busso, 2004; Gorbán, 2005 y 2014; Dimarco, 2010; Gurrieri Castillo, 2020; Maldovan Bonelli, 2014; Molina, 2017; Paiva, 2008; Perelman, 2010; Portugheis, 2020;

Schamber, 2008; Shammah, 2009; Sorroche, 2016; Suárez, 2003 y 2016; Tagliafico 2021, Villanova, 2015.

In that sense, the creation of a new government department dedicated to deal with waste pickers, not only raised questions about who was responsible for paying these new expenses , but the empowerment of this key actor highlighted the local nuances within the controversies generated by packaging waste regulations [the same appears to happen with the term "circular economy", another idea of recent global emergence that affects waste management and develops new attributes in this context (Gutberlet, et.al, 2017; Rutkowski, 2020)]. As detailed below, the role of waste pickers (and their organizations) would have in the implementation of a PWM bill occupy a prominent place in the discussions in Argentina.

It should be specified that the proposed regulations invariably define two instruments or mechanisms that P/P can use for PWM, while a few projects propose in addition a third option. The first mechanism exists, and in fact is commonly used and known as Deposit Return Schemes (DRS), which is an old system that covers a small share of the packaging universe, limited to the beer industry and some well-known brands of carbonated drinks. The mechanism takes into account the selling price of the product plus an additional sum of money as a deposit for the container. When returning the empty container to the retailer, the consumer receives the deposit back. Although major players in the industry are considering again the implementation of this system - known in Spanish as "envases retornables" (Donato, 2019; Sanguinetti, 2018), the historical trend is to avoid this type of scheme for the high logistics costs involved, they argue.

An alternative option is known as Integrated Management System (IMS), which, according to the law projects presented, is assigned to a specific territory or jurisdiction and is the mechanism to which all P/P are required to adhere with the exception of those who have in place DRS for their containers and packaging. Each IMS is responsible for the collection and storage of packaging waste, which is then delivered to either the respective P/P for re-utilization, or to the recycling industry or authorized waste pickers. This scheme is funded by the participant P/Ps, each contributing according to the type and quantity of packaging put on the national market. Likewise, other actors of the value chain that do not sell their products directly to the consumers, would act as advance payment agents (we will expand on this idea below) and are required to submit the funds collected to the IMS. The additional costs that the local governments incur in waste management (packaging waste containers, sorting plants, personnel, voluntary collection points, etcetera) will be financed by the IMS, after subtracting the savings produced by the reduction in the final disposal of packaging waste in a particular territory. Considering that the IMS would be responsible for the administration of the funds, its governance model is the axis of controversies and disputes between the private and state sectors. To execute the role of IMS, most projects propose the creation of a private non-profit consortium. Conversely, other projects put forward a state-run, decentralized and collegiate body to act as an IMS, composed of members of the Executive and Legislative branches, the waste pickers cooperatives and the private sector.

As an alternative to DRS and IMS, there are projects that contemplate in addition a third mechanism, known as Best Available Practices for Waste Management (BAPWM). Even though its characteristics are not specified, this alternative needs to prove its efficacy and its suitability to meet the objectives and goals that the regulation establishes as basic principles. However, beyond this clarification, some actors understood that this alternative could include the waste valorization and surreptitiously the incineration of domestic recyclable solid waste. For that reason they requested that the objective of the law explicitly excludes incineration techniques for waste materials that have other viable valorization options.

Graphically, DRS, IMS and BAPWM schemes can be shown in Figure 1.

1.3 Glossary and acronyms

- BAPWM: Best Available Practices for Waste Management
- DRS: Deposit Return Schemes
- EPR: Extended Producer Responsibility
- IMS: Integrated Management System
- IMSSI: Integrated Management System with Social Inclusion

FIGURE 1: Different schemes for Packaging Waste Management.

- P/P: Producers/Packagers (Manufacturing companies that produce packaged goods)
- PWM: Packaging Waste management

2. METHODOLOGY

This article aims at the systematization of the key aspects of the above discussion, evaluating the main features of the different waste packaging law projects presented to the National Congress, for which a number of secondary information sources were consulted (in addition to the most relevant law projects, several documents were analysed including technical reports published by public entities and NGOs, public records and bibliography related to this subject). Visiting the official website of the National Congress became an essential source of information, as its search engine allows access to all files recording the law projects presented in both the upper and lower houses.

As for the primary sources of information, various interviews with key actors in this area were conducted including: environmental lawyers specialising in this subject, technical experts, executives from the plastics industry, legislative advisors that have drafted the law projects, representatives of three waste pickers' cooperatives, members of parliament and civil servants and technical staff that have occupied positions in the environmental areas of the national government of different administrations. The majority of the interviews were conducted face-to-face, and only a few were done through the phone. The method of recording responses was in writing, and on a few occasions the interviews were recorded in audio. The analysis of the data obtained from the primary and secondary sources was based on our reading and interpretation, and digital tools were only used for the development of Table 1.

In our investigation, the intention was to identify the most controversial and antagonistic positions among key stakeholders. Thus we could understand that, even though there is a high consensus that packaging waste management is an activity that should no longer be a State's responsibility (through taxpayers' contributions), and becomes instead an internalized cost of the parties responsible for putting packaged products in the market, prejudices and mistrust prevail among the different actors, with regards to how this transition would be organized and the fundamental features that the management systems would have. Perhaps this article could be a contribution for those non-profit institutions, equidistant from the conflicting interests at stake, who have the technical expertise and the organizational experience, to bring positions closer in order to facilitate the much-needed transformation that would definitely translate into environmental benefits with social integration (Rutkowski, 2020).

3. RESULTS

We have identified that since 1999 there are records of packaging waste projects presented at the National Congress (Exp. D 1843/99 del diputado Francisco García, Exp. D 3017/02 diputada Graciela Gastañaga, Exp. 7646/02 diputado Daniel Esain). However, the projects that with their respective variations and nuances are similar to the model shown in Figure 1, including the DRS and IMS schemes, started to be introduced just days before the sanction of the Minimum Standards of Environmental Protection for Domestic Waste Management Law, N° 25.916 (year 2004). Since then, no less than 20 projects have been presented to the National Congress to date (through either the Upper or Lower House) for the sanctioning of a bill related to the issue of packaging waste.

Table 1 shows the number of law projects for packaging waste management presented to the National Congress, according to the year of presentation and the corresponding chamber where the project was introduced.

For this analysis, we have searched the following relevant keywords: “packaging”, “containers” and “waste”. We have selected only those projects that were relevant to the management of packaging waste from domestic sources, that is packaging and/or containers of FMCG (fast-moving consumer goods). We have not considered other projects that have a focus on a specific packaging type (e.g. “PET bottles”, “compostable packaging”), neither we have considered projects that have a broader scope (e.g. “Extended Producer Responsibility”, “Circular Economy”). Although the table counts projects that have a unique file ID, it is complex to establish a precise number of unique proposals, since it is common practice to present again the same project by another author, a tactic used to maintain the project´s parliamentary status.

A sharp observer of this long process, a former advisor of an institution that promotes the packaging industry, described during an interview the start point of the succes-

TABLE 1: Law projects for Packaging Waste presented to the National Congress by year and house.

Year of presentation	National Congress House		
	Lower	**Upper**	**Total**
2002	1		1
2004	2	1	3
2006	1		1
2007		1	1
2008	1		1
2009	2		2
2010	2		2
2011	2	2	4
2012	1	2	3
2013	3	1	4
2014	1	1	2
2015	2		2
2016	3	2	5
2018	4	2	6
2019	2	1	3
2020	1	3	4
2021	3		3
Totals	**31**	**16**	**47**

sion of presentations, and highlighted a significant change in the contents of those law projects from the year 2014. In his own words he stated:

> *"Packaging waste laws (projects) were adopted from overseas. It was observed that the main countries started to enforce them, and vanguardist legislators took the idea. They were gestures with little probability of success, the initiative of 2 or 3 lawmakers with good intentions and well informed, but with not enough real support. They were environmentalists or linked to an NGO that approached them for this purpose. The proposals reached the core of political power but did not prosper. You could see that some legislators said 'yes', but then they would vote against the law. Someone helped them to change their decision, or they changed their minds for whatever other reasons, and the law projects didn't come through. You could see certain arrogance and outrageous self-sufficiency from the companies that had to 'put up the dough', and hear them saying 'this will not succeed, we've got everything sorted out, this is not happening…"*

It should be noted that these types of regulations have been imposed on the countries within the EU with the sanctioning of Directive 2008/98/CE by the European Parliament and European Council on November 19th 2008. More recently it was established that "a definition of extended producer responsibility (EPR) regime must be introduced to clarify the meaning of numerous measures adopted by the member states, in order to require to manufacturers the compliance of their financial and management responsibilities, with regards to the waste phase in the life cycle of a product, including the activities of collection, sorting and treatment of the materials. This obligation can as well include the organizational responsibility and the duty to promote waste reduction and recycling and reuse of materials. Manufacturers can comply with the obligations of the EPR regime in a collective or individual way." (ECD 851, 2018)

The same expert mentioned above quoted:

> *"There were more laws (projects) coming through, and more frequently. They were no longer legislators from the periphery. There was volume. And when Capitanich was around, we saw for the first time a law initiative propelled by the Secretary of the Environment, part of the Executive branch, under Capitanich authority. They hired as an advisor a young specialist in the subject who did a very good job, changed the paradigm and takes the notion of EPR seriously. Different sectors were convened, and things started to move."*

The change of the national administration in December 2015 [12] didn't affect the willingness of the Executive branch to promote a law project similar to the one presented during the last part of the previous government, although on this occasion it was sponsored by the Ministry of Production rather than by the environmental area. Other people interviewed and supporting bibliography highlighted that such continuity is related to the international agenda of the new administration, given that the national government started a number of negotiations and proceedings to increase the participation of Argentina in the Organization for Economic Co-operation and Development (OECD) (Carciofi, 2017). Another author clearly states: "Today EPR can become a reality thanks to the intention of the National Government to be part of OECD… the political interest that we will soon see, the enforcement of the EPR principle, will come because of the interest of Argentina to become part of the selective group of OECD countries, and this goes beyond the projects already presented by some legislators on their own right. And in this case, being part of it (OECD) has its obligations. In this sense, perhaps we have to get used to the idea that the design and execution of environmental policies would only be possible thanks to the pressure of international organizations or the market itself, with the pros and cons that this entails." (Testa, 2017)

Nevertheless, the project presented on this occasion (Expediente 6375-D-2016) was not approved either, even considering the exceptional case that it was presented by the Executive Branch, and had the support from various sectors. Although legal experts would probably find many more differences, from our perspective there are three key issues that provoked antagonistic positions along the process, which made it difficult to approve a national packaging waste bill. These are related to a) the type of agents identified as obliged subject which generates controversies among P/Ps, b) the legal form or structure of the integrated management system (IMS), for which the P/Ps homogeneously propose a "trust fund", in opposition to some legislators that propose a state run system, and c) an additional element that became more relevant in the last few years, which is related to the role of waste pickers and the empowerment of their organizations, clear evidence of that being the appointment of some of their leaders in relevant positions in the both, the National and Buenos Aires province administrations, as well as the first seat won by a waste picker in the National Congress. It should also be pointed out that are in favour of a state-run integrated management system (IMS). These issues are further analysed below.

4. DISCUSSIONS

The large amount of law projects for packaging waste presented to the National Congress highlights the high level of consensus that exists among stakeholders that the management of packaging waste may cease to be a burden for the state (partly financed by consumers) and becomes an internalized cost for the P/P, who are the ones responsible for placing packaging containers in the market. Although this state of affairs is promising, there are not yet wide agreements on how this transition would be organized, or the essential features that the new system would have.

The most relevant controversies that have been identified among the different projects, revolve around three key issues: the obliged subjects and the advance payment of contributions, the state or private nature of the integrated management system (IMS) and the role of waste pickers and their organizations.

4.1 The obliged subjects

Independently of the way that they are called in the different projects ("economic agents", "producers", "packagers" which in this paper we refer to as "producers/packagers" or "P/P") and the fact that the same denomination could not include the same kind of actor, or could not define them with the same level of detail, all projects identify a certain type of "obliged subject". The clarity of the definition is of the utmost importance as it dictates who would become liable for the packaging waste management costs. Even though there is a lack of precision in the criteria to categorize the different actors, there is a substantial difference among the different projects regarding the suppliers/producers of raw materials for packaging production. There are some projects that not only assign responsibility to the producers/packagers (P/P), but also go upstream in the production chain, pretending that the scope of EPR also includes the suppliers/producers of raw materials for packaging. These inclusions are highly relevant because they would become "advance payment agents" of the contributions that the P/P must do.

Some of the people interviewed justified that inclusion for operative reasons, arguing that the collection of money contributions would be simplified, considering that the suppliers/producers of packaging raw materials are a small and concentrated group of companies, in contrast with a heterogeneous and wide universe of P/Ps. However, others considered that the main reason to propose the inclusion of suppliers/producers of packaging raw materials as advance payment agents, was promoted exclusively by the representatives of large producers of bottled carbonated drinks, grouped in a powerful industrial chamber. This industry organization considers that the State is conscious of the high informality that exists in this sector, meaning that the State tolerates that some players operate outside the supervision of the tax authorities, , and therefore they would not make the contributions for each packaging or bottle placed on the market (Bazzan, 2017). From that perspective, the imposition of a new financial contribution for the formal P/Ps would only make things better for the informal P/Ps, or at least for the part of their business channelled through the black market, harming those brands that are compliant with the tax laws, being them typically international "first" brands. Those "first" brands would also become less competitive, as they are normally marketed at higher prices, and the impact of the PWM contributions would make their prices even more expensive. With the proposed scheme of advance payment of contributions by the suppliers/producers of raw materials for packaging manufacturing, the informality issue in the P/P sector becomes less relevant, as the packaging waste management scheme would be financed regardless of the declaration of units sold by each company: the supplier companies are concentrated, abide by the law.

According to the same sources, this scheme is proposed by a specific sector of the P/Ps which in turn are also members of the large and strong industrial chambers such as UIA (Argentine Industrial Union) and COPAL (Coordinating committee of Industrial Food Producers). UIA and COPAL would only adopt a public position on this matter with a unanimous and consensual agreement of its members. One of the interviewees made the following comment:

> *"UIA makes statements only by consensus. 95% of the industrial companies are in favour of the law and that the funding should be provided by the P/Ps. But the remaining 5% are the P/Ps that are represented by COPAL say no. They maintain that although they are in favour of the law as well, the fees should be collected by the suppliers/producers of raw materials, and that would be an advance payment of contributions scheme. They say that there is an uneven playing field with regards to the tax impositions of formal and informal P/Ps, and the gap would be greater if they introduce a new contribution, and the state is consistently failing to fight tax evasion. So if the funds are collected from the raw materials industry, then there is no escape, and everyone pays regardless of operating in the formal or informal sectors. If instead the P/Ps are taxed, ARCOR will put the money, but Cuchuflito and Pindonga won't."*

The positioning of the different actors in the industry is not unanimous. Those who oppose to the advance payment of contributions scheme do so on the grounds that "a guiding principle of EPR is to assign responsibility to that entity in the supply chain that has the greatest power of influence in the design and selection of the packaging offered to the market- the Producer" (Stephenson y Faucher, 2018, p. 10). In that sense, the supplier of raw materials has no concern or responsibility over the criteria used by the P/Ps for the packaging design of their products. As expressed publicly by a manager of CAIP (Argentine Chamber of Plastics Industry) "neither the producers of raw materials nor the transforming industry (which is us), decide what type of packaging is used on each product that is put in the market. It is a corporate decision that is completely beyond our reach. The advance payment of contributions scheme does not exist anywhere in the world. It has been technically demonstrated that it is unfeasible and it would also generate a phenomenal distortion in anything to do with the raw materials' supply chain." (Tres Mandamientos, 2020; Revista Petroquímica, 2020). Nevertheless, in September 2019 during the debates originated on the "Dialogue roundtable for a federal packaging law", carried out in the context of "Packaging and Environment Sessions", an event annually sponsored by IAE (Argentine Packaging Institute), an idea was put forward that in the case of packaging made with PET (polyethylene terephthalate), a material used by the majority of the carbonated drinks bottles, it would be possible to implement an advance payment of contributions scheme. In the local productive chain, the manufacturers of this raw material are a small group of players, and the product that they generate is almost totally used for the manufacturing of bottles for carbonated drinks. In the own words of a specialist who participated in the roundtable:

> *"In the case of PET it is feasible. It is the only material that 95% of it goes to the packaging industry, whilst the remaining 5% goes to the textile industry. In addition to it, all manufacturers say 'yes', all P/P are my clients and they*

are my most important business. So now, all of us who participated in the roundtable and were against the advance payment of contributions scheme, we started to think about how it would be possible to implement it. We wrote an amendment text to the law that says: when in a given sector a consensus is reached among all participants, the tax authority could allow the producer of raw materials to pay the contributions in advance, and then submit that duty to the P/Ps when required. I pay in advance, and then when I sell you the raw materials I charge you the contributions that I have already paid on your behalf. This way 'you've got the informal players by the balls' because all of them go and buy the material from the same producer, which is just one (DAK. This is a complete novelty; the industry doesn't know it yet. There is 30% of the raw material that is imported into the country, we will have to see. This idea perhaps unlocks the limitation presented by the carbonated drinks industry, the hard core of resistance".

Even though this new idea gained some momentum, to date the heterogeneous group of P/Ps have not communicated a consensus position, and the majority block has been unable to prevail.

4.2 The administration of the integrated management systems

Each law project presented includes a proposal with respect to the entity responsible for the administration of the integrated management systems. Although this proposed entity is not defined with precision and clarity (delegating the task to the stage when a law already approved by the National Congress is regulated), for the purpose of establishing a classification of this universe, two types of entities can be established: state-run or private.

Among the projects that establish a state-run system, there are some that avoid the creation of a new, ad-hoc entity, and instead define that the enforcement authority should be the State organ in the highest hierarchy of the environmental competence as defined by the Executive branch of the government, being typically mentioned the National Secretariat or the National Ministry of Environment and Sustainable Development. Also among the same group, there are initiatives that propose the creation of new State organisms with collegiate management (members of the executive and legislative branches, the national science and technology agencies, P/P industry and waste pickers). At provincial level, their respective local authorities will be responsible for establishing the competent authorities responsible for the implementation of the law.

The projects that propose a privately-run system, the new entities would be integrated mostly by representatives of the P/Ps (companies and/or industry chambers from the packaging and raw materials sectors that have been defined previously as obliged subjects). They are defined as non-profit consortiums, in the form of "trust funds" or other legal figures that ensure that their members are not liable to the creditors or tax authorities.

Lastly, there is a group of eclectic projects that propose the creation of a new state-run decentralised entity, in addition to the enforcement authority, the State organ in the highest hierarchy of the environmental competence as defined by the Executive branch of the government. This new entity would be able to work in collaboration with the private sector, the municipalities, provincial authorities, the cooperatives or waste pickers' organisations, NGOs, universities and science and technology institutions.

4.3 The role of waste pickers and their organizations

The collective of waste pickers was not taken into account on the first versions of the projects presented to Congress until 2004, when they appear incidentally mentioned as actors to be considered, in the context of a list of actions that the competent authorities would conduct in each jurisdiction. That is reflected in wordings like "promote actions that take into account the integration of informal waste collection circuits", or "encourage the participation of waste pickers in PWM activities", or "support the inclusion of packaging waste pickers, promoting their registration and integration in the post-consumed market".

However, in 2016, a law project is submitted that represented the most vocal support for the inclusion of waste pickers in a leading role of a new PWM system, and is expressed in its first article: "establish the minimum standards for the protection of the environment for packaging waste management with the aim of reducing its impact on the environment, by applying the principle of extended product responsibility to the producer and giving precedence to the integration of waste pickers, in its different forms: individuals, work cooperatives, or any other form of social, associative ir cooperative format" (Exp. 6910-D-2016, reproduced two years later under code Exp. 3141-D-18, underlined our). This project proposes a state-run IMS [which in this case is referred as SIGIS in Spanish or Integrated Management System with Social Inclusion] through the creation of a ENAER (National Entity for the Administration of Packaging Waste), a legally autonomous entity with financial self-sufficiency. It is important to note that among the responsibilities of ENAER, it restricts the options for hiring the PWM: "at the moment of bidding and/or contracting the public service of collection and transportation of dry solid urban waste, ENAER must employ waste pickers and or the work cooperatives formed by waste pickers, properly registered…" in the National Programme for the Strengthening of the Waste Pickers, which is also created by this law project within the scope of ENAER (Art. 10°, Inc. J, underlined our).

Finally, on July 6 2021, the trade union Argentine Federation of Cardboard Waste Pickers, Cart-drivers and Recyclers (FACCYR) together with environmental organizations, organized a rally in front of the national congress to build momentum for a new law project that would be presented by like-minded legislators. A post on the union's Facebook page states "the law project, to be discussed during August 2021, seeks the implementation of an environmental fee for companies and manufacturers that place packaging goods in the market. The funds collected will be used for the implementation of a Recycling System with Social Inclusion across the whole country, that will allow the used packaging to return to the industry, dignifying the labour of waste pickers, women and men". (FACCYR, 2021)

In November 2021 the new law project was presented in the lower house of the national congress. Although the submission was done by incumbent legislators and officials from the Ministry of Environment took part on the legislative committee's meetings, the waste pickers union (FACCYR) recognized that they were responsible for writing and driving this initiative (Iglesias, 2022). In comparison to the previous proposals, this new project included novelties that can be grouped into two themes: a socio-economic theme related to who would be the main actors responsible for the operation and administration of the new IMS, and a technical-environmental-economic theme, related to technical specifications of the different packaging materials and containers as well as the different packaging waste collection systems, that would have a higher or lower impact on the P/P contributions to the IMS or being exempted from contributing al all.

In the socio-economic theme, the new project expressly mentions the key role that waste pickers currently play in the recycling chain. They would be the main beneficiaries of the proposed system, as their right to work would be recognized in addition to the investment in the infrastructure needed for the achievement of the recycling targets. With regards to the type of organization, the proposal is to create a National PWM System, a new entity with the authority to implement the programs for the strengthening of the Local Management Systems that work at municipal level, and would be responsible for the recycling of packaging waste and the re-introduction of those materials in the production chain. Additionally, it provides the creation of a Trust Fund for Inclusive Packaging Waste Management, the institution in charge of the administration of contributions from P/P and state funding. Both entities would operate under the authority, regulations and governance of the national state.

Under the technical-environmental-economic theme, the proposed legislation is quite specific, as it proposes the creation of an Environmental Fee to be paid by P/P, in relation to the type and quantity of packaging material that they submit to the market in a given time frame. Annex 1 of the proposed bill defines a polynomial formula for the calculation of the fee, which evaluates a number of technical features of the packaging such as: recyclability, environmental impact, percentage of post-consumer recycled material and eco-design. The objective of the formula is to reward with a lower fee those containers and packaging that minimize environmental impact and guarantee circularity. In addition to these considerations related to the shape and structure of the packaging, the proposed bill also provides a strong incentive for the implementation or the continuity of DRS by the P/P. These schemes should not pay any fee to the IMS, which clearly indicates that the proposed bill promotes the prevention of waste generation as an environmental strategy in addition to "inclusive recycling". DRS are the only alternative systems that are allowed to run alongside the IMS. Therefore, the new law will not allow P/P to establish their own packaging waste management systems in competition with the IMS.

The public debate around this new bill proposal created two different views. On one side, the legislators from the opposition parties and some of the most relevant industry chambers (AmCham, UIA, CADIBSA) strongly objected to the new project arguing that the Environmental Fee would be in fact a new tax, increasing the prices of the products, impacting negatively in consumer´s budgets, and increasing the already high tax burden that P/P face in Argentina nowadays. They have also questioned the lack of participation of the P/P or the private sector in the proposed IMS, and the lack of transparency, which could not ensure that the contributions made to the trust fund would be used for the specific objectives of increasing the recycling rate, and rather be used for "bolstering the state coffers for political purposes" In favour of the proposed bill were the plastics industry associations (CAIP, CAIRPLAS). (Infobae, 2021) In this opportunity, and unlike what happened with previous projects, there were no voices proposing or questioning the idea of advance payment of contributions by the producers of raw materials for packaging, for which we could assume is no longer an issue for the industry in general.

Up to April 2022, the proposed new law failed to get approval from the National Congress.

5. CONCLUSIONS

Whether the current debate has been originated after the social and economic crisis of 2001 in Argentina, or the domestic repercussions of a global trend for a transition to a circular economy, the definition to be made should take into account the particular characteristics of the local context, mainly the key role that waste pickers play today in packaging waste management. In contrast, it is worth pointing out that the context in which the EPR initiatives were implemented in Europe, more than two decades ago, were characterized by having a number of elements that are not easily perceived to be present in the local Argentine reality.

The proposals for advance payment of contributions by the producers of raw materials appear to be losing momentum, clearly positioning the vast and heterogeneous P/P sector as the obliged subjects of a future law.

With regards to the Integrated Management Systems (IMS), the differences among the law projects presented are related to the governance and ownership structure of the proposed entity: private, state-run or mixed. The options that propose a state-run scheme estimate that such an approach would imply significant administrative savings as it would use the current existing structure within the local governments. Those who oppose this approach suspect that it would have the propensity to attend to the urges of the local governments, and the new funds would be used for any purposes different than environmental protection. They argue that there are advantages for the collection of funds and administration through a private entity, given that such a structure would be more focused on its specific goals and further away from the other urgent needs of the community.

Another controversial aspect is the role of the waste pickers' organizations in management packaging waste. In any case, the activities of the current actors in the recycling chain (waste pickers, P/P, intermediaries) that operate in the informal sector and whose activities are not registered, should be addressed. The objective of social inclusion, together with the formalization of the actors of the recycling value chain, should not be an obstacle to the achievement of recycling rate and efficiency targets. There are no doubts that regardless of the type of IMS defined by future regulation, waste pickers will have a key role within the new entity.

A packaging waste law does not only imply the concept of extended producer responsibility (EPR). Even if it is a regulation that proposes a substantial change in the way recycling is financed, it is also an instrument for environmental protection and social inclusion. By promoting eco-design and giving incentives for collection and recycling, there is a positive effect in the reduction of landfills and open waste dumps. At the same time, it improves the working and living conditions of waste pickers and intermediaries that are already providing a service without additional incentives, a fact that strengthens the unavoidable social dimension of this type of legislation in this part of the world. In that sense, the waste pickers collective (in alliance with the incumbent national administration) has been more proactive and specific than the industrial sector by proposing regulations that give incentives to the inclusion of post consume recycling materials in new packaging, and punish those that have a bigger environmental impact.

The transfer of the contribution responsibility from the consumer to the producers/packagers is undoubtedly an opportunity for law proposals that imply environmental protection with social integration.

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NAIL VARNISH PACKAGING AND RESIDUE WASTES: PROBLEMS AND SOLUTIONS

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ABSTRACT

As global consumption of nail varnish grows, quantities of packaging waste and residues will increase. End-of-use decisions for nail varnish have implications. Residues contain some hazardous substances, whilst packaging has potential for valorization. The extent of problems related to the disposal of end-of-use nail varnish products remains unclear. This study aimed to gain evidence to inform potential management measures. An inventory of hazardous substances in nail varnish products available in the UK was compiled to ascertain their potential for harm to the environment and/or human health. Discarded samples of end-of-use nail varnish products were then collected from volunteers in the UK to determine weights of packaging materials and residues. A parallel survey explored UK consumers' means of disposal and opportunities for positive change. All the brands examined contained at least one hazardous substance; over a hundred hazardous substances were identified in total for the 24 samples audited. Hazards to human health were most common. On average, 7.2g of nail varnish remained in bottles at the point of disposal. Average weights for glass bottles and plastic lids/applicators were 28.5g and 5.6g, respectively. End-of-use products thus generate small quantities of residues and packaging, but the cumulative impacts are potentially substantial. Disposal of end-of-use products with general household waste was evident: this leads to contamination of non-hazardous waste and fails to valorize the materials lost. Collection schemes for end-of-use nail varnish offer a possible way forward, but would benefit from fuller understanding of the potential for consumer engagement.

1. INTRODUCTION

1.1 Cosmetic consumer products

Cosmetics are ubiquitous and utilized every day across the globe for hygiene and beauty. Article 2 of Regulation No. 1223/2009 on cosmetic products (European Commission, 2009) defines them as, 'any substance or mixture intended to be placed in contact with various external parts of the human body…or with the teeth and the mucous membranes of the oral cavity with a view to exclusively or mainly to cleaning them, perfuming them, changing their appearance and/or correcting body odours and/or protecting them or keeping them in good condition' (European Commission, 2009).

Makeup, a sub-group of cosmetics, became popularised in the early 20th century through theatre, ballet and Hollywood (Chaudhri & Jain, 2009). Originally, cosmetic products were predominately available to wealthier members of society, but chemical and business innovation resulted in an increased accessibility for the mass market consumer within Europe and USA (Butler, 2000). There is a growing emerging market in economically-developing countries (Chaudhri & Jain, 2009). Inevitably, the observed increase in consumption of cosmetics is accompanied by increased consumption of resources and generation of waste, with associated adverse impacts upon the environment.

Ensuring cosmetic products are safe for consumer health has long been a priority amongst legislators and traders. Regulation No. 1223/2009 on cosmetic products (European Commission, 2009) bans specified substances from products and ensures product safety checks. However, the environmental impact of cosmetics has, to an extent, been overlooked despite greater awareness in recent years, evidenced by the banning of micro-plastics in popular cosmetic products across Europe (DEFRA, 2018). Across the wide range of cosmetics products, those used for the enhancement of finger and toe nails are of concern due to their potential impacts on the environment.

1.2 Nail varnish

Nail varnish, otherwise known as nail lacquer or nail polish, is a popular cosmetic product applied to nails to deco-

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rate them and, in some cases, protect nails (Draelos, 2016). Decorating nails can be traced to as early as 10,000 BC in Ancient Egypt, in which henna or a clay (red ochre) mixed with water were utilized to dye fingernails red, orange or yellow (Chaudhri and Jain, 2009). Similarly, the practice of staining fingernails using gum Arabic, gelatine, beeswax and egg, was present within China in approximately 3000 BC. Despite this long history, the use of products that would be recognised as nail varnish in modern society were not available until the 1920s, and originated from early nitrocellulose-based car paint technology (Draelos, 2016).

Within the last decade the consumption of nail varnish products has grown globally, with the market size reaching US$ 9190 million in 2018, overtaking sales of lipstick to become an economic indicator (Kenton, 2018; Trend, 2019). This trend is predicted to continue, with a compound annual growth rate (CAGR) of 8.5% (Sun et al., 2015). The observed increase in the use of nail varnish products can be attributed to multiple factors, such as increased availability, better quality products coupled with affordable prices and increased disposable income (Kestenbaum, 2017). As a consequence of increased nail varnish purchasing, the volume of nail varnish waste, including the liquid residues and accompanying packaging, has and will continue to increase. Therefore, it is important to understand if, and what, the environmental implications of this waste are and how to manage them.

1.3 Potential impacts of nail varnish disposal

The formulation of nail varnish varies between products, however the overarching composition is typically 70-80% volatile solvent, 15% nitrocellulose and differing quantities of a diverse range of other substances (Trend, 2019). Some of the ingredients contained within nail varnish products are classified as hazardous substances, i.e. have "… the potential to cause harm to humans, animals and the environment" (Institute of Hazardous Material Management, 2004).

The presence of some hazardous substances, such as toluene and formaldehyde, has undergone scrutiny (Quach, 2015; Sainio et al., 2006; Yokota et al., 2007). Attention thus drawn to the presence of such substances in nail varnish products has resulted in (1) toluene and formaldehyde being omitted from these products and (2) customers avoiding products containing these substances. However, nail varnish may still contain hazardous substances and many consumers may be unaware of this situation. Furthermore, many nail varnish bottles do not exhibit hazard symbols or information to inform consumers of a product's hazardous nature. Consumers themselves thus have to check a product's material safety data sheet for information, which can be hard to obtain (Environment Agency, 2014a). This is particularly problematic when consumers dispose of nail varnish.

Nail varnish products can be classified as hazardous waste or hazardous household waste if they contain hazardous substances under legislation (Table 1) and therefore require special disposal with strict control measures, similar to other hazardous wastes, as described by the Hazardous Waste Directive 2008 (Table 1; European Commission, 2018a). Moreover, it is 'illegal to mix a hazardous waste with either non-hazardous or another hazardous waste' under the Hazardous Waste (England and Wales) Regulations 2005, thus incorrect disposal is an offence (Environment Agency, 2014a). Regulatory controls for hazardous substances, however, do not apply to all sectors. For example, domestic waste is excluded from controls specified by The Hazardous Waste (England and Wales) Regulations 2005 unless "it comprises asbestos waste or is collected separately." (United Kingdom Government, 2005).

Despite this situation, hazardous components of household waste are poorly understood. There is a general consensus that amounts are small, thus disposal risks can be assumed to be negligible (Slack et al., 2005). There are concerns, however, that this assumption is not supported by robust evidence (Slack et al., 2005). Furthermore, as the consumption of nail varnish and other household products increases, the quantities of hazardous substances in the household waste stream will likely increase (Inglezakis and Moustakas, 2015).

There is a lack of literature pertaining to the environmental impacts associated with hazardous waste contamination of household waste streams. However, Slack et al. (2005) suggested that hazardous substances from improperly disposed hazardous waste can lead to hazardous leachate from landfills causing water pollution, and identified a need for research to quantify contamination resulting from household waste. Research pertaining to household hazardous waste has explored identification, volumes and sources of household hazardous wastes rather than the impacts associated (e.g. Slack et al., 2004, 2009). The volume of hazardous waste will inevitably increase and understanding these wastes is important for their effective management.

In addition to concerns regarding the fate of hazardous substances in nail varnish, there are implications regarding the fate of nail varnish packaging. As "high end" products (i.e. relatively small quantities of product at relatively high price), packaging is designed partly to ensure safe containment of the product and partly by the desire for the product to be attractive to customers. The quantity of

TABLE 1: Legislation relating to hazardous substances in the UK context.

Legislation	Notes
Hazardous Waste Directive 2008/98/EC	Defines hazardous waste as waste that has substances that are " harmful to human health or the environment" (HSE, 2019)
Hazardous Waste (England and Wales) Regulations 2005	Defines hazardous waste. States "It is illegal to mix a potentially hazardous waste with either a non-hazardous waste or another hazardous waste" (EA, 2014a)

packaging relative to the quantity of the product tends to be rather high and there is potential to recycle packaging materials.

1.4 Aims of the study

In view of the background to and content of nail varnish products, this study aimed to elucidate (1) the hazardous substances contained within nail varnish, in order to gain an understanding of the hazardous substances that could arise and contaminate household waste streams, and (2) the quantities of packaging associated with end-of-use or end-of-life nail varnish products and thereby the opportunities for recycling and implications of failure to do so, and (3) consumer behaviours relating to disposal of nail varnish products with a view to identify opportunities and challenges to improve management of residues and packaging. On the basis of information acquired, the study provides commentary on means to effect positive change.

2. MATERIALS AND METHODS

2.1 Overview

Data collection was undertaken in two phases. The audit of hazardous substance in nail varnish products was carried out first. In addition to providing information in focus, this audit also informed completion of the risk assessment and Control of Substances Hazardous to Health (COSHH) assessment for the subsequent evaluation of nail varnish packaging and residues in the laboratory. A questionnaire survey was undertaken to determine consumers' disposal behaviours, as well as their receptiveness to potential management initiatives or interventions. All elements of this study involving human subjects (sections 2.3 & 2.4) received ethical approval from the University of Southampton (reference number ERGO II 47510). All data collection focused on nail varnish products available in the UK. Samples of nail varnish packaging and residues were provided by citizens living in the UK. The questionnaire survey was distributed only to UK residents.

2.2 Audit of hazardous substances in nail varnish products

In order to ascertain an indicative profile of the types of hazardous substances contained within nail varnish products and their associated hazards, an on-line audit of ingredients contained within nail varnish products was performed (February to March 2019). Boots the Chemists Ltd (trading as Boots) and Superdrug Stores plc. (trading as Superdrug) are the UK's largest and second largest pharmacy, health and beauty chains respectively, each with a wide range of self-application nail varnish products (Boots UK, 2018; Superdrug, 2019). One nail varnish product from each brand sold by these stores was selected for this audit. Lists of products for each brand sold by each store were located on the stores' websites and the first pigmented product listed for each brand on the website was selected for scrutiny. The full list of contents provided within the description of each product was used as the basis for the hazardous substances audit. Any brands in which a list of contents could not be obtained were omitted from the study.

An internet search for a Material Safety Data Sheets (MSDS) for each ingredient present in the randomly selected nail varnish for each brand was conducted. If a Material Safety Data Sheet with a hazard statement was found, the ingredient was considered a hazardous substance and recorded. The associated hazard(s), both physical and health, were recorded for each ingredient. A matrix then was constructed to map hazard statements in relation to brand. This matrix was used to determine the number of brands containing each hazardous substance, and the percentage of brands audited that contained each hazardous substance.

2.3 Packaging and nail varnish residues

A combination of opportunistic and convenience sampling was used to obtain unwanted bottles of nail varnish from individuals in Southampton and London over a five month collection period (Lavrakas, 2008) from November 2018 to March 2019. A modified triple rinse procedure, originally used to clean pesticide containers, was utilized to remove residues of nail varnish from nail varnish packaging. This method was selected as it is an accepted, widely used cleaning method and it is suitable for glass and plastic containers used for nail varnish (Miles et al., 1983). All work had to be carried out within a fume cupboard due to the presence of volatile substances within the nail varnish residues.

Each bottle of nail varnish was weighed prior to the triple rinse procedure, using a balance to three decimal place accuracy. The nail varnish bottle was inspected for any hazardous warning information and/or disposal information. All symbols and information were recorded. The volume of the product displayed on the bottle was recorded. The bottle was then inverted for one minute with the lid tightly secured. The nail varnish residues were then decanted into a lidded waste container until drips of residues were no longer observed. An aliquot of 2ml of acetone (ca. 10% of the average volume contained in nail varnish products) was pipetted into each bottle. Acetone was selected as the bottle cleaning agent because the main constituents of nail varnish are organic compounds and acetone is an effective organic solvent (Remler, 1923). Once the lid was firmly re-secured, the nail varnish bottle was shaken vigorously by hand for 30 seconds and the mixture decanted into the waste container. The presence of metal balls, which facilitate mixing of the nail varnish prior to use, was recorded. The addition of acetone in 2ml increments was repeated twice more, or until nail varnish residues were no longer evident by visual inspection. In the vast majority of cases, there were no visible residues remaining after three rinses. A paper towel was used to remove nail varnish residues from the brush applicator. If nail varnish residues remained on the brush applicator, the brush was cleaned by immersion in 5ml of acetone until residue were no longer present upon visual inspection. Once all residues of nail varnish were removed from the brush applicator, 5ml of distilled water was pipetted into the nail varnish bottle and the lid re-secured. The bottle was vigorously shaken for 30 seconds and the liquid discarded. The lid was then removed from the nail varnish bottle and the bottle was weighed. The plastic lid/brush applicator was also weighed. Although some brush bristles can be natural animal hair, all brushes within this study were assumed to be synthetic plastic fibres as nail varnish brushes produced for the mass market predominately consist of plastic bristles. The metal ball in one nail varnish was captured and weighed. Descriptive statistics for both glass and plastic packaging were calculated, and the ratio of glass to plastic packaging derived.

Volumes of nail varnish residues were obtained by first determining the density of nail varnish. A full nail varnish bottle, with a known volume, was weighed, 5ml of nail varnish was removed using a pipette, the bottle lid was re-secured and the bottle re-weighed. The residue in each bottle was used, in conjunction with the full volume, to calculate the percentage of waste nail varnish product remaining in the bottle.

2.4 Consumer questionnaire

A questionnaire was designed comprising closed and open questions to determine (1) awareness that nail varnish contains hazardous substances, (2) individuals' current disposal of nail varnish products, (3) how awareness of nail varnish's hazardous nature could change future disposal behaviours, and (4) consumers' receptiveness to potential management initiatives or interventions.

Questionnaires were completed using online methods and on a face-to-face basis. Individuals were only permitted to participate if older than 18 years, were fully capable to give consent, lived within England and used nail varnish products. Participants in the consumer questionnaire comprised a convenience sample and were recruited (1) via social media for online surveys, and (2) by approaching members of the public in Southampton city centre for face-to-face surveys. Participants recruited via social media comprised acquaintances and contacts of the researchers: participants in face-to-face interviews were all unknown to the researcher and duplicated responses are therefore unlikely. All consumer questionnaires were completed over the period March to May 2019.

Descriptive statistics were derived to profile the demographics of the population sample and responses related to awareness of nail varnishes hazardous nature, nail varnish disposal at end-of-use or end-of-life, and receptiveness to collection schemes.

TABLE 2: Hazardous substances present within the majority of nail varnish brands audited. CI indicates "colour Index" (pigment) substances (N=24).

Hazarous substances	Percentage of brands present in
Butyl acetate, Ethyl acetate, Nitrocellulose	100
Isopropyl alcohol, Trimellitic Anhydride	90-99
Acrylates Copolymer, Adipic Acid, Titanium Dioxide (CI 77891), Acetyl Tributyl Citrate, Neopentyl glycol, Yellow 5 lake (CI 19140)	80-89
Red Iron Oxide (CI 77491)	70-79
Aluminium Powder (CI 77000), Black iron oxide (CI 77499), Red 6 (CI 15850)	60-69
Benzophenone-1, Red 7 lake (CI 15850), Stearalkonium hectorite, Styrene	50-59

3. RESULTS

3.1 Hazardous substances in nail varnish products

The online hazardous substance audit was completed for samples of 24 nail varnish brands. All nail varnish brands were found to contain at least one hazardous substance and 107 hazardous substances were identified in total across all brands. The number of hazardous substances per brand ranged from 10 to 45. Several hazardous substances were noted to be present in the majority (>50%) of products sampled (Table 2).

Seven of the hazardous substances found in the majority of products (Table 2) are utilized for pigmentation and are accompanied by a five digit CI (Colour Index) code. The remaining substances are utilized within nail varnish as a solvent, suspension agent, adhesive, film former or stabilizer.

Table 3 lists the hazard statements present for all of the 24 brands of nail varnish audited. The hazards identified concern human health hazards. Hazard statements that were not present for all of the brands audited mainly concerned hazards to human health. Only one hazard to the environment was recorded: 'harmful to aquatic life with long lasting effects' was stated for over 90% of the products examined.

3.2 Packaging and nail varnish residues

Product residues and packaging (glass and plastic) were considered in relation to the volume of product as sold (Figure 1). In total, 59 end-of-use nail varnish bottles were analysed, from 28 different nail varnish brands. For

TABLE 3: Stated hazards present for all nail varnish brands audited (N=24).

Stated hazards
Above 22°C explosive vapour/air mixtures may be formed
Acute toxicity (oral, dermal, inhalation)
Combustible
Cough when inhaled
Difficulty breathing if inhaled
Dizziness if inhaled
Drowsiness if inhaled
Flammable. Headache if inhaled
Highly flammable
Irritation of eyes
Irritation of skin
May cause eye irritation
May cause skin irritation
Nasal irritation
Nausea if ingested
Nausea if inhaled
Sore throat if inhaled
Unconscious if ingested
Unconscious if inhaled
Vapour/air mixtures are explosive
Vomiting if ingested

some of the brands within this sample, more than one bottle was thus collected. Of the sample, 35 nail varnish bottles displayed a hazard warning and/or disposal information. A flammable warning symbol or written warning was present on 31 of the 59 bottles (59%). A recycling symbol was shown on only two nail varnish bottles, and a recycling symbol coupled with a 'Don't put in a general waste bin' symbol was present on only one bottle. One bottle alone had a 'Don't put in a general waste bin' symbol. No hazardous warning/symbols or disposal symbols/information were present on 24 of the 59 bottles (41%).

The mean mass of nail varnish residue within the donated nail varnish bottles was 7.2g ± 0.5g; the range of residue mass (maximum-minimum) was 14g. The mean mass of glass for glass bottles was 28.5g ± 1.1g, and the range (maximum-minimum) was 32.12g. The mean mass of plastic contained in nail varnish bottles was 5.6g ± 0.3g and the range of plastic packaging (maximum-minimum) was 8.4g. It was also noted that quantities of packaging and residues were not necessarily related to product volume. Products of 9ml, for example, were noted to have higher levels of residues than some larger products and also had relatively high weights of glass and plastic packaging (Figure 1). The ratio of residues to product volume was also highest for products of 9ml (Figure 1).

3.3 Consumer disposal behaviours and collection scheme receptiveness

The total number of respondents was 109; of these, 3 participated in the survey via face-to-face interviews and 106 took part in the on-line survey. All respondents identified themselves as female and were predominately aged 18-25 years (42%). Self-application of nail varnish products (as opposed to use of professional services) was reported by 96% of respondents; 59% of respondents stated that they were aware that nail varnish contains hazardous substances.

The number of nail varnish bottles reported to be disposed by respondents within a year ranged from 0 to 15; most respondents (55%) claimed to dispose of nail varnish once a year or less often. The majority of respondents (86%) dispose of nail varnish bottles with residues of less than 30% of the initial product volume; 29% of respondents claimed to discard bottles containing less than 10% of the original product volume and a further 29% claimed to discard bottles with less than a fifth of the original product.

At the point of disposal, most (85%) respondents appear not to check the safety or disposal information of products; only 14% do. The method of nail varnish disposal at end-of-life or end-of-use varies with the volume of residue (Figure 2).

The majority of respondents expressed an interest in participating in a nail varnish collection scheme (89%). Respondents indicated a wide range of incentives or initiatives that would potentially encourage them to participate in scheme(s) to collect nail varnish residues and packaging of which the most likely to appeal to consumers was the offer of monetary incentives in the form of discounts for further purchases (Figure 3).

4. DISCUSSION

The hazardous waste audit revealed that all nail varnish brands audited contained hazardous substances to varying degrees (Table 2), confirming that nail varnish, depending on its content, is classified as hazardous waste at the point of disposal. Due to its hazardous nature, nail varnish should in principle not therefore be mixed with other waste materials (Table 1), although the Hazardous Waste

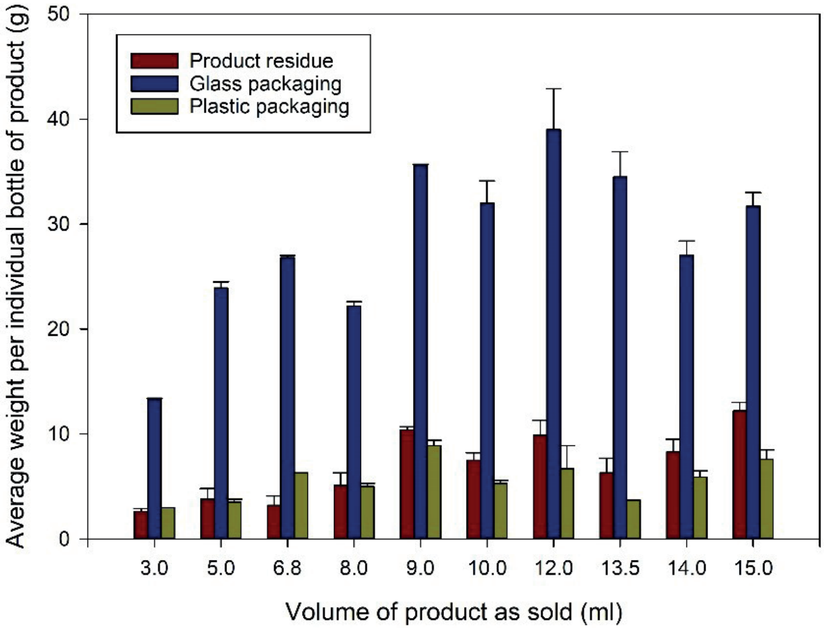

FIGURE 1: Descriptive statistics (mean ± standard error) for nail varnish end-of-use residues and packaging. Data are grouped by product volume as sold; only data for samples with three or more of the same product volume are shown. Products of 6.8ml, 9.0ml and 13.5ml each comprised samples of only one brand.

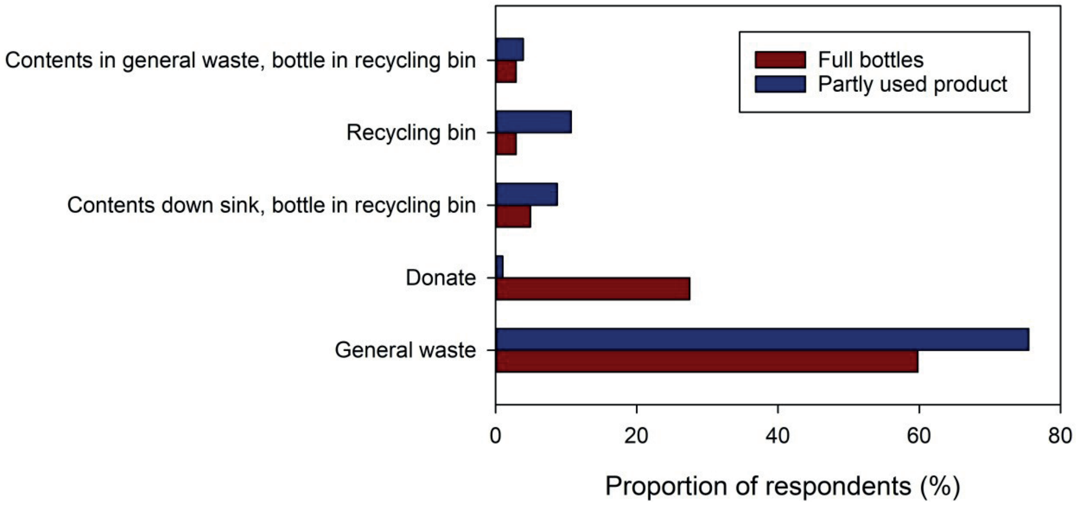

FIGURE 2: Disposal methods employed by respondents as stated in responses to the questionnaire survey. "Full bottles" refers to nail varnish products that are discarded without having been used; "partly used product" refers to nail varnish products that have been used but contain some residues. Items places in either recycling bins or discarded as general waste comprised bottles containing residues or unused product. "Donate" includes giving to known persons or organizations (N=109).

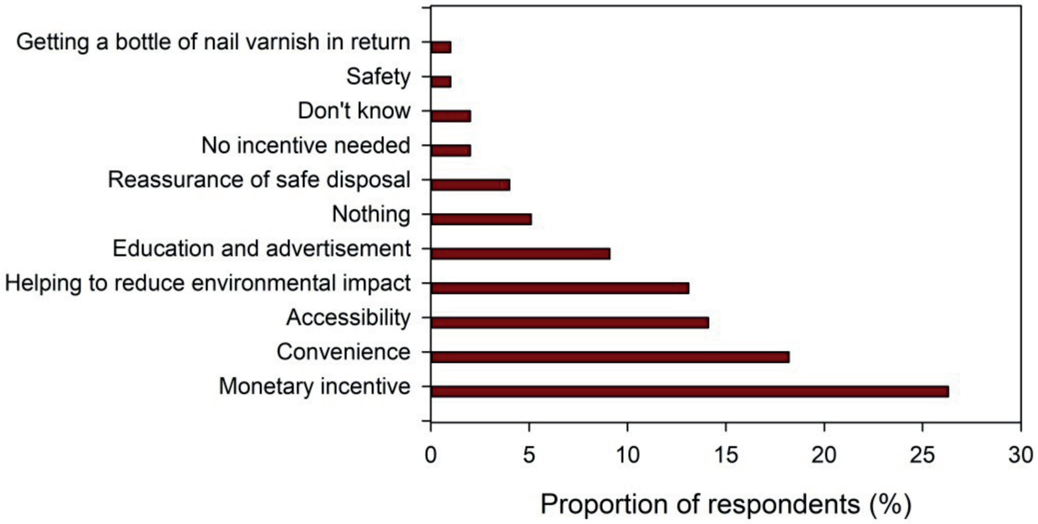

FIGURE 3: Suggestions for incentives likely to encourage engagement of consumers in nail varnish collection schemes, as identified via questionnaire surveys (N=109).

(England and Wales) Regulations 2005 (UK Government, 2005) stipulate that controls on hazardous wastes in domestic waste would not apply unless separate collection for nail varnish residues were in place. We contend that waste management authorities should ideally implement systems to capture and separate nail varnish waste from household waste to ensure safe disposal with minimal environmental impact.

The number of hazardous substances varied greatly between brands. Only three hazardous substances (Butyl acetate, Ethyl acetate and Nitrocellulose) were present within 100% of the brands audited (Table 2). Consequently, the number and type of hazardous substances that would contaminate waste varies greatly, but the highest volume associated with these will most likely be the aforementioned three substances. Most of these hazards are related to human health and fewer are associated with environmental harm. Arguably, the hazardous substances that raise the most concern for waste management coordinators, particularly with respect to contamination, are combustible, flammable and explosive substances. Fire is a common and detrimental hazardous event within waste management (Oygard et al., 2005). The presence of these substances could cause, accelerate or intensify such events, potentially causing injury, death, air, soil and water pollution, and resource consumption (Martin et al., 2016; Weichenthal et al., 2015).

Substances that are 'harmful to aquatic life with long lasting effects' are of potential concern, as hazardous substances possessing this property could accumulate in landfill leachate and potentially damage aquatic life if the leachate enters a watercourse (Salem et al., 2008). Further research into the quantities of such hazardous substances within nail varnish residues and the mean volume of nail varnish remaining in disposed bottles, in concert with knowledge of disposal, could contribute to better understanding of the risks in this regard. Product-specific information would be needed to achieve this due to the variable hazardous substance content of nail varnish products. Moreover, research into the actual volume of nail varnish needed to cause aquatic harm is also required if the possible impacts from nail varnish waste at current waste volumes are to be determined.

The low quantities of residues relative to original product volumes (Figure 1) may be interpreted as an indication that the volumes of nail varnish waste and associated risks are negligible. However, the cumulative volumes of nail varnish in waste are currently unknown. If it is assumed that the risks of hazardous substances increases with their quantity, the risk associated with hazardous substances in nail varnish remains unknown. Given the widespread use of nail varnish and its predicted increase, any risks are likely to be of concern when aggregated to population level and likely to increase. Arguably, identifying the extent of this issue is a matter of urgency, particularly in light of likely increases in use.

If a precautionary approach were to be adopted, means would be implemented to (1) avoid the placement of nail varnish residues containing hazardous substances with other household waste, and (2) capture associated packaging to facilitate recycling of glass and plastics. A possible solution would be to capture and separate nail varnish waste from households for safe treatment: establishing a nail varnish collection scheme, similar to reverse logistic collection programmes in the USA (USEPA, 2019), offers potential in this regard. However, the success of such schemes relies heavily on their economic viability. This requires high participation rates and for the collected material to have profitable market value (Pulidindi & Chakaborty, 2018; Tonglet et al., 2004).

Glass and plastic are the main component materials of nail varnish product packaging: the mean glass and plastic was found to be 3.5g and 0.65g per ml or product. The market value of recycled glass is growing, particularly clear glass of which most nail varnish bottles are composed (WRAP, 2019). Moreover, glass is considered an endlessly closed loop material as it can be recycled endlessly, but its quality does not depreciate significantly when recycled. The value of plastic packaging may be lower than glass, as the value of plastic varies with plastic resin type. The mass of plastic available to recycle is also lower than glass (Figure 1). Further analysis and research needs to be undertaken to establish the actual value of glass and plastic contained in nail varnish products, in conjunction with participation models for a collection scheme.

In tandem with systems and facilities for the collection of end-of-use nail varnish products, due consideration is needed with regard to the treatment of the residues and packaging collected. First, residues must be separated from packaging to enable recycling of glass and plastic. Given the risks to human health associated with substances in nail varnish residues (Table 3), exposure of processing operatives to residues should be minimised or avoided by the pre-treatment of contaminated packaging prior to recycling operations. Use of suitable solvents (e.g. acetone; Remler, 1923) combined with a rinsing procedure (Miles et al., 1983) could provide a suitable means to remove residues. We note that such procedures have two key weaknesses (1) manual operations of this nature (see section 2.3) are onerous and time-consuming and do not offer a viable prospect at scale, and (2) waste products in the form of a mixture of acetone, water and nail varnish are generated. Larger-scale and automated cleaning operation may be technically feasible, but the treatment of large volumes of washing residues is potentially problematic. In addition to the hazardous substances contained in nail varnishes, acetone itself is a hazardous substance and should be treated and/or disposed of as such. Waste acetone can be recycled and reused (Weires et al., 2011), but residues of nail varnish product would nonetheless remain. Landfill has been used widely as a disposal route for hazardous substances and may offer an option for nail varnish washing residues, but seepage or leakage of hazardous substances from landfills poses a risk (Slack et al. 2005). We note that in the case of nail varnish residues, the majority of hazards are to human health rather than to the environment (Table 3). Incineration – thermal destruction - of hazardous wastes arising from nail varnish residues and the cleaning of packaging might present the best opportunity for their disposal (Dempsey & Oppelt, 2015).

Ultimately, efforts to enhance recycling of glass and plastic packaging necessitate participation by consumers. Responses to the questionnaire survey indicate that engagement in collection schemes might be fostered by making available monetary incentives. Arguably, it is difficult to envisage how such schemes would operate given the often infrequent need to dispose of end-of-use or end-of-life products. Over half of the questionnaire respondents reported discarding nail varnish once a year or less often (section 3.3) and the monetary incentive would have to be commensurate with the monetary value of the product and materials. Convenience and accessibility were highlighted as important facets of collections schemes (Figure 3). Collection schemes could, for example, utilize collection facilities located in high street retail outlets. Convenient locations of this type may foster high participation rates; collection points do not have to be located at the place of purchase. Means to foster high levels of participation should recognise convenience and availability (Ongondo and Williams, 2011). Studies of nail varnish collections schemes already implemented, such as those in the USA, could be used to guide research and to design collection schemes within the UK. We recommend that researching, investing in and implementing such schemes should commence sooner rather than later, given the potential risks identified. Moreover, we propose that efforts to change consumer efforts and behaviour should consider waste avoidance as a priority in accordance with the aims and principles of the waste hierarchy (Williams, 2015). As a priority, full use of nail varnish products should be promoted to avoid generating residues. If achievable, elimination of residues (see Figure 1) would simultaneously make fuller use of the energy and resources consumed in the manufacturing, packaging and distribution of nail varnish products, whilst reducing the volume of residues requiring treatment and disposal.

Finally, we note that the hazardous nature of substances used in nail varnish (Table 3) imply that nail varnish residues potentially constitute hazardous waste. In principle, therefore, separation of nail varnish waste from other household waste streams along with safe treatment may be legally required (Table 1). Even if further research into the hazardous nature of nail varnish waste were to determine that the risks to human health and environmental risk posed by nail varnish waste are negligible and specialised treatment is neither warranted nor economically viable, there remains a need for a management system to capture resources that would otherwise be wasted (Tudor et al., 2011). The importance of recovering materials used for the packaging of products for reuse or recycling has become increasingly important, evidenced by the implementation of producer responsibility through the Packaging and Packaging Waste European Directive (94/62/EC), amended in 2015 (European Commission, 2018b; Environment Agency, 2014b). Consequently, establishing a collection scheme for end-of-use nail varnish to reuse and recycle the plastic and glass within nail varnish bottles would contribute to efforts towards circular economy approaches within the nail varnish industry.

5. CONCLUSIONS

The global consumption of nail varnish products will likely continue to grow, inevitably producing higher volumes of nail varnish waste. Due to the hazardous substances used for its production, it is likely that all nail varnish waste produced is potentially hazardous waste: waste management systems to capture and separate nail varnish waste from household wastes are required. However, there remain unknowns regarding the volume and composition of nail varnish waste generated, the hazardous substances within this nail varnish waste, and the associated risks.

In order to successfully implement an appropriate management scheme for end-of-use nail varnish residues and packaging, there is a need to understand consumer awareness regarding both the hazardous nature of substances present in nail varnish products and the presence of recyclable packaging materials. Information regarding the hazardous nature of waste, the impact of nail varnish waste and how to dispose of it correctly are of potential benefit. Accurate estimates of the volumes and type of nail varnish waste that requires disposal would also assist in efforts to establish suitable collection systems. Furthermore, implementing a nail varnish management scheme, such as a collection scheme, is not just a matter of immediate human health and environmental impacts but also sustainable consumption. Implementing a nail varnish collection scheme would facilitate the capture of resources such as plastic and glass, reducing the waste of both upstream and material resources. However, more research is required to establish the actual value of resources.

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COMPOST HEAT RECOVERY SYSTEMS: GLOBAL WARMING POTENTIAL IMPACT ESTIMATION AND COMPARISON THROUGH A LIFE CYCLE ASSESSMENT APPROACH

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ABSTRACT

Compost Heat Recovery Systems (CHRS) represent an innovative technology to provide domestic decentralized thermal energy, recovering the heat naturally produced during the aerobic biodegradation of waste biomass, coming from gardening/farming/forestry activities. CHRSs represent an alternative to centralized grid-connected power systems and are usually installed (combined with most traditional systems) to power underfloor heating systems (UHS) or domestic hot water systems (DHWS), lowering impacts and costs of thermal energy production. In this study, the Global Warming Potential (GWP) of CHRSs (measured as kg$_{CO2-eq}$/kWh) was investigated using life cycle assessment (LCA) approach, considering the whole life cycle of an average plant. CHRSs showed a negative Net value of GWP impact, equal to -0.268 kg$_{CO2-eq}$/kWh, as full balance of positive (0.062 kg$_{CO2-eq}$/kWh) and negative (-0.329 kg$_{CO2-eq}$/kWh) emissions. Negative emissions are related to avoided primary materials, replacement of natural gas used as traditional thermal energy production and replacement of mineral fertilizers. Considering only the positive emissions (0.062 kg$_{CO2-eq}$/kWh), CHRSs emerged to be in line with Solar Hot-Water Systems (0.061 kg$_{CO2-eq}$/kWh mean value) and slightly higher than Geothermal Systems (0.019 kg$_{CO2-eq}$/kWh mean value). Along with GWP impact, other midpoint and endpoint impact indicators were assessed and all showed a negative Net value: Particulate Matter PM (-2.36E-5 kg$_{PM2.5-eq}$/kWh), Fresh Water eutrophication FWE (-6.78E-06 kg$_{P-eq}$/kWh), Fresh Water ecotoxicity FWec (-2.10E-01 CTUe/kWh), Human Toxicity cancer effect HTc (-5.68E-09 CTUh/kWh), Human Toxicity non-cancer effect HTnc (-3.51E-09 CTUh/kWh) and Human Health HH (-5.22E-08 DALY/kWh). These results demonstrate that CHRS is extremely convenient considering both environmental and human health consequences.

1. INTRODUCTION

Aerobic biodegradation of biomass is an exothermic biological process able to release considerable heat, about 20,000 kJ/kg of organic matter degraded, rising the material temperature up to > 65° C (Di Maria et al., 2008; Themelis and Kim, 2002). At current level of technological and industrial development, this heat is usually dispersed into the environment. Previous studies already investigated the possibility of recovering such heat. Di Maria et al., (2008) reported the possibility of recovery from 4,000 kJ to 5,000 kJ per each kg of treated biowaste by the exploitation of a heat pump. In investigating 45 compost heat recovery systems (CHR) in 16 different areas, Smith & Aber (2017) reported the possibility of recovering about 7,000 kJ/ kg o dry matter by means of hot water for domestic purpose. First examples of CHR were documented in china sin 2000 years ago (Brown, 2014) but most recent concept of CHR were reported by Jean Pain with his book The Methods of Jean Pain: Another Kind of Garden wrote in 1972 (Pain and Pain, 1972). It basically consists of an iron welded mesh external structure, containing a compost pile usually made on woody biomass, inside which a heat exchanger is placed. CHRSs are traditionally insulated on the external part with straw bales and with a waterproof membrane placed at the bottom.

The biomass exploited inside CHRS plants is biologically decomposed through microbial respiration. When

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organic materials are metabolized by microorganisms, O_2 is consumed and CO_2 is liberated, while heat is metabolically generated. Numerous systems to recover this heat for domestic purposes have been tested over the years. The four main systems to extract heat from CHRS were summarized and schematically illustrated by Malesani et al., 2021. In the present study, the life cycle assessment was performed according to the traditional plant configuration pioneered by Jean Pain, that uses a conduction-based approach to recover heat with coiled PE pipes filled with an exchange fluid.

The involved biomass is usually lignocellulosic residual biomass that comes from gardening activities, pruning activities and landscape maintenance activities. Trees, grasses and harvest residues from food crops are the major sources of lignocellulosic biomass. According to Dahmen et al., (2019), every year are worldwide produced 4.6 billion tonnes of lignocellulosic biomass as agricultural residues, of which only 25% are used. CHRS systems improve the on-site reuse of this kind of biomass residue that otherwise should be transported, treated and disposed, with all the related costs and emissions.

At the end of the biodegradation process, the remaining material is compost material. This is an added value for CHRS, considering that compost is an excellent soil amendment to improve agricultural properties, since it increases both the soil organic carbon content and the soil nutrients content (Lord and Sakrabani, 2019). Moreover, it also improves the texture of the soil, allows to increase the water holding capacity of soils and sorb metals, having important impacts on physico-chemical properties of soils and enhancing plant growth and soil health in long terms (Mazumder et al., 2021).

Enhancing the on-site reuse of waste material and producing a still exploitable material to re-reuse on-site, while providing thermal energy, CHRS can be considered a technology that perfectly implements the principles of the circular economy. Normally defined as a new and sustainable model of production and consumption, circular economy involves sharing, leasing, reusing, repairing, refurbishing and recycling existing materials and products as long as possible, extending their life cycle (Butti, 2020). In Figure 1, how CHRS puts into action the principles of circular economy, is schematically illustrated.

The transition to a circular model of economy is indeed relates to the ever-increasing problem of waste accumulation, along with the continuous exploitation of virgin resources, that are promoting the transition from the linear concept of "take-make-dispose" to a model designed in such a way that all the material and waste flows are reintegrated into a cycle (Tamburini et al., 2021).

The circular economy paradigm is based on the concept of the Life Cycle Thinking (LCT), as an approach that seeks to identify possible improvements to goods and services in the form of lower environmental impacts and reduced use of resource, assessing their whole life cycle from the extraction of raw materials to the end of their life (Lazarevic et al., 2012). Among the tools of LCT, the Life Cycle Assessment (LCA) has become a central instrument in environmental management to model, assess and evaluate the impact of a product or a process on ecosystems, natural resources of human health throughout its life cycle (VanderWilde and Newell, 2021). From a general point of view, LCA approach results able to account for the whole process "from cradle to grave" returning both environmental and health impacts by specific indicators (Di Maria et al., 2021).

Since CHRS are plants characterized by a low technology (low-tech), made of materials readily available in the place of construction and installation and they involve in their process the capture and storage of CO_2 with related benefits on the environment, the main objective of this study was to investigate CHRSs whole life cycle impacts on the environment. In LCA, all environmental impacts may be assessed. In the previous research (Malesani et al., 2021a), the authors investigated CHRSs overall costs considering the whole life cycle of the plant, while regarding the carbon dioxide emissions only the plant management

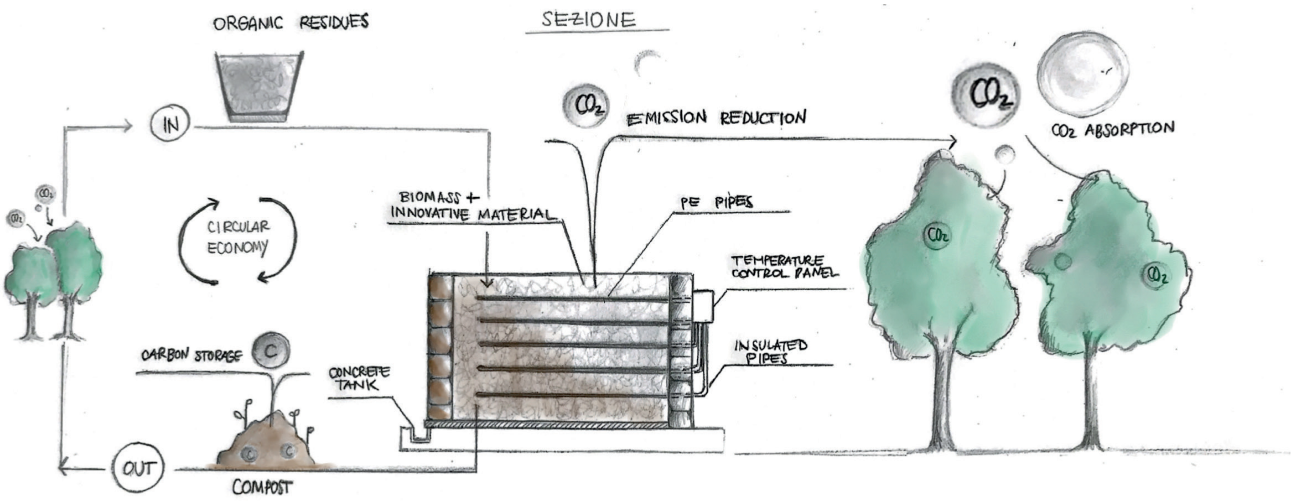

FIGURE 1: Schematic representation of the conceptual model of CHRS implementing the principles of circular economy, through the valorization of the residual biomass.

phase was considered. This further study allowed to estimate the overall emissions related to the whole life cycle through LCA approach of an average CHRS, since these are extremely interesting plants due not only to their very low $CO_{2\text{-eq}}$ emissions associated to every step of the life cycle, but also to their ability to store carbon in soil and avoid $CO_{2\text{-eq}}$ emissions thanks to the compost production and the biomass use. These multiple aspects of CHRSs are deeply interesting considering not only the global need to properly manage waste in a circular way, but also bearing in mind that the major challenge of this century is climate change and that to tackle this challenge, it is necessary to find measures to limit the temperature rise significantly reducing global CO_2 emissions and to reach carbon neutrality within the next 20 to 30 years (Rosenfeld et al., 2021).

Usually, LCA focuses on two key assessment variables: GHG emissions and the primary energy usage. This study focused on GHG emissions, that are mainly carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O). Other GHGs such as sulphur hexafluoride (SF6) and chlorofluorocarbons (CFCs) are not so relevant for energy systems, though SF6 is used to test oil and natural gas pipelines for leaks. Global Warming Potential (GWP) is used to express the contribution of different GHGs to global warming (Bird et al., 2006). In this study, several impact categories were assessed, but just the GWP impact category was deeply discussed and used to perform a comparison with the GWP values coming from literature about LCA studies performed on other technologies for sustainable thermal energy production, that are solar thermal panels and geothermal plants. The GWP data collected from literature, underwent a two-steps screening and were processed in order to have all the values expressed in $kg_{CO2\text{-eq}}/kWh$. A further statistical analysis was performed to better discuss the collected data.

2. MATERIALS AND METHODS

In this study, an evaluation of a traditional CHRS from a life cycle perspective was performed through an LCA study. The study was carried out with the final objective of comparing a CHRS to other sustainable plants for providing domestic thermal energy in terms of environmental impacts.

The evaluations and calculations were based on information deriving from real scale CHRS applications, designed with a traditional configuration.

2.1 CHRS description

The analysed CHRS essentially consists of a heap of raw woody biomass placed inside a cylindrical container system made of welded iron mesh. Since the role of the CHRS is to recover the heat produced from the composting process it is necessary to limit the heat dispersion, therefore the containing cylinder is surrounded by hay bales performing insulation. Water is needed by the system since its functioning is based on a biological process where an appropriate water content is essential to the microorganisms, so a waterproof bottom membrane placed at the bottom allows to collect the leachate produced, send it to a concrete well housing a recirculation pump and recirculate it from the top of the heap. The dimensions of traditional CHRS are usually in the range of 35 to 55 m^3, although this may vary from 25 to 170 m^3, according to Native Power association data (Native Power, 2019). In this study an average dimension of 55 m^3 was chosen to perform the calculations, meaning that the plant has the dimensions reported in Table 1. To ensure the adequate aeration (and oxygen required by microorganisms), perforated polyvinylchloride pipes are placed vertically inside the cylinder promoting airflow by means of a chimney effect (statically).

The generated heat is recovered by means of spirally arranged PE pipes fixed on wire mesh and placed at different heights inside the cylinder. The number of layers depends on the dimension of the plant. In this specific case, 5 layers of PE pipes were considered for a total amount of 500 meters of pipe.

Inside the PE pipes an exchange fluid flows, usually water. This fluid is then sent to an external puffer. The evaluation of the CHRS ends before the connection of the system to the building.

In Figure 2, a schematic representation on an average traditional CHRS is provided.

Since the biomass exploited inside a CHRS is biologically decomposed through microbial respiration, the plant works independently for a limited period. According to monitoring data of real scale plants performed by the current research group (Malesani et al., 2021b), this period usually lasts between 12 and 16 months. After that, the plant needs to undergo a maintenance service to be dismantled and quickly rebuilt with new biomass. During the dismantling of the plant the remaining biomass is extracted and valorized as compost.

This process can be done for several years reusing all the components of the plant; in this LCA study a 15-year CHRS lifetime was considered.

2.2 CHRS thermal power output

The thermal power generated over time by a CHRS can be evaluated from several available data of prototype plants and real scale domestic plants performing heat extraction from composting processes.

Over the past decades, many research papers about the heat generated from composting have been published. Table 2 reports some literature values of the thermal energy that can be recovered from composting processes involving woody biomass, together with further information collected from Native Power and Biomeiler organizations. Data were collected and processed to be homogenized.

TABLE 1: Characteristics of the considered CHRS.

Variable	Unit	Value
Diameter	m	5
Height	m	2.8
Circumference	m	15.7
Volume	m^3	55
PE layers	n	5
PE length	m	500

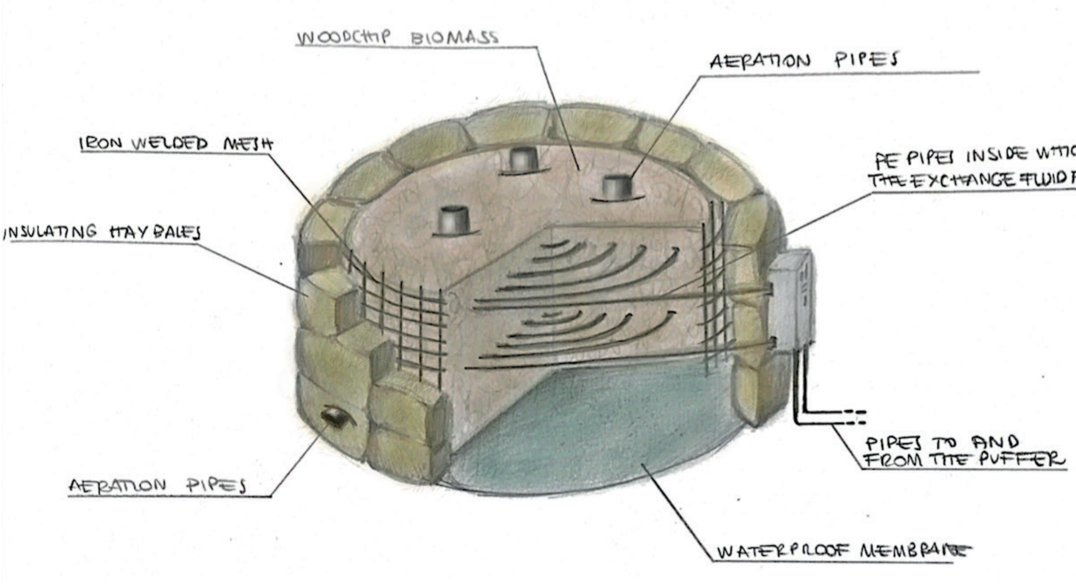

FIGURE 2: Graphic representation of a traditional CHRS.

The volumes of the plants that were considered, range from 0.09 m^3 (pilot scale) to 197 m^3 (real scale). Data about the thermal power output in kW per m^3 of biomass, range from 0.02 to 0.23 kW/m^3. Data can vary a lot depending on the volume of the plant and the duration of the biodegradation process, besides the kind of biomass used inside the plant.

Considering just the real scale plants (more than 30 m^3) that worked for about one year, the thermal power output of a CHRS ranges between 0.05 and 0.1 kW/m^3. This means that implementing a plant of 55 m^3, its power can range between 2.75 and 5.5 thermal kW.

Considering an average plant filled with woodchipped material, working 12 hours a day, with 0.1 kW/m^3 thermal power output, the total thermal energy that can be provided yearly is about 24,000 kWh.

2.3 Real scale CHRS data collection

In order to perform a LCA study, a list of materials and machineries used to install the plant and an estimation of electricity and operations needed to yearly maintain the plant, were necessary. All these data, when possible, were collected during the implementation, installation and operation of a real scale CHRS plant by the research group (Malesani et al., 2021a). Other data were retrieved from the databases used to perform the LCA study.

With regards to the materials involved in the installation of the plant and the need for use of specific equipment, means of transportation and electricity, they were assessed through the real scale plant design and construction and are reported in Supplementary materials A – CHRS list of materials. Machinery consumption has to be considered in the installation phase and in the yearly maintenance activities. Indeed, every year the biomass needs to be chipped before being put inside the plant, a wheel loader needs to be used to put the biomass inside the plant and a truck is necessary to transport the biomass from the place of its production till the plant installation site. Moreover, a recirculation pump for leachate works in the beginning phase, while a recirculation pump for exchange fluid recirculation works several hours a day for the whole lifetime of the plant. Trucks of 7 tons capacity were considered for the biomass transport and for material transport.

2.4 LCA study

The environmental impacts were assessed using an LCA approach according to the ISO 14040 (2006), ISO 14044 (2018) and the ILCD Handbook guidelines (EC, 2010,2012). All the calculations were performed using the SimaPro 9 software.

This study is a "cradle-to-grave" LCA, i.e., it mainly covers all relevant process steps from raw material production to the final waste treatment, recycling or disposal.

In a second step, the GWP results of the LCA were used to perform a comparison between CHRSs and other system to provide sustainable thermal energy. A literature review was performed in order to collect GWP data coming from several LCA conducted and published for two thermal energy generation technologies: solar thermal panels and geothermal plants. Having these LCA wide ranging results, due to technologies evaluated (i.e., differing system designs, commercial versus conceptual systems, system operating assumptions, technology improvements over time) and LCA methods and assumptions, a two-step rigorous screening evaluating the completeness, validity, and data quality of each study was necessary; nine studies were selected as representative, providing 44 data in kg_{CO2}/kWh that were used to perform the comparison.

2.4.1 Goal and system boundaries

The goal of the LCA study was to assess environmental impacts related to a CHRS, from the extraction of the raw materials, through its operative lifetime, to the final disposal or reuse of main components. First of all, the LCA study was performed on an average CHRS and the analysed system includes two main fluxes:

TABLE 2: Thermal power output from small scale composting processes; data coming from literature and elaborated by the research group.

Material	Energy recovered (Er)					Operative conditions				
	kW/m^3	kWh/m^3	MJ/m^3	kWh/kg	MJ/kg	Operative time (h)	Moisture Content (%)	Bulk density (kg/m^3)	Volume (m^3)	References
Woodchips	0.05	406.1	1462	1.015	3.7	8640	70%	1000	55	(Native Power, 2019)
Woodchips	0.05	343.4	1236	0.859	3.1	6480	70%	1000	79	(Biomeiler, 2019)
Woodchips	0.16	112.5	405	0.113	0.4	720	70%	1000	16	(Zantedeschi, 2018)
Chipped brushwood	0.19	820.8	2955	1.492	5.4	4320	45%	550	75	(Pain and Pain, 1972)
Chipped brushwood	0.02	129.6	467	0.324	1.2	6480	NF	NF	197	(Schuchardt, 1984)***
Horse manure, sawdust, woodchips	0.14	83.0	299	0.175	0.6	600	60%	475**	0.9	(Chambers and Supervisor: Allen, 2009)
Bamboo	0.06	56.0	202	0.112	0.4	1008	46%	500	50	(Seki et al., 2014)
Woodchips	0.09	777.6	2799	1.637	5.9	8640	NF	475**	31	Brown, 2014***
Horse manure, woodchips, fresh grass	0.10	85.7	308	1.003	3.6*	864	58%	603	6.7	(Bajko et al., 2019)
Cow manure, grass, sawdust	0.23	70.8	255	0.149	0.5	312	58%	477	2.8	(Mwape et al., 2020)
Green waste	0.10	864.0	3110	1.819	6.5	8640	NF	475**	60	(Cuhls et al., 2020)***
Green waste	0.10	864.0	3110	1.819	6.5	8640	NF	475**	150	(Cuhls et al., 2020)***
Wood	0.06	560.5	1699	1.180	4.2	8760	60%	475**	134	(Kimman et al., 2019)

The factor used to convert MJ in kWh and vice-versa is 3.6 MJ/kWh. kWh/kg and MJ/kg are expressed in terms of kg of biomass. NF= Not Found.
** MJ/kgDM; ** assumed values of biomass density, when not available; ***cited after Zimmermann, 2020*

- On one side, the plant production starting from the extraction of raw materials for the production of main components of the plant, till their final reuse, recycling or disposal;
- On the other side, the production of raw organic biomass to use inside the plant, from the sowing of plant seeds, till the plant growing and harvesting to produce woodchipped biomass to feed the plant.

These components concur to the building and operation of the plant. A life span of 15 years was assumed. After each aerobic treatment cycle, the compost generated by the biomass has been assumed to be exploited on soil as organic fertilizer. Since it cannot be addressed to the specific CHRS all the energy consumption along with the efficiency in energy exploitation outside the CHRS facility were not include in the boundaries of the system (Figure 3), that include the production processes of the components that are necessary for the construction of the plant, the electricity and fuels necessary for its management, the energy consumption during the final composted material management. Direct emissions from the composting processes and indirect emissions due to the plant construction and management are considered and included. Given the multifunctionality of the system that involves energy and material transformation, system boundaries were expanded. Main backgrounds were represented by the activities of extraction of raw materials, manufacture of the main components of the plant, the activities of plants cultivation, their pruning and the wood chipping of the organic residues. Main foregrounds were represented by the CHRS plant operation, energy and organic fertilizer production.

This study was performed for research purpose, with the final objective of comparing the CHRS to other plants in terms of environmental impacts.

For taking into account the multi-functionality of the system that involves energetic and material transformations, the boundaries of the system were expanded (Figure 3).

2.4.2 Functional unit, reference flow and life cycle inventory analysis

The function of the system was the generation of 1 kWh of heat for civil heat needs. This was also assumed as reference flow. The reference flow is not able to significantly affect the backgrounds.

For the electricity consumed, the Italian energetic mix was considered referred to 2019, consisting of the following: natural gas 39%, oil 1%, coal 8.6%, other fuels 4%, hydroelectric 14.7%, geothermal 1.8%, wind 5.3%, photovoltaic 6.8%, bioenergy 5.8%, imports 13.2% (Terna, 2019).

Since the functional unit is considered to be constant the life cycle inventory (LCI) framework is attributional. Heat from Natural Gas fueled heat generator was considered replaced by the one recovered by the CHRS. Also, through the use of compost as soil fertilizer, the mineral fertilizers urea, phosphate P_2O_5 and potash K_2O were considered to be substituted. The amount of organic carbon stored in soil after compost application was considered to be the 15% of the compost total organic carbon, as average value between the values reported by Hermann et al., (2011) reporting that approximately 23% of the organic carbon remains in the soil as humus and Franz et al. (2009), according to whom about 8.2% of the organic carbon supplied with the compost would still be available in the soil over long periods (10-100 years).

Data obtained from measurements and calculations

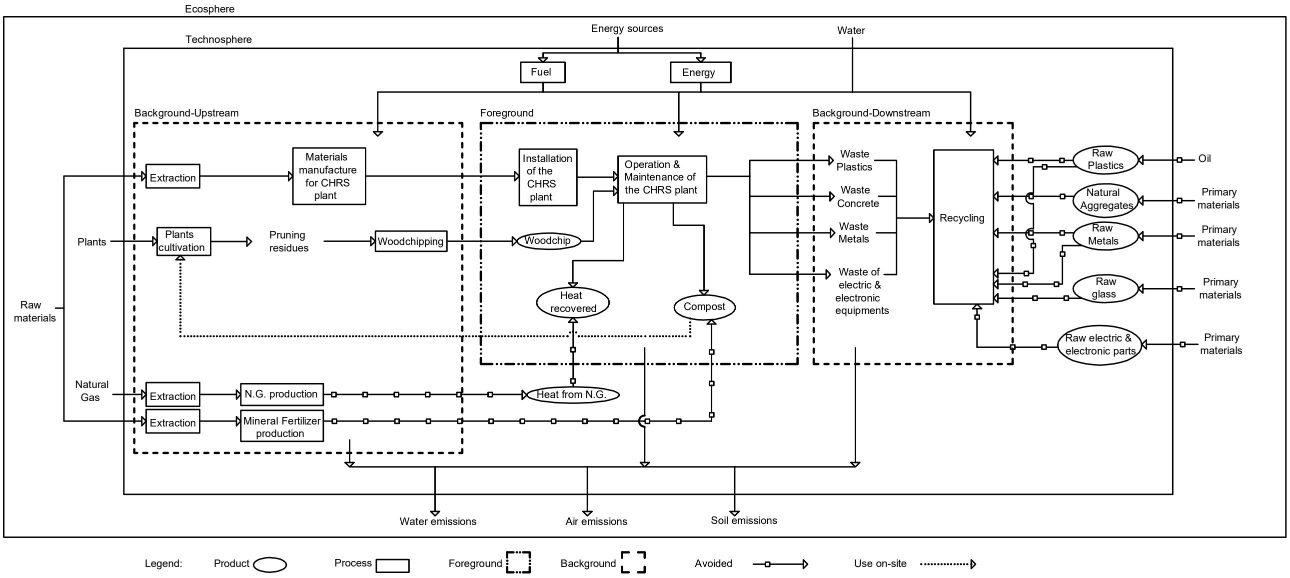

FIGURE 3: Boundaries of the considered system.

performed on the analyzed system, were used for adjusting the interested inventories retrieved from the Ecoinvent 3.7 database. All the materials involved in the installation of a real scale CHRS plant were considered based on the experience of the research team with the installation and monitoring of real scale plants.

2.4.3 Impact indicators and assessment method

Both midpoint and endpoint impact assessments methods were adopted. For the midpoint the ILCD 2010+ method (EC, 2012) was used considering the following impact indicators: Global Warming (GWP at 100 years) (kg_{CO2eq}); Particulate Matter (PM) ($kg_{PM2.5eq}$); Fresh Water Eutrophication (FWE) (kg_{Peq}); Fresh Water Ecotoxicity (FWec) (CTU_e – Comparative Toxic Unit for ecosystems); Human Toxicity, cancer effects (HT_c) (CTU_h – Comparative Toxic Unit for Human Health); Human Toxicity, non cancer effect (HT_{nc}) (CTU_h).

For the Human Health (HH) endpoint (DALY – Disability Adjusted Life Year), the IMPACT 2000+ (Jolliet et al., 2003) assessment method was used.

Since the goal of this study was mainly to assess, evaluate and describe the GWP impacts in terms of $kg_{CO2\text{-}eq}$/kWh, all the other calculated impact indicators are reported in Supplementary materials B – LCA impact indicators, including their graphic representations.

2.4.4 Uncertainty assessment

The margin of error related to the LCA results obtained during the present study, was assessed through a simplified standard procedure based on a pedigree matrix that takes pattern from a work published by Weidema & Wesnæs (1996) and based on the use of basic uncertainty factors (U_b) reported by Frischknecht et al. (2007). The basic uncertainty factors come from expert judgements.

Data sources were assessed according to the following characteristics: reliability, completeness, temporal correlation, geographical correlation and further technological correlation. Each characteristic is divided into five quality levels with a score between 1 and 5, that is assigned to each indicator of the pedigree matrix according to the quality of the available data. On the basis of the assigned score, each indicator corresponds to a given uncertainty factor: U_1=uncertainty factor of reliability, U_2=uncertainty factor of completeness, U_3=uncertainty factor of temporal correlation, U_4=uncertainty factor of geographical correlation, U_5=uncertainty factor of other technological correlation (Di Maria and Micale, 2014). These factors are also based on expert judgements and are reported by Frischknecht et al., (2007).

Once all these values were determined, the square of the geometric standard deviation was then calculated, with a 95% confidence interval, according to Equation 1:

$$SD_{g95} = \sigma_g^2 = exp^{\sqrt{[\ln(U_1)]^2+[\ln(U_2)]^2+[\ln(U_3)]^2+[\ln(U_4)]^2+[\ln(U_5)]^2+[\ln(U_b)]^2}} \quad (1)$$

2.5 Literature data collection and screening approach

Positive emissions of equivalent CO_2 related to the whole life cycle of the CHRS (GWP impact) were finally used to perform a comparison with literature data collected from several LCA studies conducted on other sustainable thermal energy production system: solar thermal panels and geothermal plants.

To collect data from LCA studies, a comprehensive search of the literature was performed. Studies collected were reviewed and screened in two steps.

The studies that passed the first screening step should:

- Be written in English
- Be published after 2010
- Be published as: journal article, conference proceeding, book or book chapter
- Cover thermal energy technologies from solar or geothermal sources

- Report qualitative results from an LCA or review results from multiple LCAs

The second screen consisted of three main criteria:

- Quality: the study had to employ a currently accepted LCA methodology (e.g., following ISO 14040 series standards [ISO 2006]).
- Transparency: the study must report the method transparently providing a description of the system and reporting impacts qualitatively.
- Relevance: the study must evaluate a modern technology, relevant to current or near future.

The studies that passed the two screening steps provided 44 estimates of life cycle CO_2 emissions for the aforementioned technologies and were included in this analysis. Multiple emission estimates from a single reference were considered when different technologies were analysed. Indeed, several references that passed the two screens, provided more than one estimate value, based on either alternate scenario or alternate technologies, e.g., Martinopoulos et al. (2013) performed LCA on a variety of typical Domestic Solar Hot Water System (DSHWS) providing 28 data, Comodi et al. (2016) performed LCA on two different DSHWS, glazed and unglazed, A. S. Pratiwi & Trutnevyte (2021) quantified the life cycle impacts of 6 heating and cooling configurations with geothermal wells providing 6 data and others.

The total amount of 44 emission data drawn from 9 reference, then underwent a processing process necessary to express all of them in kg_{CO2}/kWh. To be considered, studies were required to either directly report life cycle emissions in kg_{CO2} per functional unit or to provide sufficient information to calculate it without using exogenous assumptions. Even rigorous studies did not report emissions in kg per functional unit. To limit errors, the results had to be reported numerically, not only graphically, as happens in many LCA studies.

Duplicate estimates from one study quoting another or from the same author group publishing the same estimate multiple times were not included.

Finally, key parameters values had to be reported to be considered for comparison. The required parameters were:

- Operative lifetime
- Thermal load (in MJ/y or kWh/y). When it was reported in MJ/y, it was converted using a conversion factor of 3.6 MJ/kWh
- Functional unit

Data retrieved from literature were processed and elaborated to achieve uniformity of the unit of measure into kg_{CO2}/kWh. Specifically, Martinopoulos et al. (2013) reported the load covered by Domestic Solar Hot Water System (DSHWS) in MJ/y and the environmental impacts of DSHWS divided into CO_2, CH_4 and N_2O for a total amount of kg of CO_2, kg of CH_4 and kg of N_2O emitted in 15 years. They also reported the kg of CO, NMVOC, NO_2 and SO_2 but they were not considered since they are climate-altering gases with an indirect effect on climate, not directly contributing to Global Warming. So, first the load covered by DSHWS was converted into kWh/y with a conversion factor of 3.6 MJ/kWh, and then a total amount of kg of equivalent CO_2 emitted in 15 years were calculated converting CH_4 and N_2O emissions into emissions of equivalent CO_2 considering the 100-year global warming potentials (GWP100) equal to 28 for CH_4 and to 265 for N_2O. Having the total thermal load covered by DSHWS in 15 years expressed in kWh and the total emissions of $CO_{2\text{-eq}}$ in 15 years expressed in $kg_{CO2\text{-eq}}$, the values were expressed in $kg_{CO2\text{-eq}}$/kWh. The same procedure was applied for data reported by Comodi et al. (2016); the thermal load expressed in MJ/y, was converted into kWh/y with 3.6 MJ/kWh conversion factor and, regarding the global warming potential of DSHWSs already reported in $kg_{CO2\text{-eq}}$, only emissions of CO_2, CH_4 and N_2O were considered. Having the total thermal load expressed in kWh and the total emissions expressed in $kg_{CO2\text{-eq}}$, the values were expressed in $kg_{CO2\text{-eq}}$/kWh. Albertí et al. (2019) directly reported the values of the Global Warming impact in kg_{CO2}. All of the other cited studies (Frick et al., 2010; Karlsdottir et al., 2014; Lacirignola and Blanc, 2013; Pratiwi et al., 2018; Pratiwi and Trutnevyte, 2020, 2021) reported the values of the Global Warming impact in $kg_{CO2\text{-eq}}$/MWh so the values were just expressed in $kg_{CO2\text{-eq}}$/kWh.

2.6 Statistical assessment and comparison

Data collected according to the approach described in the previous paragraph, were statistically analysed and graphically represented using Microsoft Excel. The descriptive statistic was performed (calculating mean, median, minimum, maximum, first and third quartile and interquartile range) and the resulting descriptive statistics were further summarized graphically through boxplots. Boxplot representations are among the most widely used exploratory data analysis (EDA) tools in statistical practice and they allow to quickly visualize the degree of dispersion of the data and they intuitively reflect outliers. Since the adoption of real-life data set, the Boxplots methods, quartiles and interquartile range are chosen as the basis of judgment, which will not be affected by outliers (Li et al., 2016).

The results of the statistical analysis were used to evaluate the variability and the central tendency of the dataset collected from literature, to determine how reliable the comparison it is.

3. RESULTS AND DISCUSSION

GWP impact values in $kg_{CO2\text{-eq}}$/kWh were assessed and used to perform a comparison with other environmental damages data found in literature about other domestic heating systems from renewable sources.

3.1 Environmental effects

The LCA indicated that all the environmental impacts related to the implementation of a CHRSs due to direct and indirect emissions (positive emissions) are lower respect to the benefits due to the avoided primary materials, the replacement of natural gas used as traditional thermal energy production and the replacement of mineral fertilizers (negative emissions). Indeed, from all the impact categories considered emerged a final net negative value, since positive emissions are lower than negative ones. This can be clearly seen from the graphs reported in Supplementary materials B – LCA results.

In Figure 4, the GWP impact for each step of the life cycle of CHRS in terms of kg$_{CO2\text{-}eq}$/kWh is shown. As can be seen, the negative net value indicates the benefits related to CHRS implementation.

Main contribution to GWP impact were by direct and indirect emissions from the plant management phase (operative lifetime of the plant), with a total amount of positive emissions of 0.056 kg$_{CO2}$-eq/kWh, mainly due to the gasoil use for:

- the wheel loader use, for plant filling (37%)
- the woodchip material transport (33%)
- the wood-chipping machine use (29%)

The percentages are referred to the total amount of positive emissions related only to the plant management phase and all the detailed positive and negative emissions with related percentages can be found in supplementary materials.

Emissions from the materials manufacture resulted quite limited too, with a total amount of 0.005 kg$_{CO2\text{-}eq}$/kWh, mainly related to the production of the:

- polyethylene pipes inside which the exchange fluid flows (29%)
- stainless steel welded mesh for the external structure of the plant (25%)
- pumps for recirculating water inside the PE pipes and leachate from the bottom of the plant to the top of it (12% and 12%).

The contribution of the end-of-life stage was practically negligible (0.0002 kg$_{CO2\text{-}eq}$/kWh) since every component of the CHRS is reused or recycled except for the waterproof membrane that is sent to an incineration system with energy recovery. The negative value related to avoided primary materials (avoided thanks to material reuse ore recycle) is equal to -0.006 kg$_{CO2\text{-}eq}$/kWh.

The main contribution to the avoided emissions, is related to the avoided heat production (-0.275 kg$_{CO2\text{-}eq}$/kWh). To evaluate it, it was considered to substitute with a CHRS, a traditional gas heating system. The avoided emissions related to the avoided mineral fertilizer are equal to -0.048 kg$_{CO2\text{-}eq}$/kWh, and it is mainly due to the carbon stored in soil in 15 years of CHRS use (88%).

The combination of the benefits achieved avoiding the use of mineral fertilizers, avoiding the heat production from natural gas and avoiding the primary materials use thanks to material recycle and reuse, leaded to a negative net value of -0.268 kg$_{CO2\text{-}eq}$/kWh.

The numerical values of the GWP impact for each stage are reported in the graphical representation in Figure 4.

From the outputs of the whole LCA, observing all the considered impact categories (reported in Supplementary materials B – LCA results), emerged that CHRS implementation is clearly beneficial in terms of equivalent carbon dioxide emissions, particulate matter emissions, human health, human toxicity, fresh water ecotoxicity and fresh water eutrophication. The findings suggest indeed that, on average, the impact on environment and human health of both direct and indirect emissions from the use of CHRS with heat recovery and compost production, are definitely lower than those due to the production of the same amount of mineral fertilizer, the same amount of heat with natural gas and virgin materials replaced.

For further comparison with other technologies, just the positive emissions were considered, equal to 0.062 kg$_{CO2\text{-}eq}$/kWh.

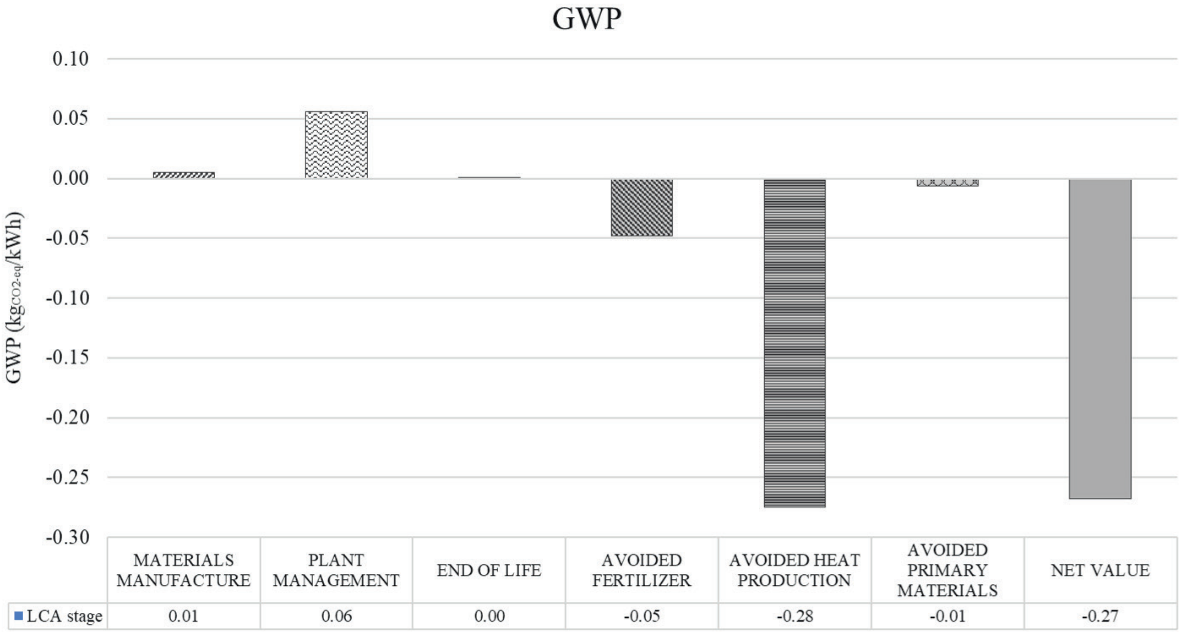

FIGURE 4: Schematic representation of the conceptual model of CHRS implementing the principles of circular economy, through the valorization of the residual biomass.

3.2 Uncertainty assessment

Only the main emissions contributing to the GWP impact category were considered for the uncertainty evaluation. Due to the independency assumed for the background from the system foreground, only direct emissions generated by the processes were considered. Indirect emissions and emissions not influenced by the main process of plant functioning, were not considered (i.e. Materials manufacture phase). Moreover, due to the marginal effect that End of life phase has on the overall GWP impact of the CHRS (contributing for 0.4% to the total emissions), the uncertainty analysis of the related database was omitted.

The CHRS plant was simulated based on the expertise developed by the research group during a real scale plant construction and monitoring with respect to the year 2018. On field data were collected during one-year plant operation and, when not fully available, data were retrieved from literature (i.e plant thermal power output). Woodchip machine use, recirculating pumps functioning, and wheel loader use are real data so high-quality data corresponding to low scores. Regarding the material transport, real data were not available, so an average distance of 20 km was assumed because CHRSs are plant meant to be implemented close to the place where residual biomass is produced. Data were collected from one plant that operated for one year, so the data from the remaining years of operation were estimated on the base of the first year of operation. The LCA was performed considering the real geographical area under study and the electricity mix considered for electricity consumption, is the Italian energetic mix referred to year 2019. For all these considerations, the values associated to the quality indicators of the pedigree matrix are 2, 4, 2,1 and 1 respectively.

On the basis of the data reported in Frischknecht et al. (2007) for Uncertainty factors and on the basis of the Eq (1), the variance was evaluated. Uncertainty factors scores and values assigned are showed in Table 3, together with the final variance value equal to 1.13.

Results show that CO_2 emission s_g^2 reflects substantially the basic uncertainty factor U_b, indicating that the uncertainty contribution introduced by the pedigree matrix is not relevant. In fact, s_g^2 equal to 1.13 emerged to be just 7,5% higher than U_b, equal to 1.05.

The higher Uncertainty factor is the second one, related to Completeness of the data, since the plant was monitored for a shorter period of time respect to the operative lifetime considered for the LCA.

The main reason, contributing to the resulting variance, is the fact that data comes from real scale plant designed, built and monitored by the research group, hence uncertainty factors related to geographical correlation and further technological correlation are the lowest ones.

4. COMPARISON WITH OTHER TECHNOLOGIES

Positive direct and indirect emissions of equivalent CO_2 related to the whole life cycle of the CHRS (GWP impact) were used to perform a comparison with literature data collected from several LCA studies conducted on solar thermal panels and geothermal plants. Negative emissions (avoided emissions) were not considered in this comparison, since data collected from literature only concerned the kg of $CO_{2\text{-eq}}$ produced during the life cycle of the plants.

4.1 Data collection and statistical assessment

Collected values were expressed in $kg_{CO2\text{-eq}}$/kWh in order to be foreseeably comparable to the values obtained from the current LCA performed on CHRSs. The values found in literature that passed the screening approach and were considered for statistical assessment and comparison, are resumed in Supplementary materials D – literature data collection. A total amount of 44 values were considered, 31 related to SHWS and 13 related to GHS.

When the values were not expressed in $kg_{CO2\text{-eq}}$/kWh, they were processed or converted with adequate conversion factors. Processed values are marked in the table reported in supplementary materials.

A statistical assessment of variability and central tendency of the processed dataset was performed. Central tendency is reported using both the medians and arithmetic means od the datasets. The variability of the datasets is also described using multiple parameters, including the range (difference between maximum and minimum values) and the interquartile range (IQR) bounded by the 25th and the 75th percentile values. Figure 5 displays box plots for the life cycle carbon dioxide emission estimates of the two technologies considered in the present study and numerical results are provided in Supplementary materials E – statistical analysis.

From the Boxplot statistical representation (Figure 5), it emerges that the datasets about Solar Hot Water System (SHWS) and Geothermal Heating Systems (GHS) in $kg_{CO2\text{-eq}}$/kWh have a low variability, especially for SHWS, meaning that data are well consistent and do not vary a lot from the mean value equal to 0.06 $kg_{CO2\text{-eq}}$/kWh It also emerges from the interquartile (IQ) ranges, equal to 0.006 for SHWS. Moreover, on one side, as can be seen from the graph, SHWS data follow a quite normal distribution where

TABLE 3: Scores and values assigned to uncertainty factors and resulting variance for GWP impact category.

	Reliability	Completeness	Temporal correlation	Geographical correlation	Further technological correlation	Basic uncertainty factor CO_2	Square of the standard deviation
Uncertainty factors	U_1	U_2	U_3	U_4	U_5	U_b	
Scores	2	3	2	1	1		
Values	1.05	1.10	1.03	1	1	1.05	1.13

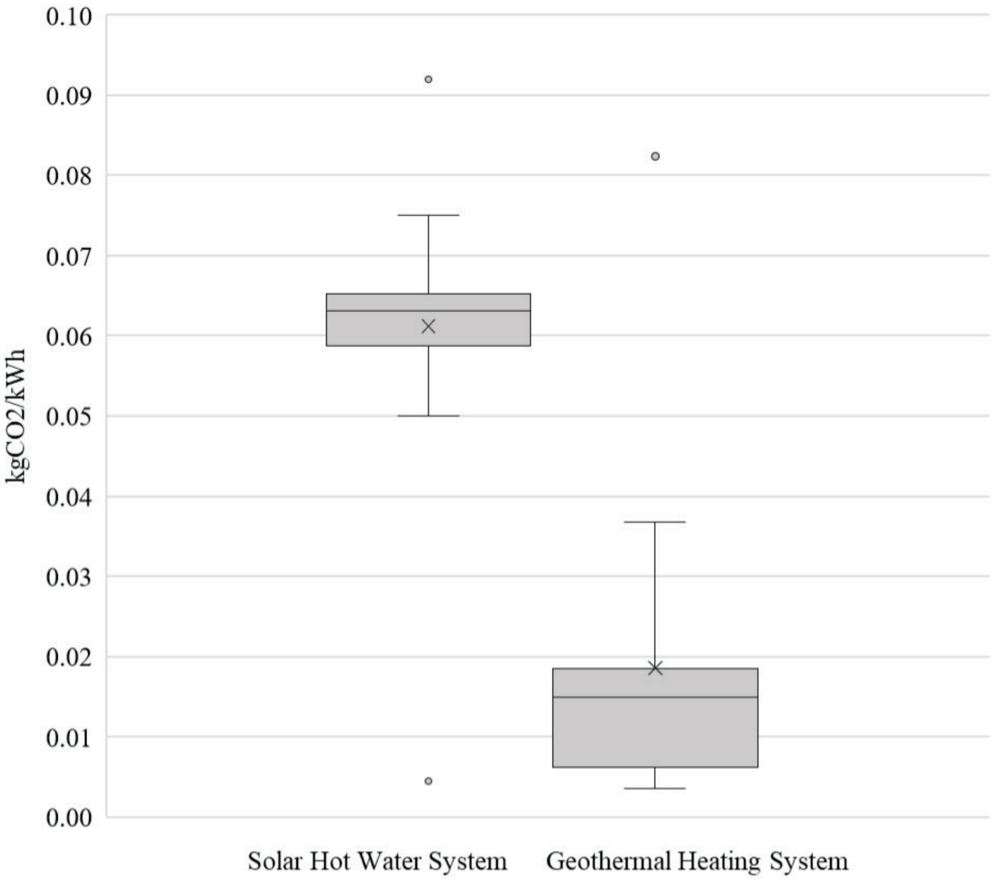

FIGURE 5: Box plot of the GWP impact in $kg_{CO2\text{-}eq}$/kWh for Solar Hot Water Systems and for Geothermal Heating System (literature data). The line within the box shows the median value, the box denotes the range of 50% of data, X stands for the average value and dots stand for outliers.

mean and median values are almost equal (0.061 and 0.063 $kg_{CO2\text{-}eq}$/kWh respectively), well describing the behaviour of the dataset. On the other side, GHS distribution emerges to be asymmetric, rightly skewed, with a median value equal to 0.015 $kg_{CO2\text{-}eq}$/kWh lower than the mean value equal to 0.019 $kg_{CO2\text{-}eq}$/kWh.

4.2 Data comparison and discussion

Just considering the positive emissions assessed through the LCA study equal to 0.062 $kg_{CO2\text{-}eq}$/kWh, CHRSs is in line with the literature data collected for SHWS (mean value 0.061 $kg_{CO2\text{-}eq}$/kWh), while it is slightly higher than the values collected for GHS (mean value 0.019 $kg_{CO2\text{-}eq}$/kWh).

Finally, it is worth noting that CHRSs compared with traditional systems to provide domestic heating is way more sustainable in terms of $kg_{CO2\text{-}eq}$/kWh. Indeed, from literature some data about the use of Electricity and Natural Gas Systems (NGS) for domestic heating were also collected: from Martinopolous et al. emerged a value of 1.41 $kg_{CO2\text{-}eq}$/kWh if the thermal load is entirely covered by electricity, using country specific data for electricity production in Greece (Martinopoulos et al., 2013) and Pratiwi reported a value of Bayer et al. of 1.18 $kg_{CO2\text{-}eq}$/kWh as electricity mix average carbon emission in Poland (Bayer et al., 2013; Pratiwi and Trutnevyte, 2020). Moreover, Albertì et al. reported 0.264 $kg_{CO2\text{-}eq}$/kWh for domestic hot water production from NGS (Albertí et al., 2019).

5. CONCLUSIONS

CHRS is in a position to compete on the market with both traditional and green technologies to provide sustainable domestic heating. Malesani et al. (2021) reported a comparison between different scenarios involving CHRS, pellet combustor, natural gas condensing boiler, solar thermal panels and geothermal plant to heat a farmhouse located in northern Italy and demonstrated that the implementation of CHRS allow to decrease the cost in €/kWh provided considering not only the operative phase, but also the capital costs (design, construction, installation) and the dismantling costs.

This study represents a further research about the emissions related to the full life cycle of CHRS by means of a LCA study, to evaluate the feasibility of installing CHRS as alternative to other heating system with advantages in both economic and environmental viewpoints. The CHRS was analyzed as a system to produce heat in order to be comparable with other heating system, considering the kWh of thermal energy produced by the system.

Considering both positive and negative emissions related to all the life cycle stages, CHRSs present a negative Net value of GWP indicator emissions equal to -0.268 $kg_{CO2\text{-}eq}$/kWh.

Compared with Solar Hot Water Systems and Geothermal Heating Systems considering only the positive emissions, CHRSs emissions of $CO_{2\text{-}eq}$ emerged to be in line with SHWS and to have slightly higher emissions than GHS. Considering the traditional technologies for domestic heating (Electricity and Natural Gas Systems), CHRSs appear to be far more environmentally sustainable in terms of emissions.

To conclude, the utilization of renewable technologies as alternatives to conventional heating systems generally results in low emissions of greenhouse gases. GWP impact values ranging from 0.004 to 0.092 $kg_{CO2\text{-}eq}$/kWh were found for SHWS and ranging from 0.004 to 0.082 $kg_{CO2\text{-}eq}$/kWh were found for GHS though a literature review and data collection process. A positive value equal to 0.062 $kg_{CO2\text{-}eq}$/kWh and a negative value equal to -0.329 $kg_{CO2\text{-}eq}$/kWh were calculated through LCA for an average CHRS. Positive values are especially related to the use of specific machineries for yearly plant maintenance, while the negative value mainly represents the emissions avoided thanks to the use of a CHRS respect to the use of a conventional heating system and thanks to the use of compost as fertilizer respect to the use of conventional mineral fertilizers. The negative Net value underlines the highly beneficial aspects related to the implementation of a CHRS considering its overall life cycle emissions.

A further possible evaluation of the system could consider the kWh of thermal energy available for a domestic utilization and has to be implemented to specific cases considering detailed aspects of a plant, among which: the distance of the plant from the final user, the specific type of heat exchanger, the temperature levels required by the users' building and so on.

This first step of environmental impacts and advantages related to the implementation of a CHRS allowed to demonstrate that it is worth investigating in more depth and detail the real-scale implementation and utilization of CHRS, especially comparing it with other system to provide domestic thermal energy, since it emerged of having great potential to enter the market of sustainable heating production with very low costs and low environmental impacts.

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COMPOSTING OF VEGAN KITCHEN WASTE: APPLICABILITY ASSESSMENT

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ABSTRACT

Kitchen waste from vegan households can be a raw material for home or backyard composting. However, the use of waste without components like eggshells or meat can constitute several problems, including the decrease of the process temperature and the content of Ca, N, K, Mg, Mn, Na, and S. The scientific aim of the study was to investigate possibility of production and applicability of vegan compost (VC) produced from kitchen waste during 12-weeks backyard composting. The VC properties have been compared to the traditional compost (TC), made from plant- and animal-origin waste materials. Analyses showed that VC and TC have similar properties, reaching a pH close to 7.5, respiratory activity AT_4 <2 $mgO_2 \cdot (g\ dry\ mass)^{-1}$and dry matter content of ~79%. VC didn't show any phytotoxic effect on garden cress; it was characterized by the highest seed germination (100%) and it stimulated the growth of plants' roots. It was characterized by a higher content of phosphates, P and K, and achieved a lower BOD/COD ratio, demonstrating its maturity and low rotting potential. VC also contained less chloride, Ni, and Pb and showed a lower potential for nitrates leaching. The values of contaminants contained in VC samples didn't exceed the limit values for organic fertilizers. Therefore, studies indicated that plant-based kitchen waste can be a valuable substrate to produce compost and proved that vegan households, restaurants, and cafes are not disqualified from implementing a circular economy by using them assecondary material.

1. INTRODUCTION

Among the factors influencing the choice of a vegan diet, in addition to religious or ethical beliefs regarding animal welfare, environmental aspects are increasingly mentioned (Christopher et al., 2018). They pertain to the reduction of the negative impact of the livestock industry, consisting largely in greenhouse gas emissions and deforestation (Ruby, 2012). Growing awareness is leading to the increase in the number of vegans in the world, especially among the younger generations (Aavik, 2019). A vegan diet, defined as the complete relinquishment of animal products such as meat, dairy, eggs, and honey (Norman & Klaus, 2020), has become common in most Western countries (Dolan, 2016). For example, in the UK the number of people who declare to follow a plant-based diet increased in 2006-2016 by 350% (Marsh, 2016). The Vegan Society, a charity promoting veganism, saw a 20% increase in membership over the 3 years from 2011 to 2014 (The Vegan Society, 2014). This trend is also visible in the media, which defined 2014 as "the year of the vegan" (Rami, 2014). The elimination of animal products in the diet also has the backing of institutions such as The United Nations, who argue that plant-based diets benefit the environment (Carus, 2010).

Growing popularity of veganism is reflected in increasing production of vegan food, the supply of meat alternatives, and the number of restaurants serving plant-based dishes (Norman & Klaus, 2020). Therefore, these products have become a potential composting material, whereas vegan restaurants or cafes – become sources of feedstock for producing vegan compost. At the same time, kitchen waste from vegan households can be used as a raw material for home or backyard composting, a procedure that has become more important in residential waste management in recent years (Faverial & Sierra, 2014).

The main idea of composting is to produce a valuable product – a material that has a high potential for use in

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agriculture as a soil amendment and may replace or reduce use of synthetic fertilizers (Radziemska et al., 2019). For this purpose, a final product which is made of a wide range of substrates, such as green waste from gardens and parks, urban solid wastes, or agricultural waste such as manure, is used (Pellejero et al., 2017). The benefits of applying compost as an organic fertilizer are due to many related effects of this material on the soil. By the direct impact on its macrostructure, it changes the pore volume, leading to a more favorable moisture distribution, and consequently gas exchange (Pellejero et al., 2017). The use of compost also reduces the apparent density, increases the stability and permeability of soil aggregates, as well as water retention capacity, thus contributing to the inhibition of land erosion (Zebarth et al., 2011). In addition, compost, as a source of humic substances, promotes favorable conditions for plants' growth, resulting from the increase of the buffer capacity of the soil and cation exchange capacity (Pellejero et al., 2017). It is related to fertilization through the increased carbon and other nutrient supply (like N, P, K, Ca, Fe) (Kowaljow & Mazzarino, 2007). This increases the activity of microorganisms (Pellejero et al., 2017) and may increase the yield, quantity, and quality of the desired plants, incl. fruit (Giannakis et al., 2014). Additionally, the use of compost has an environmental justification; it reduces greenhouse gas emissions by increasing soil's carbon storage (Cerda et al., 2018) and what is more, composting might offer the opportunity for energy recovery (de Souza Lima & Mahler, 2020).

However, composting vegan waste that doesn't contain elements such as eggshells or meat may lead to several problems. The lack of the first component may reduce the content of calcium necessary for plant growth (Quina et al., 2017), while the lack of meat – lowering the process temperature and the content of N, K, Mg, Mn, Na, Sr and S (Storino et al., 2016). Moreover, home and backyard composting of vegan waste may lead to the formation of excessive leachate, which is associated with the high proportion of fruit and their potential to fast degradation and thus the production of organic acids (Chanakya et al., 2007). An important aspect related to the use of compost as a crop additive is also its phytotoxicity. In order to secure its functions as a material supporting seed germination and plants' growth, the material stabilization during the composting process is necessary to remove possible phytotoxic substances (Illera-Vives et al., 2015). Thus, it is necessary to use the effective process technologies that enable the production of a good quality product, to keep a sustainable household level circular economy.

Therefore, the following question arises: Is the vegan backyard compost applicable for home-garden purposes like a traditional compost? The scientific aim of the study was to investigate the possibility of production and applicability of high-quality vegan compost from kitchen waste during backyard composting. The properties have been compared to traditional compost, made from plant- and animal-origin waste materials.

2. MATERIALS AND METHODS

2.1 Composted waste

Kitchen waste of plant origin (fruit and vegetable remains, peelings, coffee grounds, tea bags – vegan compost) and their mixture with animal-origin waste (eggshells, cheese, skins, yoghurts – traditional compost) were obtained from a 3-person household from May to July 2020 (spring and summer period, Poland). Substrates were collected twice a week throughout the composting process, stored in a freezer at T=-18°C, separately for vegan and traditional samples. After completion of the field tests of backyard composting, they were transported in a portable freezer to the Biomass and Waste Laboratory, Wrocław University of Environmental and Life Sciences, Poland. Properties of raw materials are presented in Figure A.1 (Supplementary content).

2.2 Experimental set-up

Vegan and traditional waste samples were backyard composted in two designed composters, built of plastic crates (26 dm^3 each). To increase the volume of each composter to 122 dm^3, a garden net based on a wooden bracket was attached. The air flow into the waste mass was provided through the perforation of crates walls. Additionally, to facilitate aeration, the bottom of each box was covered with three layers: 20 cm high layer made from small branches and sticks, then a layer of garden soil, and finally a layer made of mowed grass with mass ratio 3:1 for the last two (Figure 1). The composters were placed in a shaded, wind-protected place without water accumulation. On each day of the process, air temperature (°C), its humidity (%), and the amount of precipitation (mm) were measured at five-hour intervals (9 am, 2 pm, and 7 pm).

The backyard composting process was carried out outdoor in May-July 2020 for 12 weeks with mixing the material after 8 weeks in order to homogenize the sample and equalize the moisture content. The substrates were dosed twice a week, resulting in the accumulation of ~9 and 12 kg of vegan and traditional material, respectively (Figure 2).

2.3 Composts Characteristics

2.3.1 Sieve analysis

The vegan and traditional composts produced during the 12-week process were sifted separately without layers of branches and sticks, soil and mowed grass by laboratory shaker with electromagnetic drive MULTISERW-Morek LPzE-4e, Marcyporębie, Poland, using sieves with a mesh size of 16, 8, and 3.15 mm after shaking for 15 minutes. The fractions >16 mm, 16-8 mm, 8-3.15 mm, <3.15 mm were then weighed with the laboratory scale RadWag WPT/R 15 C2, Radom, Poland, with an accuracy of 0.01 g.

2.3.2 Dry Matter and Organic Matter Content

Plant- and animal-origin waste added each week to the composting process, as well as the samples of obtained vegan and traditional composts, were analyzed for dry matter content at 105°C for 24 hours in three replications using the laboratory dryer WAMED, model KBC-65W, Warsaw, Poland, by PN-EN 14346:2011 (Polish Committee

FIGURE 1: Backyard composters bottom preparation: a) branches and sticks layer; b) mowed grass layer; c) covering garden soil; blue crate - vegan compost, black - traditional compost.

for Standardization, 2011a). After that, all these samples were analyzed for organic matter content at 550°C using the muffle furnace Snol 8.1/1100, Utena, Lithuania, in accordance with PN-EN 15169:2011 (Polish Committee for Standardization, 2011b).

2.3.3 Physicochemical Composition and Heavy Metals Content

For vegan and traditional compost, in accordance with the PN-Z-15009:1997 standard (Polish Committee for Standardization, 1997), water extracts were made in three repetitions. Briefly, dry matter content was determined in the collected materials' samples in accordance with the procedure described in section 2.3.2. Then the samples were prepared in a 1:10 ratio of compost to water (m/m). Three conical flasks with a volume of 0.5 dm^3 were prepared, 41.66 g of vegan and traditional compost were placed in each, and 333.33 ml of water of purity grade 3 was added. After 1 h, the flasks were closed and shaken for 4 hours using digital orbital shaker ELMI DOS-20L, Calabasas, USA. Then the flasks were uncovered and placed for 16 h under static conditions. After this time, they were closed again, and shaking was repeated for another 4 hours. After that, the samples were left for another 2 h for solids sedimentation. The obtained water extracts were filtered using 0.45 μm filter paper and then subjected to the physicochemical analyzes listed in Table A.1 (Supplementary content).

2.3.4 Respiratory Activity

AT_4 respiratory activity was determined in triplicates for vegan and traditional compost using the OxiTop®Control measuring system, Weilheim, Germany, in accordance with Binner et al. (2012); Kilian & Macedowska-Capiga (2011). Briefly, ~50-55 g material samples in 2.5 L glass vessels were placed for 7 days in a WAMED KBC-65W, Warsaw, Poland, climatic chamber at 20°C. Each day the vessels were opened for 15 min to aerate the samples.

Based on the Equation 1, the total oxygen consumption was calculated for the samples of vegan and traditional compost:

$$OD = \Delta p \cdot \frac{M_{O2}}{R \cdot T} \cdot \frac{V_{ges} - V_{abs} - V_{sample}}{m_{d.m}} \tag{1}$$

where:

OD: oxygen consumption, $mgO_2 \cdot g\ d.m.^{-1}$,

Δp: pressure difference, mm hPa,

M_{O2}: molecular weight of oxygen, M_{O2}=31988 mg mol^{-1},
R: general gas constant, R=83,14 dm^3 hPa (K mol^{-1}),
T: temperature, K,
V_{ges}: total volume of the measuring vessel, dm^3,
V_{abs}: volume of the absorber and internal auxiliary equipment, dm^3,
V_{sample}: sample volume, dm^3,
$m_{d.m}$: mass of dry matter, g.

According to Kilian & Macedowska-Capiga (2011), the so-called lag-phase, which ends when the mean of the 3-hour measuring interval reaches 25% of the mean of the 3-hour interval with the greatest oxygen demand, was substracted.

2.3.5 Composts Phytotoxicity

The experiment was carried out using water extracts of vegan and traditional composts prepared according to the procedure described in section 2.3.3 with seed germination and early growth microbiotest with higher plants – Phytotoxkit (MicroBioTest Inc., 2004).

First, a test of water holding capacity of the reference soil was performed by filling a 100 cm^3 cylinder with 90 cm^3 of soil sifted through a sieve with a mesh size of 2 mm and 50 cm^3 of distilled water. It was mixed until the soil was completely saturated with water. After reaching the equilibrium state (complete saturation of the soil and formation of a water layer above the soil surface), the supernatant was poured into a 50 ml measuring cylinder. The operation was repeated after a few minutes. The volume of water required to completely saturate the reference soil was calculated as the difference in the volume of water added to the soil and the volume of the supernatant recovered in the measuring cylinder. Then for each treatment, the 90 cm^3 of reference soil was placed on a transparent PVC test plate composed of a bottom part separated by a middle ridge into two compartments and a flat cover (Figure 3). Subsequently, 30 cm^3 of water extract was introduced into the soil with a syringe. The material was evenly leveled to obtain a uniform layer thickness. A paper filter was placed on the surface of the soil, and at its top edge 10 seeds of *Lepidium sativum* were placed in one row at the same intervals from each other. The plate was closed and placed in a holder for 3-days vertical incubation in the dark at 25°C in a climate chamber ST 3 BASIC, POL-EKO-APARATURA, Wodzisław Śląski, Poland. The experiment was performed in 3 variants: vegan compost, traditional compost, and control (reference soil with distilled water) in triplicates. The image of the seed germination capacity in each plate was recorded with a camera after 3 days, and the root length was measured with the ImageJ software, Wayne Rasband.

Based on the Equation 2, the percentage of seed germination was calculated:

$$A = 100 \cdot \frac{(a-b)}{a} \tag{2}$$

where:
A: seed germination, %,
a: total amount of seeds,
b: number of non-sprouted grains.

The inhibition of root elongation was calculated according to:

$$I = 100 \cdot \frac{(C-T)}{C} \tag{3}$$

where:
I: inhibition of root elongation, %,
C: root length value measured for samples with reference soil, mm,
T: value of the root length measured for samples with the tested material, mm.

3. RESULTS AND DISCUSSION

3.1 Physical and Chemical Properties of Composts

Physical and chemical properties are not the only important elements determining compost quality; they also influence its utilization and handling (Agnew & Leonard, 2003). One of them is the particle size distribution of the product of the process. Vegan and traditional compost differed in particle size distribution, the former being distinguished by a higher share of the largest and the smallest fraction (45.90 and 27.88% for groups >16 mm and <3.15 mm, respectively), while in the case of animal-based material, the largest share was recorded for the first two com-

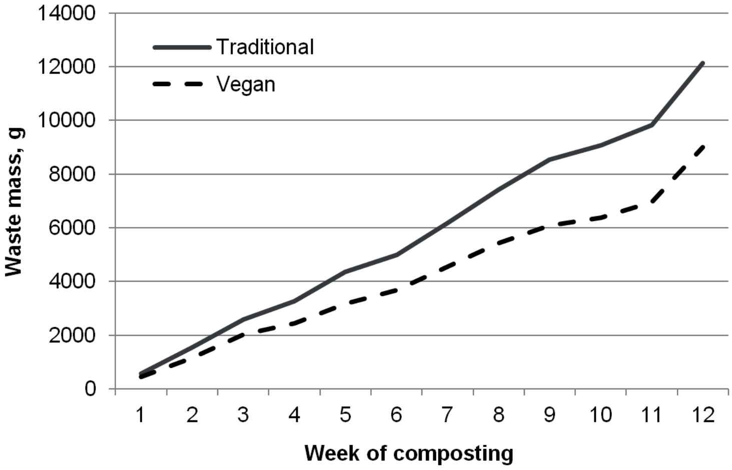

FIGURE 2: Cumulative mass of traditional and vegan waste added to backyard composting.

partments with the largest particle diameter (35.34 and 26.49% for fractions >16 mm and 16-8 mm, respectively, Figure 4). The greatest percentage of the >16 mm group in vegan compost was related to the type of its main building components, including large-sized peelings derived from vegetables such as zucchini, potatoes, celery, but also from turnip, radishes, and lettuce leaves; the high proportion of the fine fraction can be associated not only with greater susceptibility to decomposition of waste of plant origin, but also with their form, e.g. coffee or tea grounds. Although these kinds of wastes were also subjected to traditional compost, a smaller differentiation between the groups was observed here, which may result from the presence of eggshells, which were initially combined into coherent conglomerates, separated during the sieve analysis, and passed to each of the granulometric fractions.

The initial water content in the substrates changes during the composting process due to the decomposition of organic compounds available in the waste and related heat generation and vaporization (Liao et al., 1997). The final dry matter content in the conducted studies was similar for both types of compost, averaging 80.00 and 79.08% for traditional and vegan samples, respectively (Table 1). A similar level was reported for composts made from cattle manure and its mixtures with herbal plants residues (Khater, 2015). The obtained result could have been influenced by the location of the composters in a place where no water accumulated, but also by the duration of the process. The lower moisture level of vegan and traditional samples compared to the reported by (A. Kalamdhad & Kazmi, 2008) in the case of compost made of cattle manure, sawdust and vegatable waste in the rotary drum composter (~20 and ~50% respectively), may be due to the fact that the processes carried out differed significantly in duration. In our experiment, during the 12 weeks of backyard composting, moisture could have been lost as a result of a number of processes such as convective air movement, water vapor diffusion in high temperatures areas or during material turning after 8 weeks (Agnew & Leonard, 2003), which may not have happened or occured to a lesser extent during the 20 days of the composting of (A. Kalamdhad & Kazmi, 2008).

Vegan and traditional compost were characterized by similar pH (7.5 and 7.8, respectively, Table 1). Values not exceeding pH=8 were also recorded for the compost pro-

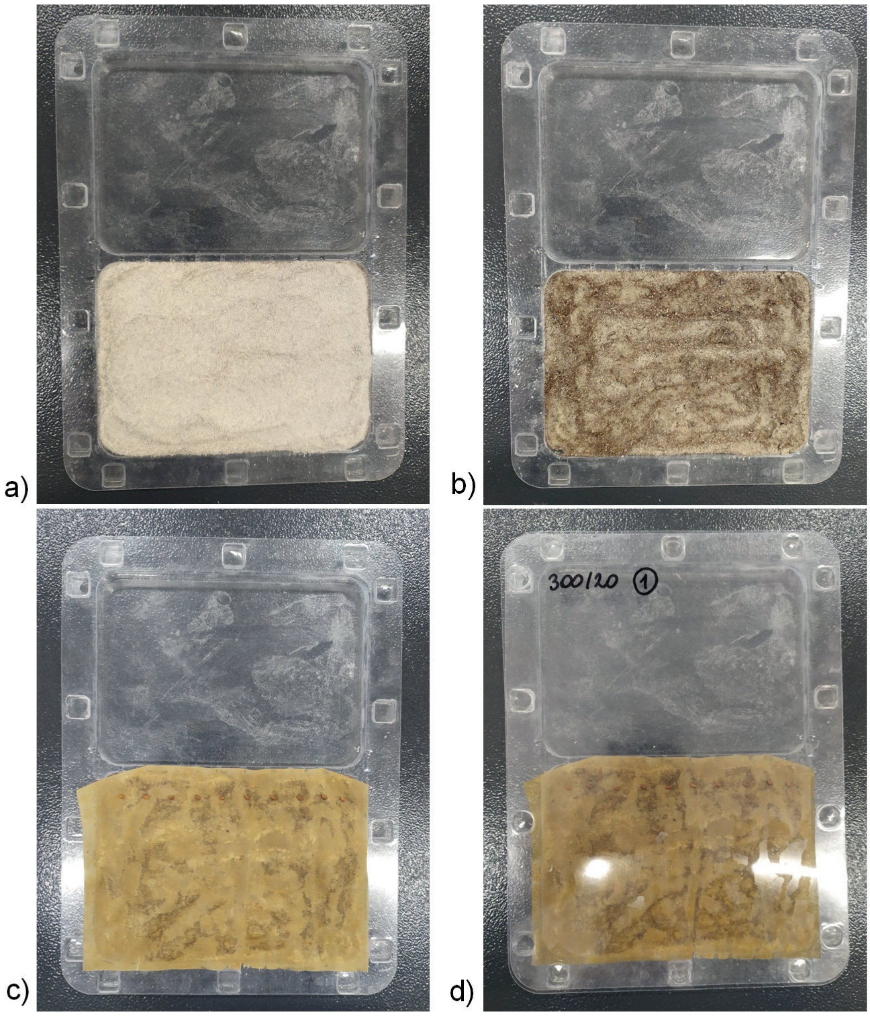

FIGURE 3: Procedure of using the Phytotoxkit test: a) reference soil on a test plate; b) soil saturation with distilled water, c) Lepidium sativum seeds on a paper filter; d) closed test plate prepared to incubation.

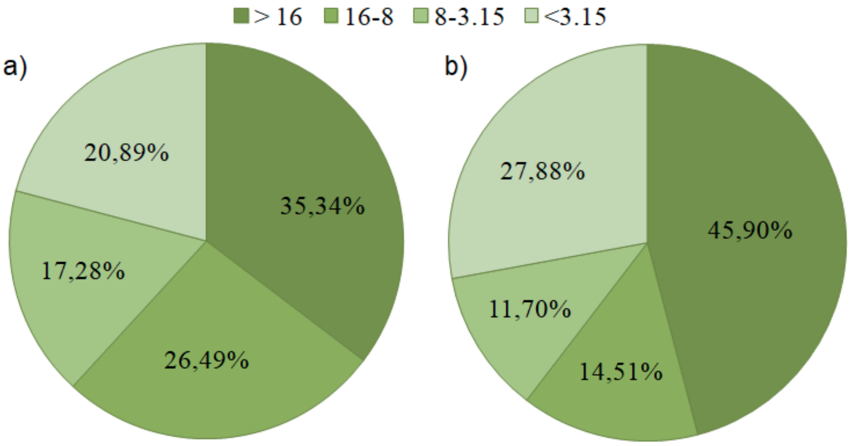

FIGURE 4: Particle size distribution (weight, %): a) vegan compost; b) traditional compost.

duced by (Kalamdhad & Kazmi, 2009b) from cattle manure, mixed green vegetable waste, food waste, grass cuttings, paper waste and sawdust, where the pH at the second and the third run of composting was equal to 7.92 and 7.72 respectively, and for composts tested by (Khater, 2015) (pH ranging from 7.2 to 7.8 for cattle manure and sugar cane plants residues, as well as for herbal plants residues, respectively). The similarity between samples obtained as a result of backyard composting conducted in this research was also seen in case of turbidity of water extracts. A lower index was recorded for vegan compost (16.2 compared to 17.6 NTU – Nephelometric Turbidity Units – for traditional compost, Table 1). These values are significantly lower than obtained by other researchers; turbidity of raw compost leachate tested by (Tawakkoly et al., 2019) reached 670 NTU; after processing it significantly decreased, but still exceeded approx. 14x the values noted in this experiment (252 NTU). Even higher value was obtained by (Brown et al., 2013), where the initial compost leachate was characterized by turbidity at the level of 1370 NTU. Such low results for vegan and traditional composts compared to those available in the literature indicate the advantage of their safe use. Compost leachate, due to the content of excessive nutrients and other substances, can become dangerous and harmful to the earth's surface and groundwater. Therefore, it is particularly important to manage properly the composted mass of waste and the final product.

TABLE 1: Physical and chemical properties of final composts; d.m. – dry matter.

	Vegan compost	Traditional compost
Dry matter content, %	79.08 ± 1.84	80.00 ± 0.05
pH	7.50	7.80
BOD_5, mg O_2·dm^{-3}	19.20	20.00
COD (Cr), mg O_2·dm^{-3}	109.60	94.80
BOD/COD	0.18	0.21
AT_4, mg O_2·g d.m.$^{-1}$	1.70 ± 0.5	1.90 ± 0.4
Conductivity, μS·cm^{-1}	450.00	537.00
Turbidity, NTU	16.20	17.60

An important aspect related to the use of compost as an organic fertilizer is its stability, which will ensure that biological processes do not continue (to a large extent) after its application, leading to a number of undesirable effects, such as the formation of odors, hazardous substances and pathogens and the associated occurrence of insects and flies (Kalamdhad & Kazmi, 2008). Therefore, it is important to know the amount of readily biodegradable organic substances (Bernai et al., 1998). The indicators used for this purpose, such as Biological Oxygen Demand (BOD), Chemical Oxygen Demand (COD) or respiratory activity AT_4, can significantly facilitate the interpretation of the possibility of safe use of the produced compost. The analyzed samples of vegan and traditional materials were characterized by low levels of BOD_5 and COD_{Cr}, while in the case of the latter indicator the difference between composts was greater, and the plant-based material showed a higher level (109.60 mg O_2·dm^{-3} compared to 94.80 mg O_2·dm^{-3}, Tab. 1). Nevertheless, it can be noted that the compost samples produced during the 12 weeks of the process differed significantly in terms of these two parameters from the results obtained by other researchers. BOD values of vegan and traditional composts were approx. 5.4x lower than the final result for vegetable waste, cattle manure and sawdust composted for 20 days (Kalamdhad & Kazmi, 2009b), 6.6x lower than during poultry litter composting for 30 days (Gupta & Doherty, 1990), 10.3x lower compared to the food mix and vegetable waste with grass cuttings and 17.1x lower compared to compost made from cattle manure, food waste, vegetable waste, paper waste and sawdust (20 days for both experiments, Kalamdhad & Kazmi, 2009b). Slightly lower variability can be observed when comparing the COD index; other researchers obtained about 4.5-5x higher values (Kalamdhad & Kazmi, 2008; Kalamdhad & Kazmi, 2009b). Importantly, however, the vegan and traditional composts produced as a result of the research were characterized by a low BOD/COD ratio, reaching 0.18 for the vegan compost and 0.21 for the traditional compost. These results prove that the produced composts consist of organic matter with low potential for further decomposition, i.e. they are microbiologically stable (Mangkoedihardjo, 2006). This is confirmed by (Kalamdhad & Kazmi, 2008), considering their BOD/COD rate of 0.23 at the end of the process as a sign that only non-biodegradable parts remained in the compost. Higher requirements for the stability of this organic material are made by (Borglin et al., 2004), considering the BOD/COD ratio ≤0.1 limit as safe.

Considering the respiratory activity of the analyzed compost samples expressed as the AT_4 index, a similar conclusion can be reached as in the case of the BOD/COD ratio. Vegan and traditional materials showed low AT_4 levels, on average close to 1.70 and 1.90 mg O_2·g d.m.$^{-1}$ respectively (Table 1). This means that after 12 weeks of the process, the intensive decomposition of easily biodegradable organic matter and the related development of microorganisms slowed down, and the composts had a greater proportion of mineral fractions, attesting to the stabilization of this organic material (Kilian & Macedowska-Capiga, 2011). This is also highlighted by the comparison of the obtained results with the guidelines developed for the processing of the biodegradable fraction of municipal waste, according to which the limit value for stabilized waste is AT_4 <10 mg O_2·g d.m.$^{-1}$ (Siemiątkowski, 2012). Moreover, the respiratory activity is always connected with moisture content in feedstock (Villaseñor et al., 2011). In this study low AT_4 level was observed when dry matter content was ~80%, which did not provide an optimal environment for the further development of microorganisms. The obtained values are also consistent with the own research conducted previously on a technical scale, during which, in the spring and summer period, turned piles of bio-waste with sewage sludge obtained AT_4 below 2 mg O_2·g d.m.$^{-1}$ after 43th day of the process (Stegenta et al., 2019).

Electrical conductivity (EC) is the most frequently mentioned parameter of compost samples (Agnew & Leonard, 2003), indicating total salt/electrolyte content in this organic material (Epstein, 2017). Here, EC was higher for traditional compost (537.00 μS·cm^{-1} compared to 450.00 μS·cm^{-1} for vegan sample, Table 1). Similar values were also obtained for pruning waste compost (Benito et al., 2006), however, in most studies, the conductivity range for waste materials is much higher, reaching from 1.1-1.9 mS·cm^{-1} in the case of yard waste compost (Campbell & Tripepi, 1991), through 2.8-4.6 mS·cm^{-1} for aerobic sewage sludge (García et al., 1991) up to 5.42 mS·cm^{-1} for separated cattle manure (Inbar et al., 1993), 7.8-9.8 mS·cm^{-1} for organic city refuse or even 7-12 mS·cm^{-1} for urban refuse composts (García et al., 1991; Villar et al., 1993), respectively. The tested traditional and vegan composts with a significantly lower EC level showed no phytotoxic effect (section 3.3), which is in line with the suggestion of (Epstein, 2017), that the limit value of non-toxic impact on plants is 5 mS·cm^{-1}. This is also in assent with the results obtained by Ringer et al. (1997). In this research, the extract from manure compost showed an EC of 0.7-1.5 mS·cm^{-1}, while having no phytotoxic effects in the soil. The analyzed samples also fall within the range indicated by the authors.

3.2 Heavy Metals Content

The content of heavy metals (Ni, Cd, Cr, Pb) in the compost and their possible release into the soil during its application as a fertilizer raise the greatest doubts about the ecological effect of this organic material, especially in the case of its excessive use in arable fields (Iwegbue et al., 2007). Danger resulting from the accumulation of heavy metals in plant tissues and the related contamination of the human and animal food chain has led in many countries to the development of strict guidelines for the use of composts, specifying limit values for the level of these elements in the material (Iwegbue et al., 2007). However, certain heavy metals are essential elements for plant growth as micronutrients, although they show very low levels in soil under natural conditions (Rosen & Chen, 2014). That is why it is so important to control the product of the composting process in terms of the content of these elements and thus create optimal conditions for the development of crops.

Traditional and vegan compost did not differ greatly in their heavy metal content; the largest difference between the samples was noted for nickel (6.06 and 4.20 mg·kg d.m.$^{-1}$, respectively, Table 2). Traditional compost also showed a higher concentration in the case of lead, but with the minor difference (11.76 mg·kg d.m.$^{-1}$ in case of the first and 11.55 mg·kg d.m.$^{-1}$ for the latter). Of the four non-nutrient heavy metals measured, lead was also the element having the largest content in the produced composts; cadmium was the least recorded, with levels close to 3 mg·kg d.m.$^{-1}$ for both materials. Comparing the obtained values with the legal requirements for composts in Poland (The Ministry of Agriculture and Rural Development, 2008), none of the tested heavy metals exceeded the permissible values, indicating that both types of compost are ready to be used as an organic fertilizer (Table 2). Moreover, the content of Ni, Cd, Cr and Pb in the vegan and traditional compost samples were much lower than those reported by other researchers. Green waste (GWC) and municipal solid waste (MSWC) composts analyzed by (Farrell et al., 2010) were characterized by high levels of lead, nickel and chromium; in the case of MSWC they exceeded, respectively, approx. 77x, 14x and 7.5x the maximum values obtained in this study, while for GWC, despite lower pollution levels, the exceedances were 3.4x, 5.2x and 5.8x, respectively. A similar trend can also be observed in the case of the Ni and Pb content studied by (Kalamdhad & Kazmi, 2009b) in three types of produced compost. The values for the first element reported by the authors ranged from 18.9 to 39 mg·kg^{-1}, while in the case of lead from 19.2 to 142 mg·kg^{-1}.

The biggest difference that was noted between the two types of compost in the case of Ni can be related to the granulometric composition of both materials. The higher content of this heavy metal in traditional compost, char-

TABLE 2: Heavy metals content.

	Vegan compost, mg·kg d.m.$^{-1}$	Traditional compost, mg·kg d.m.$^{-1}$	Limit value, mg·kg d.m.$^{-1}$ (The Ministry of Agriculture and Rural Development, 2008)
Ni	4.20	6.06	60.00
Cd	3.03	2.87	5.00
Cr	6.42	6.25	100.00
Pb	11.55	11.76	140.00

acterized by a higher proportion of fine fractions (<3.15 mm), is consistent with the reports by (Kupper et al., 2014). In the conducted research, the authors concluded that the content of heavy metals in the material sieved to the fraction <20 mm is higher when comparing the results with unsieved material. Additionally, the results obtained by (Paradelo et al., 2011) and (Veeken & Hamelers, 2002) agree with this. However, the outcomes reported here do not follow the trend observed by (Qiao & Ho, 1997) that Ni changes to a more accessible form at lower pH; traditional compost, for which the content of this metal was higher at the same time was characterized by higher pH than in the case of vegan material.

The obtained differences in the content of heavy metals between vegan and traditional compost could also be influenced by the substrates used in their production. The higher levels of chromium in the plant-based material were probably associated with the high proportion of vegetables. Heavy metals are accumulated in the tissues of these plants, both in their edible and inedible parts (Guerra et al., 2012). This is related to the uptake of these elements by crops from the soil, but also from the air - sediments can form a layer of pollutants directly on plant tissues (Zurera-Cosano et al., 1989). Related problems are reported by researchers around the world who are warning of the need to monitor the heavy metal content of vegetables and fruits, especially with the reference to vegetarian and vegan diets based on these products (Guerra et al., 2012; Islam et al., 2007). This need may be justified by the results obtained by (Guerra et al., 2012), who during their research noticed exceeding the permissible Cr content in 44.2% of the tested vegetables, mainly in green ones. In turn, in the analysis conducted by (Islam et al., 2007) it was observed that Cd can accumulate both in roots and in shoots of Chinese cabbage, winter greens and celery. The vegan compost produced here was based on the use of vegetables, including the types mentioned by the authors, which may explain the increased concentrations of Cd and Cr compared to traditional compost.

When considering traditional compost, a higher Pb content might be associated with the addition of animal waste, such as dairy products (yoghurt, cheese) and eggshells. Dairy products, although not containing many heavy metals in its primary form (milk) may be contaminated with them during product processing and packaging (Enb et al., 2009); a possible reason for the presence of elements such as Ni or Pb in dairy products may also be the supply of previously contaminated feed to dairy animals (Abou-Arab, 1997). According to a study of (Enb et al., 2009) on heavy metals behavior during processing of milk products such as yoghurts made from buffalo's and cow's milk, lead is one of the main contaminating elements. This is also confirmed by the analysis of (Shahbazi et al., 2016), who found Pb not only in yoghurt, but also observed a significant content in the case of white cheese. The authors explained this relationship by the presence of casein, a milk protein that tends to bind Pb. A similar effect was noted by (Meshref et al., 2014); the lead content in their milk and dairy products samples exceeded the permissible limit in all cases. Apart from dairy products, the presence of eggshells may also be responsible for the higher Pb level in traditional compost. (Waegeneers et al., 2009) concluded that the major source of this element in eggs is contaminated soil ingested by chickens; the average concentration of Pb in fresh eggs may be even 10x higher than in the case of the content of Cd (Fu et al., 2014). However, the problem of heavy metals content in eggshells is not fully understood; most authors claim that different paths of contamination should be checked before defining the main source (Grace & MacFarlane, 2016).

3.3 Physicochemical Composition

The issue of nitrogen transformation in the waste mass during the composting process is discussed, in particular, due to the possible associated emission of harmful gases, such as NH_3 and N_2O, but also due to the loss of this element during the decomposition of organic matter and the reduction of the fertilizing properties of the final product (de Guardia et al., 2010). In the conducted tests, a higher total nitrogen content was found in traditional compost (38.15 mg N·dm^{-3} compared to 27.30 mg N·dm^{-3} for vegan samples, Table 3). However, for both types of materials, nitrate nitrogen was predominant over organic and ammonium nitrogen, reaching for traditional and vegan compost 34.69 N_{NO3}·dm^{-3} and 21.93 mg N_{NO3}·dm^{-3}, respectively. Considering the Kjeldahl nitrogen for both types of material, it

TABLE 3: Physicochemical composition of vegan and traditional composts.

Indicator	Vegan compost	Traditional compost
Total nitrogen, mg N·dm^{-3}	27.30	38.15
Kjeldahl nitrogen, mgN·dm^{-3}	5.01	3.44
Organic nitrogen, mg N_{org}·dm^{-3}	4.37	3.15
Ammoniacal nitrogen, mg N_{NH4}·dm^{-3}	0.64	0.29
Nitric nitrogen, mg N_{NO3}·dm^{-3}	21.93	34.69
Nitrite nitrogen, mg N_{NO2}·dm^{-3}	0.36	0.02
Phosphates, mg P·dm^{-3}	11.15	8.61
Total phosphorus, mg P·dm^{-3}	13.82	9.36
Dissolved oxygen, mg O_2·dm^{-3}	7.50	7.80
Total dry residue, mg·dm^{-3}	450.00	490.00
Dry mineral residue, mg·dm^{-3}	230.00	200.00
Dry organic residue, mg·dm^{-3}	220.00	290.00
General dissolved substances, mg·dm^{-3}	415.00	460.00
Dissolved mineral substances, mg·dm^{-3}	225.00	190.00
Dissolved organic substances, mg·dm^{-3}	190.00	270.00
General suspensions, mg·dm^{-3}	35.00	30.00
Mineral suspensions, mg·dm^{-3}	5.00	10.00
Organic suspensions mg·dm^{-3}	30.00	20.00
Sulphates, mg SO_4·dm^{-3}	20.60	19.70
Chlorides, mg Cl·dm^{-3}	7.60	14.80
Sodium, mg Na·dm^{-3}	1.90	7.40
Potassium, mg K·dm^{-3}	95.80	70.60
Magnesium, mg Mg·dm^{-3}	7.30	11.20

can be observed that approx. 85% and 90% was organic nitrogen for composts based on plant substrates and plant plus animal origin, respectively. This could be explained by the activity of microorganisms which, initially carried out the ammonification reaction, then immobilized the formed dissolved ammonium and, by using it as a nitrogen source and the associated transformation, returned it as organic nitrogen (Sánchez-Monedero et al., 2001). This is in line with the observations of (Li et al., 2013) who noted an increase in NH_4-N content in the initial stage of composting (day 3 and 4 of the analysis), which quickly turned into a decline until the end of the process. In turn, the high content of nitrate nitrogen in the analyzed total nitrogen of the composts produced may indicate the occurrence of favorable conditions for the activity of nitrifying bacteria during the 12 weeks of the process, such as temperature below 40 °C and adequate access to oxygen (Sánchez-Monedero et al., 2001). A similar ratio of N_{NO3} to N_{NH4}, where the former significantly exceeded the amount of the latter, was also noted in the analysis of (Bueno et al., 2008). The observation of such a trend in traditional and vegan composts highlights the possibility of their effective use in agriculture, due to the best bioavailability of this form of nitrogen for the plant root system (Sánchez-Monedero et al., 2001). For both types of materials, the lowest share was nitrite nitrogen (approx. 1.3% for vegan compost and 0.05% for traditional compost), which is consistent with the observations mentioned earlier (Li et al., 2013).

Phosphorus is one of the key elements influencing plant development and quality; due to its scarcity, the yields of 30-40% of arable land in the world are below optimal (Malhotra et al., 2018). Its comprehensive activity is based on its participation in every stage of plant development. The content of this element plays an important role in seed germination, where grains with initially more P help the seedlings grow and mature faster (Y.-G. Zhu & Smith, 2001). Additionally, it contributes to cell enlargement and division, influencing plant height or the number and area of leaves (Assuero et al., 2004). Thanks to P, plant reproduction is also increased, resulting from the stimulation of flower and seed formation (Malhotra et al., 2018). However, composting, apart from increasing the availability of this element for plants, may also be responsible for the environmental risk associated with its leaching (Hashimoto et al., 2014), based mainly on the eutrophication of water bodies (Eneji et al., 2003) or erosion. It is therefore necessary to control the P content during decomposition of organic matter in the compost to ensure sustainable distribution practices for this element.

Among the tested samples, vegan compost showed a higher content of both phosphates and total phosphorus, while for the second factor the difference between the composts was greater (13.82 mg P·dm^{-3} compared to 9.36 mg P·dm^{-3} for the traditional material, Table 3). The obtained values were much lower than those recorded by other researchers, which, as in the case of material moisture, can be explained by the relatively long duration of the composting process and the progressive loss of organic phosphorus through mineralization and its microbiological consumption (Haug, 1993) or leaching. The values indicated in the literature are several dozen times higher, but in the case of composting of vegetable waste with cattle manure and sawdust lasting 20 days, the difference is smaller than for the same substrates within 15 days (approx. 3.5 g·kg^{-1} and 3.8 g·kg^{-1}, respectively (Kalamdhad & Kazmi, 2008; Kalamdhad & Kazmi, 2009a), which may indicate the dependence of the P content on the duration of the process and its progressive decrease. This is also confirmed by the research conducted by (Wei et al., 2015), where the availability of P showed a decrease from 44% to 36% after the thermophilic phase of composting in each of the six prepared composts (made of pig and chicken manure, municipal solid waste, green waste, straw and fruits and vegetables waste). This decrease was related to the presence of available phosphorus in the treated waste (water soluble P, Olsen P, citric acid P); at the same time, an increase in the content of moderately available and inaccessible variant of this element was observed (from 48% to 59%). Low P content in vegan and traditional compost probably related to its transformation into a more resistant and less soluble fraction, as (Eneji et al., 2003) confirmed in the analysis, which leads to the conclusion that longer maturing compost, compared to fresh material, may limit the spread of P impurities after its application.

Vegan compost was characterized by a higher content of total suspended solids, with more organic solids (1:6 ratio of mineral to organic suspended solids), which was also visible in the case of traditional compost (ratio 1:2, Table 3). The maximum value recorded for the material based on plant waste (35.00 mg·dm^{-3}) did not exceed the values indicated in the literature (approx. 3x and 9x higher results obtained by Gupta & Doherty, 1990 for poultry litter compost). Contrary to the total suspended solids, the traditional compost was characterized by a higher total dry residue (490.00 compared to 450.00 mg·dm^{-3} for vegan compost) and total dissolved solids (460.00 and 415.00 mg·dm^{-3}, respectively). However, a similar trend was noted for both groups, where vegan compost is characterized by a higher proportion of mineral, and the traditional material – organic compounds.

Among the analyzed sulphates, chlorides, sodium, magnesium and potassium, vegan compost was characterized by a higher content of sulphate and potassium (Table 3). This may be an important aspect taking into account its fertilizing nature and the supply of these nutrients to soil poor in these compounds. Sulphates are important biogenic elements in the primary metabolism of plants; they take part in many basic processes in their cells, including photosynthesis and the metabolism of carbon and nitrogen (Droux, 2004). Similarly, K plays many roles inside the plant (including cell turgor building and cell membrane potential control) and is present in a number of its activities, being responsible, among others, for the regulation of stomatal aperture or leaf movements (Nieves-Cordones et al., 2016). Therefore, it is worth noting that despite the vegan nature of the compost, the content of sulphate and potassium did not decrease, contrary to the experience of (Storino et al., 2016) who attributed an increase in these ions to the presence of meat in the composted waste. On the other hand, in the case of Na and Cl there was agreement with the authors' observations; for these elementsthe greatest difference between the samples was noted (values of about 3.9x and 1.9x higher for traditional material). It is related to the substrates of the traditional compost the meat, poultry, and eggs that build it, which are considered to be one of the three main groups of sodium-contributing food (Engstrom et al., 1997). Much of this is due to the widespread use of sodium chloride in meat processing to improve its taste, texture and shelf life (Ruusunen & Puolanne, 2005). In addition, animal-based material had a higher Mg content than vegan compost, which in turn can be explained by the lack of eggshells in the latter. Magnesium, right after Ca and carbonate, are their main inorganic component (Cusack et al., 2003), and they enrich the final product of the composting process.

3.4 Phytotoxicity

Phytotoxicity is considered to be one of the most important elements in assessing compost's quality and usefulness for the potential use of it as a plant support agent in agriculture and horticulture (Barral & Paradelo, 2011). Biological tests based on the use of plants with rapid germination and growth, such as *Lepidium sativum L.*, are used to assess the overall impact of compost on the plant, resulting from a number of possible factors such as heavy metals, salt, contaminants and other phytotoxic factors (Emino & Warman, 2004).

In comparison to the control sample, which was the reference soil soaked with distilled water, traditional and vegan compost stimulated the growth of *Lepidium sativum* roots, which reached the length longer by an average of 6.36 and 5.08 mm (Table A.2, Supplementary content). When analyzing the effect of water extracts from composts on plant germination, it was noted that in the case of vegan compost all the seeds used have grown up; a lower and thus equal result was obtained for the traditional compost and the control sample (93.33%). The difference between plant- and animal-based material may be due to their different conductivity; the ions present in the compost may influence the regulation of the osmotic potential and the redox potential, thus influencing seed germination (Ibrahim, 2016). According to (Zhu et al., 2016) the high concentration of salt ions has an inhibitory effect on this process, which may happen with traditional compost with a higher EC value. The lack of phytotoxic effects of compost from plant waste is also consistent with the observations of (Gavilanes-Terán et al., 2016). Inhibition of the seed germination can also be a a result of nitrogen imbalance, when it is not in equilibrium (Song et al., 2021). A significant difference in nitric nitrogen levels between the vegan and traditional compost samples were noted here (21.93 and 34.69 mg N_{NO3}·dm^{-3}, respectively, Table A.2); at the same time other nitrogen forms were at similar level, which could contribute to the problem with germination.

The trend towards a 100% germination efficiency of plants from the use of compost has already been reported in the literature (Ait Baddi et al., 2004; Jouraiphy et al., 2008; Yangui et al., 2009). As in the case of vegan compost, the researchers observed the highest level of germination in the final product, thus recording the lowest phytotoxicity at the end of the process (He et al., 2009), including the *Lepidium sativum* used in this study (Selim et al., 2011). Researchers associated this effect with the compost cooling phase (2nd and 3rd month of composting), during which not only the number of phytotoxic lipids is reduced, but also the amount of stable organic matter and humic substances increases (El Fels et al., 2014). The humic acid fraction is indicated as the one that incorporate and immobilize phytotoxic substances such as heavy metals and pesticides through sorption and complexation (Smith, 2009; Zucconi et al., 1985).

4. CONCLUSIONS

Due to its pro-environmental character, composting has become a technology perceived as one of the basic tools of the circular economy, where the amount of directed waste has recently increased (Ros et al., 2006). Along with the growing trend of using a vegan diet observed in more and more countries it can be expected that the popularity of this method of kitchen and food waste processing will not decrease, and will also develop towards home composting. However, to ensure the environment friendly nature of this process, it is important to control the quality of the final product in order to provide valuable fertilizer material that can be used for home-garden purposes, and to introduce a household circular economy.

Plant-based kitchen waste without the addition of animal-origin product has been proven to be a valuable substrate to produce vegan compost during home and backyard composting. Studies have shown that vegan compost meets the criteria of an organic fertilizer and does not contain increased concentrations of heavy metals. Due to the lack of phytotoxic effects and to the stimulation of plants' growth, it can be successfully and safely used for their cultivation. Compared to the traditional compost, it is also characterized by a higher content of phosphorus, which is crucial for plant development, as well as by lower leachability of nitrates. Respiratory activity of this material after 12 weeks of the process indicates its good stability. In addition, it does not tend to rot anaerobically, which eliminates any problems associated with odors. Moreover, the process does not require a technological system or technologically advanced composters. Considering the content of cadmium and nickel which verged on the limit values, it is recommended that the share of vegetable waste from the cruciferous and celery families shoud not be dominant and, at the same time, it is suggested to use the substrates that are not too fragmented.

This preliminary research proved that vegan households, restaurants, and cafes are not disqualified from implementing a circular economy by using kitchen waste as a secondary material. However, it is worth noting that due to the possible increase in the amount of vegan waste in the coming years, this analysis should be extended to complete monitoring and controling the potential environmental impact of the vegan composts used, not only in terms of their impact on the crops grown, but also on such elements of the environment as soil and groundwater.

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SUPPLEMENTARY CONTENT

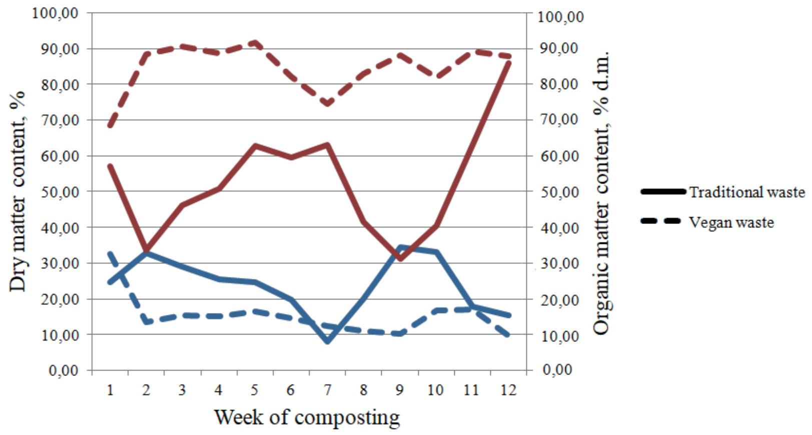

FIGURE A.1: Properties of traditional and vegan waste added to backyard composting; blue colour – dry matter content, red – organic matter content, % d.m. (dry matter).

TABLE A.1: Determination methods of water extracts physicochemical composition.

Indicator	Determination method
pH	PN-EN ISO 9963-1:2001 (Polish Committee for Standardization, 2001a)
Conductivity, $\mu S \cdot cm^{-1}$	PN-EN 27888:1999P (Polish Committee for Standardization, 1999a)
Turbidity, Nephelometric Turbidity Units (NTU)	PN-EN ISO 7027-1:2016-09 (Polish Committee for Standardization, 2016)
Total nitrogen, mg $N \cdot dm^{-3}$	The sum of all forms of nitrogen
Kjeldahl nitrogen, mg$N \cdot dm^{-3}$	PN-EN 25663:2001 (Polish Committee for Standardization, 2001b)
Organic nitrogen, mg $N_{org} \cdot dm^{-3}$	Indirectly (difference between Kjeldahl and ammonium nitrogen)
Ammoniacal nitrogen, mg $N_{NH4} \cdot dm^{-3}$	PN-C04576-4:1994 (Polish Committee for Standardization, 1994)
Nitric nitrogen, mg $N_{NO3} \cdot dm^{-3}$	PN-C-04576-08:1982 (Polish Committee for Standardization, 1982)
Nitrite nitrogen, mg $N_{NO2} \cdot dm^{-3}$	PN-EN 26777:1999 (Polish Committee for Standardization, 1999b)
Phosphates, mg $P \cdot dm^{-3}$	ISO 6878/1:2006 (International Organization for Standardization, 1986)
Total phosphorus, mg $P \cdot dm^{-3}$	PN-EN 1189-2000 (Polish Committee for Standardization, 2000)
Dissolved oxygen, mg $O_2 \cdot dm^{-3}$	PN-EN ISO 5814:2013-04E (Polish Committee for Standardization, 2013a)
BOD_5, mg $O_2 \cdot dm^{-3}$	PN-EN 1899-1:2002 (Polish Committee for Standardization, 2002, p. 2002)
COD_{Cr}, mg $O_2 \cdot dm^{-3}$	PN ISO 15705:2005 (Polish Committee for Standardization, 2005)
Total dry residue, mg$\cdot dm^{-3}$	PN-EN 12880:2004 (Polish Committee for Standardization, 2004a)
Dry mineral residue, mg$\cdot dm^{-3}$	Weight loss after ignition at 550°C
Dry organic residue, mg$\cdot dm^{-3}$	Indirectly, difference between dry total residue and dry organic residue
General dissolved substances, mg$\cdot dm^{-3}$	By weight, after evaporation of the filtered sample
Dissolved mineral substances, mg$\cdot dm^{-3}$	By weight, after roasting at 600°C
Dissolved organic substances, mg$\cdot dm^{-3}$	Indirectly, difference between general solutes and organic solutes
General suspensions, mg$\cdot dm^{-3}$	PN-EN 872:2007 (Polish Committee for Standardization, 2007)
Mineral suspensions, mg$\cdot dm^{-3}$	Indirectly, difference between total suspensions and organic suspensions
Organic suspensions mg$\cdot dm^{-3}$	By weight, after roasting at 600°C
Sulphates, mg $SO_4 \cdot dm^{-3}$	PN-ISO 9280:2002 (Polish Committee for Standardization, 2013b)
Chlorides, mg $Cl \cdot dm^{-3}$	PN-ISO 9297:1994 (Polish Committee for Standardization, 2004b)
Sodium, mg $Na \cdot dm^{-3}$	Atomic emission spectrometry (AAS, according to standards provided by Spectro-Lab Ltd.)
Potassium, mg $K \cdot dm^{-3}$	
Magnesium, mg $Mg \cdot dm^{-3}$	
Nickel (Ni), mg$\cdot$kg d.m.$^{-1}$ (dry matter)	
Cadmium (Cd), mg$\cdot$kg d.m.$^{-1}$	
Chromium (Cr), mg$\cdot$kg d.m.$^{-1}$	
Lead (Pb), mg$\cdot$kg d.m.$^{-1}$	

TABLE A.2: Phytotoxicity of composts.

	Vegan compost	Traditional compost	Reference soil (control sample)
Seed germination, %	100.00	93.33	93.33
Root elongation inhibition, %	-6.19	-7.75	–
Average root length, mm	87.13 ± 5.87	88.41 ± 7.29	82.05 ± 4.45

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VOLATILE ORGANIC COMPOUNDS FROM GREEN WASTE ANAEROBIC DEGRADATION AT LAB-SCALE: EVOLUTION AND COMPARISON WITH LANDFILL GAS

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ABSTRACT

The chemical composition of volatile organic compounds (VOCs) in landfill gas from municipal waste (MW) landfills primarily depends on the type of degrading waste. To provide first insights into the relationship between VOC chemistry (in landfill gas) and specific waste components, a lab-scale experiment on anaerobic digestion (AD) of green waste (GW) was carried out. The composition of the released C_{4+} VOCs was semi-quantitatively determined and indirectly compared to that generally expected for the overall MW landfill gas. The generated biogas from degrading GW during AD time showed up to 29 different VOCs, mainly including terpenes, followed by alkanes, alkenes, cyclics, aromatics, and halogenated compounds. O- and S-substituted compounds were sporadically detected. Overall, speciation and total concentration of VOCs fluctuated over AD time, likely due to changes in microbial populations and metabolism, as well as substrate depletion, during the AD evolution. As expected, VOC speciation in the GW biogas was lower than that of the typical MW landfill gas, since the latter is generated by a large variety of organic and synthetic waste components following different sequential degradation processes. These results highlighted that, when disposed of at MW landfill sites, the specific GW component may (i) potentially concur to the overall odour charge and toxic effects of MW landfill gas and (ii) detrimentally impact the energy exploitation of MW landfill gas by releasing terpenes, aromatics, and halogenated compounds.

1. INTRODUCTION

Landfilling has long been the prevalent practice for municipal waste (MW) management (treatment and disposal) worldwide (Duan et al., 2021; Kaza et., 2018). Once deposited in landfill, MW generates landfill gas as the result of the direct volatilization of compounds contained in the waste (e.g., plastic foam, air fresheners, detergents, etc.) and the progressive degradation of organic matter mainly based on anaerobic digestion (AD) processes (Chiriac et al., 2011; Christensen et al., 2010; Rasi et al., 2011). MW landfill gas, primarily consisting of CH_4 and CO_2, includes small fractions (normally at concentrations < 1% by vol.) of a wide range of non-methane volatile organic compounds (hereafter, VOCs) (Tassi et al., 2009), having a potential impact on air quality, local population's welfare, and landfill worker health if released to the environment (Nair et al., 2019). VOCs may also damage gas engines, reducing the functional life of equipment and the energy recovery performance (Giraudet et al., 2014; Sevimoĝlu and Tansel, 2013; Shin et al., 2002). Moreover, CH_4 oxidation occurring within landfill cover soils may decrease in presence of significant amounts of VOCs, resulting in a reduced mitigation of CH_4 release to the atmosphere (Alvarez-Cohen and McCarty, 1991; Alvarez-Cohen and Speitel, 2001).

Owing to its biodegradability and relatively high methane yield (Campuzano and González-Martínez, 2016), the organic fraction of municipal waste (OFMW) represents a significant challenge for landfill management. In fact, the OFMW (i) positively contributes to the conversion of landfill gas to energy by increasing its methane content but (ii) negatively contributes to the possible emission of toxic and pollutant organic trace gases into the environment. Nota-

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bly, the OFMW is by far the dominating fraction of MW accounting for almost 50% of the generated waste worldwide (Zamri et al., 2021) and landfilling has remained the main method for its management (Chen et al., 2020). Particularly, the specific component of OFMW represented by the green waste (GW), which includes grass clippings, hedge cuttings, pruning, leaves, and wood from private gardens, public parks, and roadside greenery, has been increasing in recent years with the growth of urbanization and related urban green spaces (Langsdorf et al., 2021; Reyes-Torres et al., 2018). For example, in Beijing (China) the amount of generated GW has increased by about 50,000-100,000 ton year^{-1} (Zhang and Sun, 2016), while an increase of 2 million ton occurred in the USA during the period from 2010 to 2018 (USEPA, 2020). Moreover, a relevant amount of 10.53 million ton of GW was destined for landfilling in the USA in 2018 (USEPA, 2020). Even in territorial areas with a considerable level of separately collected MW destined for recovery, as in the Marche Region (Central Italy), where it was equal to 70.5% in 2019, the GW still represents, with food waste and fines, the largest portion of the residual MW, accounting for 30.8% as overall organic fraction (Boccarossa et al., 2021). In the representative Marche Region, both the mechanically separated outputs from the residual MW are destined for landfilling, either directly in the case of the oversize stream or following only a partial aerobic biostabilization in the case of the undersize stream (Boccarossa et al., 2021).

In the last decades, several studies have been focused on the chemical characterization of VOCs in landfill gas to (i) identify potential odorous or pollutant compounds, (ii) verify the achievement of quality standards required to the optimized energy recovery, and (iii) assess the landfill life-cycle impact perspective (e.g., Allen et al., 1997; Beylot et al., 2013; Nair et al., 2019; Papadias et al., 2012; Parker et al., 2002; Sadowska-Rociek et al., 2009; Saral et al., 2009; Takuwa et al., 2009). On the contrary, to the best of our knowledge, only Staley et al. (2006) have focused on the characterization of VOC compositional features from few, selected waste components (paper, yard waste, and food waste) under anaerobic conditions. However, a deep analysis on the temporal evolution of VOC composition seems to be lacking. The chemical composition of the primary MW landfill gas compounds is known to be the result of the combination of the organic and synthetic waste components disposed of at a given landfill and the extension and evolution of the occurring biodegradation. However, the chemical features of VOCs released from different deposited waste components under anaerobic conditions, over time, have remained poorly known.

In this study, an AD experiment was conducted at lab-scale using GW, a key organic component of MW, as substrate to simulate the anaerobic degradation that typically occurs in landfills with the aim of investigating the VOC release in the time evolution and in the relation with potential sources. Semi-quantitative analyses of a wide range of VOCs (C_{4+}) in the biogas generated during the AD of GW were repeatedly carried out. The specific objectives of this study were to (i) compare indirectly the VOCs released at lab-scale from GW AD with those generally expected from a MW landfill and (ii) explore the potential contribution of deposited GW on MW landfill gas composition in terms of VOCs.

2. MATERIALS AND METHODS

2.1 AD experiment

2.1.1 Substrate and inoculum

The considered GW used as substrate was obtained from the collection center of the Fano town district in the Marche Region that is located at the pertaining landfill operated by the "ASET" public multi-utility group. At this facility, first the GW was visually separated from impurities (such as plastic, paper/cardboard, glass, and metal). Then, the GW was shredded by a mobile grinder and further reduced by an electric blender at the laboratory to favor the substrate uniformity in size to a few millimeters (Raposo et al., 2011).

To the purpose of the experiment, an inoculum consisting of anaerobic sludge (hereafter, INO), obtained from the anaerobic digestion treatment stage at a municipal wastewater treatment plant, was added to provide initial microbial community and improve the stability of the experimental system (Carchesio et al., 2014, 2020).

Both the substrate and inoculum were characterized in terms of moisture, total solids (TS), and volatile solids (VS), the latter being an indirect measure of the organic matter content (Tatàno et al., 2015). Moisture and TS contents were determined by loss-on-drying at 105°C, while VS content was measured by loss-on-ignition at 550°C (IRSA, 1984). The considered properties are shown in Table 1. GW moisture and VS contents fell within the ranges reported in the literature as representative for the physico-chemical quality of green waste (i.e., 21.1-67.9% FM - fresh matter - and 30.0-98.9% TS, respectively: Reyes-Torres et al., 2018). As far as INO is concerned, the resulting moisture content was at the lower limit of the range of 96.5-98% FM derivable in Londong (2006) as indicative for a well anaerobically digested sludge, while VS content remained at the upper limit of the range of 30-60% TS reported for digested sludge (Acaia and Ragazzi, 1991).

2.1.2 Experimental setup

Two parallel AD reaction lines were implemented using a mixture of GW with INO (hereafter, just GW line) and only INO as blank control, respectively. The substrate to inoculum ratio in the mixture was fixed at 2:1 on a VS basis (Carchesio et al., 2020). The AD experiment, which lasted 49 days, was performed in batch mode with two lab-scale digesters consisting of a dark glass bottle (inner volume: 1 L) equipped with a pierceable rubber cap and filled up

TABLE 1: Properties of the substrate and inoculum.

	Green Waste (GW)	Inoculum (INO)
Moisture (% FM)	49.9	96.5
Total Solids, TS (% FM)	50.1	3.5
Volatile Solids, VS (% TS)	67.1	60.2

FM = fresh matter

to approximately 0.5 L (Figure 1a). The operating temperature was set to mesophilic conditions (38°C) (Carchesio et al., 2020) and the digester contents were homogeneously mixed using heating magnetic stirrers at speed of roughly 300 rpm (Figure 1a). Small amounts of Na_2CO_3 powder (around 1 g) were initially added to both digesters to raise the pH up to approximately 8 and prevent the inhibition of AD process in the early phase of the experiment (Carchesio et al., 2020). The pH values in the GW digester evolved in time respecting the range of 6.0–8.3, where the anaerobic digestion is expected to occur (Angelidaki and Sanders, 2004). To avoid high overpressure in the digester headspaces, biogas was periodically evacuated in both GW and INO digesters, particularly after each dedicated biogas sampling.

From each digester, eight biogas samples were periodically collected (precisely, at 2, 7, 12, 16, 21, 33, 40, and 49 days after AD started). The biogas samples were collected inserting a needle, connected to a syringe through a three-way-valve, in the digester headspace from the pierceable rubber cap (Figure 1a). Thus, the biogas samples were transferred and stored into glass vials (Labco Exetainer®, 12 cm^3), equipped with a pierceable rubber cup, using an inlet and outlet needle to insert the samples and rinse deionized water filled vials, respectively (Figure 1b).

2.2 Chemical analysis

Semi-qualitative analysis of VOCs (C_{4+}) was performed using solid phase microextraction method (SPME) to pre-concentrate VOCs (Arthur and Pawliszyn, 1990) and a Thermo Trace Ultra gas-chromatograph (GC) equipped with a Thermo DSQ quadrupole mass spectrometer (MS) as analytical instrument. The SPME fibre, constituted of divinylbenzene (DVB)-carboxen-polydimethylsiloxane (PDMS), was inserted into the vials through the pierceable rubber and exposed to the gas for 30 min at 20°C. Thus, adsorbed VOCs were released, exposing the SPME fibre for 2 min at 230°C in the injection port of the Thermo Trace Ultra GC. Analytes were separated by employing a film thickness TR-V1 fused silica capillary column (30 m x 0.25 mm, inner diameter = 0.25 µm) and He as carrier gas (1.3 mL min^{-1}). The column operated at three temperature ramps: 35°C (held 10 min) to 180°C (held 3 min) at rate of 5.5°C min^{-1} and then to 230°C (held 6 min) at rate of 20°C min^{-1}. Finally, analytes were detected by the quadrupole mass spectrometer that operated in electron ionization mode (EI), with ionization energy of 70 eV and source temperature set at 250°C, and in full scan mode (35 to 400 m/z). VOC species were identified by the retention time of chromatographic peak and comparing the mass spectra with those reported in the NIST05 library (NIST, 2005). A relative quantitative analysis of VOC compounds was carried out as follows: the areas of the peaks measured in each sample were normalized to the lowest peak area (represented by the symbol "+"), determining five classes with increasing area ranges, each one represented by an increasing number of "+" symbol. For example, a VOC compound labelled with three "+" symbols showed a peak area approximately three times higher than the lowest peak area range. The relative quantities of VOCs pertaining to the same functional group were summed and the resulting percentage of each functional group was computed according to the sum of "+" symbols of the VOCs in each sample (hereafter, % ΣVOCs).

3. RESULTS

The VOC compounds identified during AD time of GW and INO digesters, respectively, are listed in Table 2. Up to 29 different VOCs were detected in the GW biogas samples, including: alkanes (5), alkenes (5), aromatics (4), cyclics (2), terpenes (5), halogenated (3), S-substituted compounds (1), as well as compounds pertaining to four different O-substituted classes (i.e., ketone, alcohol, ether, and aldehyde) (Table 2). Differently, gas samples from the INO AD at every sampling time showed only five VOCs (i.e., methylpentane, hexane, trimethylpentene, methylcyclopentane, and cyclohexane) pertaining to alkanes, alkenes, and cyclic groups (Table 2).

In the individual GW biogas samples, the number of measured VOC species ranged from 12 to 25 (Figure 2). In particular, the number of different VOC species showed a peak in the initial stage of AD (25), decreased during the next two weeks (18), then increased again up to 25 at day 21 and, subsequently, decreased to 12 (Figure 2). In gener-

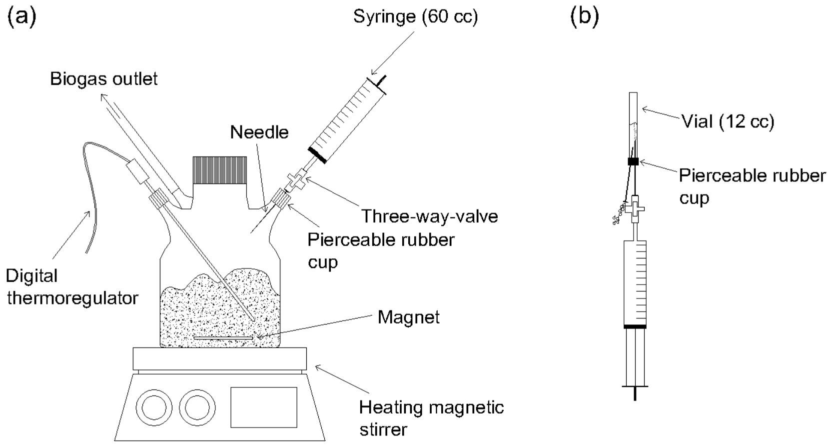

FIGURE 1: Experimental equipment used for the AD of GW and INO (a) and biogas sampling scheme (b).

TABLE 2: Summary of semi-qualitative analysis of VOC (C_{4+}) composition in GW and INO biogas samples during AD time.

	Day 2		Day 7		Day 12		Day 16		Day 21		Day 33		Day 40		Day 49	
	GW	INO	GW	INO	GW	INO	GW	INO	GW	INO	GW	INO	GW	INO	GW	INO
Alkanes																
Methylpentane	++	+	++	+	++	+	++	+	++	+	++	+	++	+	++	+
Hexane	+++	+	+++	+	+++	+	+++	+	+++	+	+++	+	+++	+	+++	+
Dimethylhexane	+															
Methyloctane	+		+		+		+		+		+		+		+	
Nonane	+		+		+		+		+		+		+		+	
Alkenes																
Methylbutene	++		++		++		++		++		++		++		++	
Hexene	+		+		+		+		+							
Methylpentene	+		+		+		+		+							
Trimethylpentene	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+
Dimethylhexene	+		+													
Aromatics																
Toluene	+		+		+		++		+++		+		+		+	
Ethylbenzene									+							
Xylene									+							
Styrene	+								+							
Halogenated																
Trichloroethylene	+								+							
Chlorohexane	+		+		++		+		++		+		+		+	
Trichloromethylpropane	+				+		+		+							
S-Substituted																
Dimethylsulfide	+															
Cyclics																
Methylcyclopentane	++	+	++	+	++	+	++	+	++	+	++	+	++	+	++	+
Cyclohexane	++	+	++	+	++	+	++	+	++	+	++	+	++	+	++	+
Terpenes																
α-Pinene	+++++		+++++		+++++		+++++		+++++		+++++		+++++		+++++	
Camphene	+++		+++		+++		+++		+++		++					
β-Pinene	+		+		++		++		++		+					
Carene	+		+		+		+		++		+					
Limonene	++		++		++		+++		+++		++		++		+	
O-substituted																
Ethylhexanol									+							
Butoxyethanol									+							
Dimethylcyclohexanone	+															
Benzaldehyde	+								++							

"+" = detected; empty space = not detected.
The number of symbols "+" indicates the magnitude of selected peak area ranges (i.e., greater number of "+" means larger peak area).

al, VOC variety in GW biogas samples was mainly provided by terpenes, alkenes, and alkanes (up to 5 different compounds for each group) (Figure 2). In the last period of GW AD, the number of VOC species of the former two groups dropped to 2 (Figure 2). These pair of compounds were detected at all GW AD sampling times among terpenes and alkenes (α-pinene and limonene, and trimethylpentene and methylbutene, respectively) (Table 2). Differently, at least four alkanes were detected during the GW AD time (i.e., methylpentane, hexane, methyloctane, and nonane) (Table 2). The prevalent speciation of aromatics (4 and 2) and halogenated (3) was observed in combination with the overall peaks in the number of species (Figure 2), whereas among these VOCs only toluene and chlorohexane were detected during the whole GW AD time (Table 2). Similarly, two cyclic VOCs (i.e., methylcyclopentane and cyclohexane) were constantly detected during the whole GW AD time (Table 2). Instead, two and three oxygenated compounds occurred at days 2 and 21 of GW AD, respectively (Figure 2). Finally, dimethylsulfide (DMS) was detected only at day 2 of GW AD (Table 2).

Concerning the relative quantities of VOC functional groups, substantial variations were observed during the GW AD (Figure 3). In particular, terpene group dominated up to day 40 (ranging from 30 to 41% ΣVOCs) and then decreased approaching the GW AD end (27% ΣVOCs) when on the contrary alkanes became dominant (Figure 3). However, α-pinene was by far the most abundant VOC detected during the whole GW AD (Table 2). Alkanes and alkenes (dominated by hexane and methylbutene, respectively: Table 2), representing large relative fractions of VOCs (overall ranging from 16 to 32% and from 11 to 19% ΣVOCs, respectively), showed the lowest relative abundance at day 21 of GW AD when on the contrary the highest relative abundance of aromatics (overall ranging from 3 to 13% ΣVOCs) and oxygenated compounds (5 and 9% ΣVOCs at day 2 and 21, respectively) occurred (Figure 3). Cycling compounds, consisting of comparable fractions of methylcyclopentane and cyclohexane (Table 2), globally ranged from 9 to 18% ΣVOCs and showed particularly an increasing trend approaching the GW AD end after day 21 (Figure 3). Some variations of relative abundances of halogenated group (overall ranging from 3 to 9% ΣVOCs) were observed during GW AD time (Figure 3). DMS found at day 2 of GW AD accounted for 3% ΣVOCs (Figure 3).

With final regard to the INO AD, the related biogas samples regularly consisted of 40% ΣVOCs each of alkanes and cyclic compounds and 20% ΣVOCs of alkenes.

4. DISCUSSION

Lab-scale AD tests are known to be a reliable tool to assess the anaerobic biodegradability and the biochemical methane potential of selected organic substrates (Carchesio et al., 2020; Raposo et al., 2011). In this study, the AD experiment was conducted to simulate, under an-

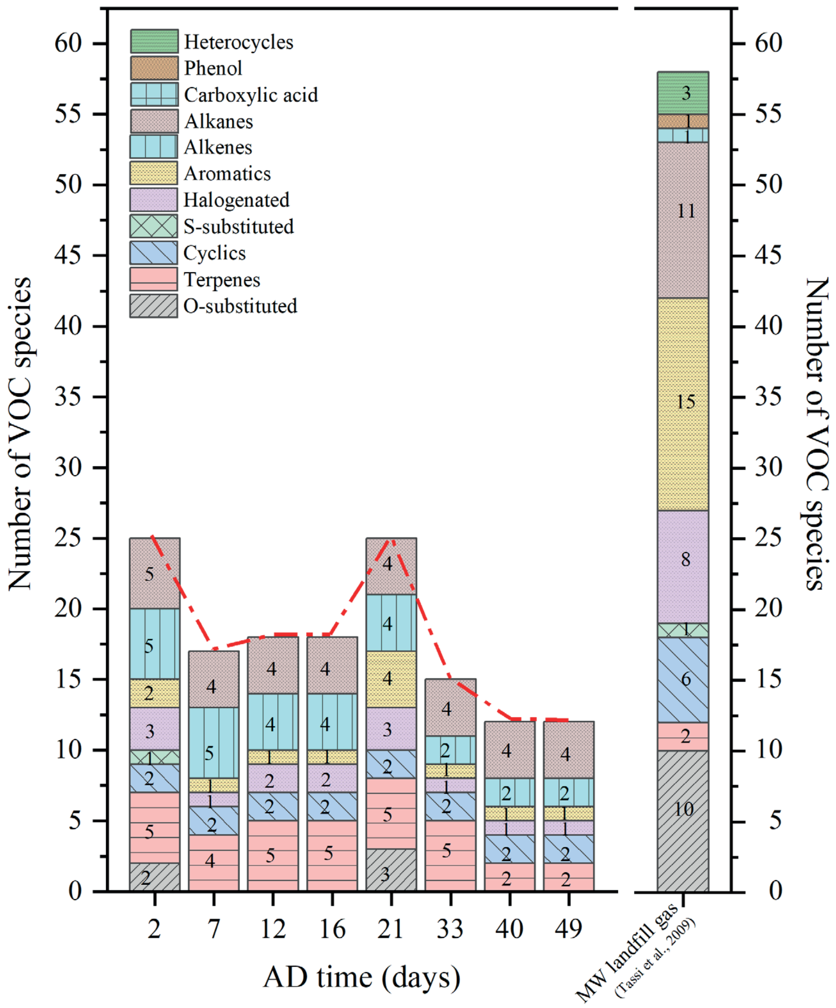

FIGURE 2: Stacked column diagram showing the evolution of the number of species in the VOC functional groups detected in GW biogas during AD time and the number of species in the VOC functional groups in a typical MW landfill gas (Tassi et al., 2009). MW landfill gas column includes both the pertaining recovery wells 1 and 2 because they correspond. Dashed red line shows the total VOC species number.

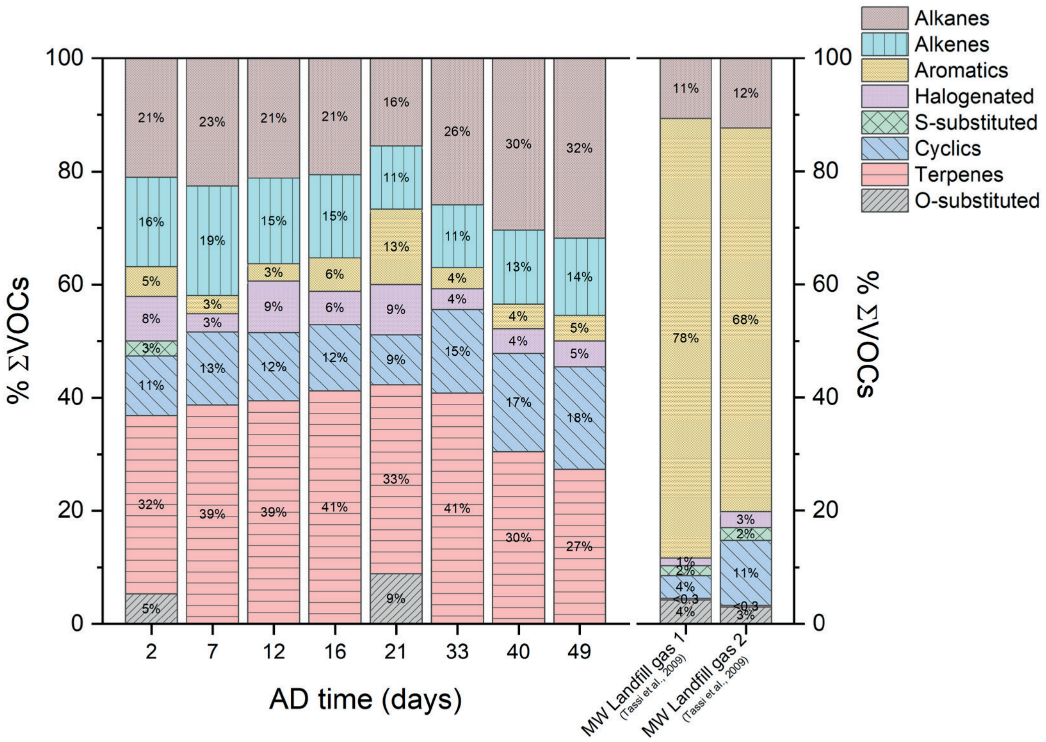

FIGURE 3: Stacked column (100%) diagram illustrating the amount of each VOC functional group in GW biogas during AD time expressed as % ΣVOCs (as indicated in Section 2.2). The relative amounts of VOC functional groups in MW landfill gas from pertaining recovery wells 1 (MW landfill gas 1) and 2 (MW landfill gas 2) (Tassi et al., 2009) were also reported and expressed as % ΣVOCs. In both MW landfill gases, terpenes, heterocycles, phenol, and carboxylic acid accounted for < 0.3% ΣVOCs.

aerobic condition and in a limited temporal scale, the digestion of a key component of MW as the short-term analogue of its landfilling (Bogner and Lagerkvist, 1997), with the aim of investigating the consequential release of VOCs. Effectively, the AD represents the predominant and long temporal phase of MW degradation expected within the landfill body and promoted by the development of anaerobic bacteria consortia (Christensen et al., 2010). Basically, the AD converts the organic matter into small molecules and finally into the main gaseous components of CO_2 and CH_4 through four sequential stages: (i) hydrolysis, (ii) acidogenesis, (iii) acetogenesis, and (iv) methanogenesis (Li et al., 2014, 2019).

4.1 VOC composition and sources in GW biogas

GW biogas samples were characterized by large relative abundance of terpenes (Figure 3), particularly dominated by α-pinene (Table 2). On the other hand, terpenes are strictly related to degradation of yard trimmings, vegetable food, and fruits (Komilis et al., 2004; Maulini-Duran et al., 2014; Wang and Ting, 2008). Generally, compounds pertaining to this functional group, including monoterpenes and sesquiterpenes, are synthesized by plants (as defence against biotic and abiotic stressors and attractor for pollinators) and stored in tissues, leaves, needles, and resins (Gershenzon and Dudareva, 2007; Mewalal et al., 2017; Ormeño et al., 2008). Terpenes are also identified in some fruits and vegetables (Aharoni et al., 2004; Muchlinski et al., 2020). The production of terpenes depends on plant species, and it changes in function of diverse vegetative components such as roots, leaves, and barks (Courtois et al., 2012, 2009). For example, conifer trees such as Pinus, Picea, and Abies predominantly emit α-pinene, β-pinene, camphene, and limonene (Bowman et al., 1997; Kopaczyk et al., 2020). Accordingly, high contents and variety of terpene species released from the experimental GW AD were likely dictated by the expected mixture of the investigated GW and the related decaying rates of the diversified matrices. It is worth noting that terpenes are known to be highly odorous compounds (Wenjing et al., 2015) as they have low odor thresholds (Gallego et al., 2012). The odor threshold identifies the charge of selected gaseous compounds or mixtures as the lowest concentration which generally produce a sensorial response to the human sense of smell (Gallego et al., 2012). In particular, the concentration of α-pinene detected by olfactometry is about two and one order of magnitude (in terms of $\mu g\ m^{-3}$) lower than camphene and limonene, respectively (Gallego et al., 2012). Moreover, α-pinene threshold is about three times lower than hexane (Gallego et al., 2012). Considering the relatively high abundance of α-pinene detected in the GW biogas samples (Table 2), this compound may play a fundamental role on the odorous charge of GW biogas. Moreover, high amounts of terpenes are known to be detrimental for materials normally employed in biogas collection and exploitation systems (such as pipelines, valves, and pumps) and to affect the removal efficiency of impurities (Arrhenius and Engelbrektsson, 2016; Bragança et al., 2020).

Short-chain alkanes and alkenes constituted a relevant fraction of the overall GW biogas composition (Figure 3).

Particularly, they may be related to the degradation of long-chain aliphatic compounds that constitute cuticular wax coating the surface of plant leaves, stems, and flowers (Eglinton and Hamilton, 1967; Mitra et al., 2020; Sharma et al., 2018). The relatively high contents of methylbutene revealed in the alkene group (Table 2) may be attributed to the reactivity and degradability of longer-chain alkenes. Differently, the resulting dominance of hexane, known to be potentially toxic, among alkanes (Table 2) may be ascribed to the relatively high stability of C_6 structure (Capaccioni et al., 1995; Randazzo et al., 2020).

Aromatics and halogenated compounds, which may pose potential concerns to the environment and human health (Dincer et al., 2006) as well as to the biogas exploitation having detrimental effects on engine systems (de Arespacochaga et al., 2014; Piechota, 2021), were detected in the GW biogas (Figure 3). Firstly, aromatic compounds were likely produced by the biodegradation of lignin, a natural polymer enveloped inside the lignocellulosic biomass that consists of hemicellulose and cellulose (Liew et al., 2012; Lubbers et al., 2019; Zimmermann, 1990). For example, it is known that styrene may be produced by decarboxylation of cinnamic acid, a precursor organic acid for lignin and flavonoid biosynthesis (Mooney et al., 2006; Salvador et al., 2013). On the contrary, no evidence was found for halogenated compounds as constituent of plant and vegetal tissues. However, it is worthy mentioning that both aromatic and halogenated compounds may be assimilated by plants from contaminated air (Cape, 2003; Keymeulen et al., 1995; Mosaddegh et al., 2014) and then released after death. Consequently, the detected presence of halogenated compounds and partly of aromatics in the GW biogas may be considered of xenobiotic origin.

Cyclic compounds were detected in all biogas samples from GW AD (Figure 2). On the other hand, leaf waxes are covered by a barrier formed by different alicyclic compounds (Andrae et al., 2019; Chen et al., 2020; Staiger et al., 2019) which may be consequently released during degradation processes.

As expected, limited variety and relative amount in oxygenated compounds were found in only two GW biogas samples (Figures 2 and 3). Typically, these compounds are intermediates or by-products of aerobic decomposition processes (Li et al., 2021; Mustafa et al., 2017; Wu and Wang, 2015). Consequently, they are efficiently produced only at oxidizing conditions, e.g., during the alternative composting process. Nevertheless, some O-substituted compound species have been recognized to occur also at anaerobic conditions, deriving from both degradation of fresh waste and direct volatilization of compounds contained in waste (Duan et al., 2021; Staley et al., 2006; Zheng et al., 2020). Particularly, benzaldehyde (Table 2), a highly odorous compound (Gallego et al., 2012), can be produced by enzymatic hydrolysis of some fruit kernels (Verma et al., 2017).

As far as finally DMS is concerned, its initial detection in GW AD (Table 2) may derive from decomposition of organic waste containing sulfur (Wu et al., 2020; Zheng et al., 2020).

4.2 Temporal evolution of VOC composition in GW biogas

Significant variations in VOC composition and relative abundance of functional groups were revealed during GW AD (Figures 2 and 3).

Initially, a first peak in VOC species (25) was observed in the GW biogas at day 2 of AD, including different compounds of aromatics, DMS, halogenated and O-substituted compounds (Figure 2). Effectively, DMS, aromatics, O-substituted and halogenated compounds are normally emitted and volatilized during fresh waste degradation (Duan et al., 2014; Zhao et al., 2015). Moreover, high amounts of oxygenated compounds were detected during the acidogenesis (Chiriac et al., 2011), suggesting that this AD stage was dominant at day 2. From day 7 to day 16, no significant changes in VOC features were observed (i.e., terpenes, alkanes, and alkenes remained the dominant groups) (Figure 2), implying a certain stability of the AD process. The second peak in number of VOC species (25), observed at day 21 (Figure 2), possibly derived from changes in microbial metabolism. It is known that microbial diversity and activity are sensitive to variations in physico-chemical conditions during AD time (e.g., variations in nutrients, particularly ammonia content) and shifts among different AD stages (Fitamo et al., 2017; Mao et al., 2019; Pan et al., 2021), metabolizing and releasing different VOC species (Zhang et al., 2020). However, the release of gas pockets trapped within the GW substrate cannot be excluded as suggested by the occurrence of xenobiotic compounds pertaining to aromatic and halogenated compounds (see Section 4.1). The decrease in variety of VOC species observed during the last temporal stage of the GW AD may be ascribed to the gradual GW substrate depletion as highlighted by the drastic reduction of terpene species (from 5 at day 33 to 2 at conclusive day 49) (Figure 2).

Methyloctane, nonane, methylbutene, toluene, chlorohexane, α-pinene, and limonene were steadily detected in all GW biogas samples and not appeared in the INO gas samples (Table 2). Consequently, they could be considered as markers of the GW AD. On the contrary, methylpentane, hexane, trimethylpentene, and each cyclic compound were also found in the INO gas samples (Table 2). However, in GW biogas the relative abundance of hexane was steadily higher than methylpentane, trimethylpentene, methylcyclopentane and cyclohexane, while their relative abundances were comparable in INO gas (Table 2). This suggests that a significant portion of hexane was direct released from the GW anaerobic degradation.

α-pinene and hexane were the most abundant species in each GW biogas sample and showed an overall relative enrichment in the AD time (Table 2) implying a relatively high release rate of these compounds from the investigated GW regardless of the evolution of AD stages. Instead, the other terpene compounds revealed in GW biogas were dependent on the proceeding of the AD experiment: in particular, the terpenes/alkanes ratios were 1.5 at the beginning of GW AD (day 2), then increased to > 1.5 from day 7 to day 33, when methanogenesis was expected (Brown et al., 2012; Liew et al., 2012), and finally dropped to ≤ 1 at the conclusive stages of AD when only α-pinene and limonene were found. The previous conditions suggest that the release of terpene species from GW AD was more affected by type and rate of biochemical process and substrate depletion than alkanes. Finally, the tendency of increasing alkanes/alkenes ratios over AD time was also observed, indicating a relative depletion of alkenes likely due to the progressive exhaustion of the GW substrate.

4.3 Comparison of GW biogas and MW landfill gas

In order to perform indirectly a coherent comparison between GW biogas and MW landfill gas, the chemical composition of MW landfill gas determined by Tassi et al. (2009) was specifically considered, according to: (i) analogous geographic, climatic, and socio-economic context (i.e., both the considered MW landfill and GW collection center are located in Central Italy); and (ii) common analytical method. Particularly, Tassi et al. (2009) reported compositional data of MW landfill gas from two recovery wells including a wide variety of 58 VOCs (C_{4+}) (Figure 2). In the considered MW landfill gas, aromatics showed the highest VOC speciation (15), followed by alkane (11), O-substituted (10, including 2 aldehydes, 3 esters, and 5 ketones), halogenated (8), and cyclic (6) compounds (Figure 2). Finally, heterocycles (3), terpenes (2), S-substituted compound (1), phenol (1), and carboxylic acid (1) were also revealed, whereas no alkenes (C_{4+}) were detected (Figure 2). In the considered MW landfill gas, aromatics constituted relatively the most abundant VOC group, followed by alkanes, cyclics, and O-substituted compounds (Figure 3). Significant contents of halogenated and S-substituted compounds (i.e., dimethylsulfoxide) were relatively observed in the considered MW landfill gas, while terpenes, heterocycles, phenol, and carboxylic acid occurred up to 0.3% ΣVOCs (Figure 3).

The lower speciation and relative abundance of terpenes in MW landfill gas characterized by Tassi et al. (2009) (i.e., only α-pinene and eucalyptol were identified with resulting incidence < 0.3% ΣVOCs), as compared with the GW biogas generated and characterized in the present experimental study, may likely be dictated by (i) large terpenes loss by volatilization in the early phases of degradation of deposited waste or (ii) relatively low presence of deposited waste containing terpenes. On the other hand, the AD experiment in the present study was conducted in a confined and controlled volume, avoiding biogas losses and using a terpenes-rich substrate (see Sections 2.1.2 and 4.1). Generally, terpenes in MW landfills may derive from both biodegradation of green and food wastes, and direct volatilization of industrial substances contained in household detergents and air fresheners (Duan et al., 2021; Allen et al., 1997). In particular, α-pinene and limonene are generally the dominant terpene species in MW landfill gas (Duan et al., 2021), indicating that they are easily released during degradation processes of organic and synthetic waste.

The relatively high speciation of alkanes recognized in MW landfill gas (Figure 2) was caused by the enrichment of long-chain alkanes (C_{7+}). Presumably, the large variety of long-chain alkanes (C_{7+}) detected in MW landfill gas (Tassi et al., 2009) was ascribed to the degradation of plastics, frequently disposed of at landfills, releasing a large variety of high molecular weight alkanes (Meunmee et al., 2016). Similarly, the higher variety of aromatic compounds in MW landfill gas than GW biogas (Figure 2) may be caused by the direct release of these compounds from anthropogenic waste commonly disposed of at landfills such as solvents, petroleum-derived products, and plastics (Duan et al., 2021). Indeed, toluene was a dominant aromatic in both MW landfill gas (Tassi et al., 2009) and GW biogas (Table 2), suggesting that this compound can be easily released by degradation processes regardless waste composition. However, MW landfill gas showed alkanes/aromatics ratio significatively lower (about 0.16) than those revealed in GW biogases (ranging from 1 to 8). On the other hand, aromatics in GW biogas are related to the decomposition of the recalcitrant lignin and the possible release as xenobiotic compounds (see Section 4.1). Differently, in MW landfill they are also produced by the direct volatilization of compounds contained in deposited synthetic waste.

Similar to alkanes and aromatics, the relatively high speciation of cyclic compounds in MW landfill gas (Figure 2) was ascribed to the wide range of waste typologies deposited in MW landfill (Tassi et al., 2009).

Although alkenes (C_{4+}) may be released during degradation of plastics such as packaging and films (Muenmee et al., 2016) commonly disposed of at MW landfills, they are not or scarcely found in MW landfill gas (Allen et al., 1997; Tassi et al., 2009). Presumably, the expected overall landfill conditions disadvantage the presence of alkenes, since these compounds are extremely reactive in an aerobic environment (Li et al., 2021). Differently, the anaerobic conditions imposed in the adopted lab-scale system (see Section 2.1.2) permitted the accumulation of alkenes in the GW biogas (Figure 2).

Halogenated compounds in MW landfill gas (Tassi et al., 2009) markedly differed from those identified in GW biogas (Figure 2). These VOCs are recalcitrant and normally considered anthropogenic contaminants contained in synthetic substances used in textile, chemical, pharmaceutical, and dry-cleaning industries (Randazzo et al., 2020). As a wide range of industrial products, such as solvents, paints, adhesives, and pesticides, are generally deposited with unsorted waste (Allen et al., 1997), relatively higher content and variety of halogenated compounds were expected in MW landfill gas than GW biogas. However, the presence of chlorohexane was revealed in all GW biogas samples (Table 2) while it was absent in typical MW landfill gas (Tassi et al., 2009). Presumably, the occurrence of chlorohexane during GW AD was related to the possible use of specific organochlorine pesticides eventually adsorbed, accumulated, and metabolized by the vegetal tissues (Gonzalez et al., 2003; Strandberg and Hites, 2001; Waliszewski et al., 2004)

The relatively low abundance and speciation of O-substituted compounds in GW biogas were likely dictated by the anaerobic conditions controlling the biogas production (Figures 2 and 3). In fact, oxygenated compounds are typically related to aerobic degradation processes occurring at the beginning of MW degradation in landfills (Liu et al., 2015; Nie et al., 2018), particularly in sites characterized by air permeation since biodegradation of alkanes to O-substituted compounds is more efficient (Randazzo et al., 2020).

S-substituted group in MW landfill gas consisted only in dimethylsulfoxide (DMSO: Tassi et al., 2009). Considering that DMS, found instead in GW biogas (Table 2), is unstable at oxidizing conditions (Taylor and Kiene, 1989), its absence in MW landfill gas may be caused by the possible occurrence of oxidation processes at aerobic conditions (Long et al., 2017; Wu et al., 2010) in the landfill body, particularly favoured by the permeation of meteoric water or atmospheric air.

Finally (Figure 2), the lack of phenols and heterocycles in the GW biogas agreed with the origin of these functional groups, being generally released from chemical products and intensely cooked foods such as meat and coffee, respectively (Randazzo et al., 2020), whereas carboxylic acids (i.e., intermediate hydrophilic products of anaerobic processes: Cao et al., 2021), were not detected likely because they were dissolved in fermentation broth (Duan et al., 2021).

5. CONCLUSIONS

The results of this study showed that terpenes dominated in both relative abundance (i.e., ranging from 27 to 41% ΣVOCs) and number of species (up to 5 compounds) the GW biogas and were strictly related to the specific waste component used as substrate for the AD experiment. Significant amounts and high speciation (up to 5 compounds) were exhibited by alkanes and alkenes accounting from 27 to 46% ΣVOCs as sum. The previous VOC groups were followed in the order by aromatics (up to 4 compounds), halogenated and O-substituted compounds (up to 3 each), cyclics (2 compounds), and S-substituted compound (1) that relatively represented, as sum, from 19 to 40% ΣVOCs. Alkanes, alkenes, cyclics, aromatics, DMS, and O-substituted compounds were to be ascribed to the degradation of plant tissues and other general vegetal organic components (e.g., fruits and flowers). Differently, a fraction of aromatics and halogenated compounds could be considered xenobiotic.

Overall, the temporal changes in number and amount of VOC species during GW AD may be caused by (i) subsequent AD stages induced by the progressive establishment of different microbial consortia, and (ii) degree of substrate conversion. Indeed, a comprehensive assessment of biodegradation processes occurring during GW AD requires complementary analyses such as main gas composition, isotopic composition of C-CO_2 and C-CH_4, and microbiological characterization.

Significative differences in VOC composition were indirectly observed between GW biogas and MW landfill gas, since MW landfill gas is expected to derive from a large variety of deposited waste components including a wide range of synthetic materials (such as solvents, plastics, paints, and dry-cleaning products). Moreover, MW landfill gas in the landfill body may be often (or at least initially) affected by oxidation processes related to air circulation possibly driven by permeating meteoric water. Differently, at the experimental lab-scale implemented in this study, whereas environmental and process conditions were fixed and steady and the investigated substrate was isolated in a closed digester, biogas oxidation was avoided or at least minimized.

The results of this study showed that the disposal of GW in MW landfills may concur to the odorous charge and potential toxic effects of the globally generated MW landfill gas, since it provides to landfill gas the content of terpenes and xenobiotic compounds pertaining to aromatic and halogenated groups. Moreover, significative charges of terpenes, aromatics, and halogenated compounds generable by the anaerobic degradation of GW may concur to negatively affect the energy exploitation of the landfill gas by potentially damaging gas collection, conveying, and engine systems. Indeed, these potential detrimental effects may be also expected in the alternative waste management option based on the possible bioenergy recovery through AD from the separately collected GW streams in biogas plants.

Future lab-scale experiments to be carried out using different waste components than GW, may be useful to predict the expected contributions (particularly in terms of VOCs) to the composition of landfill gas generated (and eventually released to the atmosphere) at MW landfill environments.

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LIQUID FLUIDS FROM THERMO-CATALYTIC DEGRADATION OF WASTE LOW-DENSITY POLYETHYLENE USING SPENT FCC CATALYST

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ABSTRACT

The widely-used plastics, especially low-density polyethylene (LDPE), have resulted in a considerable accumulation of plastics in the waste stream, causing a global environmental problem. Therefore, the research aims to examine the thermal and catalytic degradation of waste LDPE plastic using spent fluid catalytic cracking (FCC) catalyst and compare the properties of the produced liquid oils with commercial fuels. The potential of converting the most energy from waste plastics to valuable liquid oil, gaseous, and char was investigated. A batch reactor was used to thermally and catalytically degrade LDPE at temperatures 350 to 550°C and catalyst to plastic ratio of 0.10 to 0.25. The physical properties of the produced liquid oils, flash point, pour point, viscosity, API-gravity, carbon residue, density, etc., were determined using standard methods. We characterized the chemical properties of produced pyrolysis liquid oils with Gas chromatography-mass spectrometry (GC-MS). The liquid oil, gas, and char produced at catalyst to plastic ratio of 0.20 at 500°C were 92.7 wt.%, 6.1 wt.%, and 1.2 wt.% respectively. The thermal pyrolysis at 500°C gave 76.6 wt.%, 20.7 wt.%, and 2.7 wt.% for produced liquid oil, gas, and char, respectively. The GC-MS shows that the produced LDPE liquid oil contains many hydrocarbons from C_7-C_{29}. The major hydrocarbons common to LDPE are benzene, 1, 3 dimethyl benzene, and toluene. The produced liquid oil's properties compare favorably with that of commercial fuels.

1. INTRODUCTION

Plastic solid waste (PSW) is mainly composed of polyethylene (PE), polypropylene (PP), polystyrene (PS), polyvinyl chloride (PVC), and polyethylene terephthalate (PET). Polyethylene is made up of high-density polyethylene (HDPE), low-density polyethylene (LDPE), and linear low-density polyethylene (LLDPE). They account for about 40 percent of the PSW; hence it is the most common type of waste material in the waste stream (Onwudili et al., 2019). PSW causes serious environmental and waste management problems in Nigeria and other countries, including developed nations. These problems include environmental flooding, reduction in landfills space, water, and air pollution, blockage of side drains, increase in the spread of diseases, and an increase in the presence of insects and dangerous rodents (Aisien et al., 2013). However, the presence of PSW in enormous volumes in landfills, terrestrial and marine environments could be traced to its durability, less energy-efficient, inexpensive, non-biodegradable, and good applicability nature of plastics (Aisien et al., 2013). Aisien et al. (2021) reported that one of the most effective and efficient ways of managing PSW is pyrolysis. Sosa et al. (2021) and Aisien et al. (2021) stated that pyrolysis is an environmentally friendly sustainable plastic waste management process and is the most promising resource for producing commercial fuels (gasoline, diesel, kerosene). Patni et al., (2013) reported that the pyrolysis or degradation process significantly reduces plastic waste volume, lowers decomposition temperature, low capital cost, and increases recoverable energy by producing varieties of products. Plastic degradation can either be thermal or catalytic, depending on the operating temperatures and the use of catalyst. Aisien et al., (2021) reported that catalysts had been widely used in the pyrolysis process to optimize product distribution, increase product selectivity, upgrade pyrolysis products and improve hydrocarbon distribution. Besides, catalytic pyrolysis of plastics produces liquid oil whose properties are similar to that of commercial fuels (Sharuddin et al., 2016). Some researchers have studied

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thermal and catalytic pyrolysis of other waste plastics (Kunwar et al., 2018; Abbas-Abadi et al., 2013, 2014; López et al., 2017; Singh et al., 2018).

The catalytic pyrolysis of plastic waste using ZSM-5, red mud, zeolites Y, FCC, and others, produced pyrolysis oils whose properties compare favorably with commercial diesel fuel (Miandad et al., 2016; Hernandez et al., 2007). Moorthy Rajendran et al., (2020) reported catalytic pyrolysis of municipal plastic waste using HUSY, HZSM-5, HMOR, Zeolite Y, silica, and FCC catalysts produced quality fuels. The FCC catalysts are fine particles with an average size of 60 to 100 μm and a bulk density of 0.80 to 0.96 g/cm^3. Also, it has large pore sizes, high temperature, steam stability, high activity, and good resistance to attrition (Chen et al., 2019). Susastriawan et al., (2020) reported that at low temperature, the smaller the zeolite size and the higher the temperature, the higher the liquid fraction yield when zeolite catalyst was used to degrade polyethylene. Similarly, Onwudili et al., (2019) studied catalytic pyrolysis of a mixture of plastics using catalysts such as FCC, ZSM-5, and zeolites Y. They stated that the yield of the liquid fraction decreased with the addition of the catalyst. The liquid fraction had properties suitable for fuel, although the number of aromatic compounds presents increased. Similarly, Santos et al., (2018) reported that zeolite USY gave a higher amount of liquid fraction, and the major components were alkylbenzenes, naphthalenes, and olefins. Also, Kyong et al., (2003) stated that liquid oil yield from PP, PE (LDPE and HDPE), and PS pyrolysis was different.

This research is focused on the applications of low-medium temperature (350-550°C) and moderate catalyst to plastic ratio (0.1-0.25) on the degradation of LDPE. Therefore, the optimization of LDPE pyrolysis with respect to increase liquid oil yield and quality using low-medium temperature and moderate spent FCC catalyst has not been previously considered. However, the main essence of this research is to recover energy in the form of liquid oil from LDPE waste plastics and reduce this PE waste plastic and spent FCC catalyst in the environment. Hence, the study aims to investigate the thermal and catalytic degradation of waste LDPE plastic using spent FCC catalysts and compare the properties of the produced liquid oil with commercial fuels. The objectives include comparing thermal and catalytic degradation of LDPE at different temperatures and catalyst to plastic ratio. Besides, to compare the produced liquid oil properties from LDPE with commercial fuels and evaluate the composition of the liquid oil products.

2. MATERIALS AND METHODS

2.1 Materials

We collected the waste LDPE from the Irorere Obazee landfill at Evbuozegbo-Benin, Nigeria. We got the spent FCC catalyst from Warri refinery and petrochemical company, Warri-Nigeria. The surface area, pore volume, and average pore size of the spent FCC catalyst obtained are 63.42 m^2/g, 0.103 cm^3/g, and 7 nm respectively. We got the pyrolysis plant with an insulated jacketed 5 kg capacity batch pyrolysis reactor from the engineering workshop, University of Benin, Benin city. We purchased analytical grade chemicals from Sigma-Aldrich Co. Ltd. in the UK and Qualikems Fine Chemical Ltd. India.

2.2 Spent fluid catalytic cracking catalyst characterisation

The spent FCC catalyst samples were degassed at 200^{o}C for 8hr. under vacuum to remove any contaminants. The catalyst samples' surface area, pore volume, and pore size were determined from Braunauer-Emmett-Teller (BET) technique by nitrogen adsorption and desorption isotherms from the catalyst at equilibrium vapour pressure using the static volumetric method. The nitrogen adsorption– desorption isotherms were recorded at 77 K using a Micrometrics Tristar 3000. The surface area was determined by Brunauer-Emmett-Teller (BET) model (Brunauer et al., 1938), and Barrett-Joyner-Halenda (BJH) analysis (Gregg and Sing, 1982) was used to determine the pore volume, pore diameter, and pore size distribution.

2.3 Preparation of plastic materials

The waste LDPE plastics were washed with detergent and tap water to clean them. We dried the cleaned waste plastics with sunlight. We shredded, ground, and stored the ground waste LDPE plastics in a covered plastic container.

2.4 The pyrolysis studies

The experimental pyrolysis plant shown in Figure 1 was used in this study. It was designed, fabricated, and tested as described by Aisien et al., (2021). The thermal degradation process started with charging 1 kg of prepared LDPE plastics into the heating chamber of the pyrolysis reactor after evacuating it with a vacuum pump. The reactor was heated at 15°C/min until it reached the desired temperature. The temperature was regulated by employing K-thermocouple and PID controller. The produced gaseous products were made to pass through the outlet tube connected to the two ice-cold water glass condensers. The condensable liquid products and the solid residues were collected and weighted. The experiment was each conducted in triplicates for the temperature variations (350-550°C) studied. Mabood et al., (2010) formula in equations 1 to 3 was used to evaluate the yield of the products, and the calculation of the degradation products was based on the weight/mass after weighing each product on completion of each study. Standard analysis methods and GC-MS were used to analyze and characterize the physical properties of the produced liquid oil. A similar procedure was used to carry out catalytic degradation studies. The catalyst to plastic ratio varied between 0.1 and 0.25.

2.5 Analytical methods

The standard ASTM methods were used to evaluate the physical properties of the produced liquid oil. The GC-MS and ASTM distillation was used to analyse the pyrolysis liquid oil fraction's chemical composition and properties. The calorific value was determined using the bomb calorimeter.

2.6 Statistical analysis

The Kruskai-Walli H-test (Kruskai and Walli, 1952) or

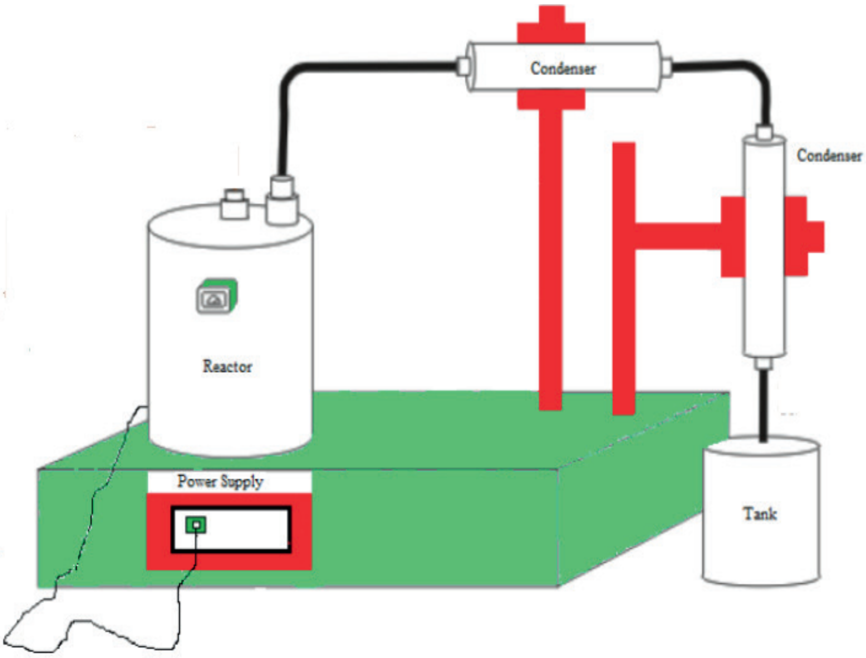

FIGURE 1: Schematic diagram of the pyrolysis process.

one-way analysis of variance by ranks using statistical package for social sciences (SPSS) version 20 with a significance level of p = 0.05 with n = 4 for statistical analysis of products yield data.

Liquid Yield:

$$Oil\ (wt.\%\ ) = \frac{Mass\ of\ oil}{Mass\ of\ LDPE}\ x\ 100\% \tag{1}$$

Residue (Char) Yield:

$$Char\ (wt.\%\ ) = \frac{Mass\ of\ residue}{Mass\ of\ LDPE}\ x\ 100\% \tag{2}$$

Gas Yield:

$$Gas\,(wt.\%) = 100\% - (Oil + Residue) \tag{3}$$

3. RESULTS AND DISCUSSION

3.1 The effect of temperature on the degradation of LDPE plastic

The effect of temperature on the yield of pyrolysis products (liquid oil, gas, and char) at temperature range 350 to 550^{o}C are shown in Figure 2. We observed that the yield of the produced liquid oil from LDPE gradually increased, while gaseous products decreased as the pyrolysis temperature increased from 350 to 500°C. The results show a significant difference ($p<0.05$) in the liquid oil and gases yields between 350 and 500°C. However, an increase in temperature to 550°C resulted in reduced liquid oil yield and an increase in gaseous product yield. The reduction in LDPE liquid oil yield at 550°C might be attributed to the fact that at 550°C, these plastics have almost wholly degraded (Chin et al., 2014; Marcilla et al., 2009; Onwudili et al., 2009). This observed trend is a function of the catalyst to plastic ratio.

Moreover, for the entire temperature variation, the char yield was minimal and decreased very slightly with temperature. There is no significant difference ($p>0.05$) in char yield with temperature. Figure 3 compares products yield with temperature for thermal and catalytic degradation of LDPE at 0.2 catalyst to plastic ratio. The T in Figure 3 represents the results of the thermal degradation of LDPE plastic. We observed that the LDPE products (liquid oil, gas, and char) yield for catalytic degradation were relatively higher than those from thermal degradation, independent of the prevailing temperature. There is a significant difference ($p<0.05$) in produced liquid oil and gas yields between thermal and catalytic degradation at 500°C. The maximum temperature for the optimization of liquid oil, gas, and char yields was 500°C. These observed results and trends were similar to those reported by Aisien et al., (2021), Marcilla et al., (2009), Onwudili et al., (2009), Jung et al., (2010), and Abbas-Abadi et al., (2013). However, some researchers reported reduced overall liquid oil yield and increased gas and char yields during pyrolysis of waste plastics (Abbas-Abadi et al., 2014; López et al., 2017; Demirbas, 2004).

3.2 The effect of catalyst to plastic ratio on the degradation of LDPE plastic

Figure 4 shows the effect of the catalyst to plastic ratio on the yield of LDPE pyrolysis products at different temperatures. As shown in Figure 4, we observed that at 350°C, the liquid oils yields from LDPE gradually increased, and the gas yields reduced as the catalyst to plastic ratio increased from 0.1 to 0.2. Also, a further increase of the catalyst to plastic ratio to 0.25 reversed this trend. A similar trend was observed with other temperatures, as shown in Figure 5. There is a significant difference ($p<0.05$) in the liquid oils and gases yields between 0 and 0.20 catalyst to plastic ratio. However, the char yield from LDPE degradation was minimal with no significant difference ($p>0.05$) as

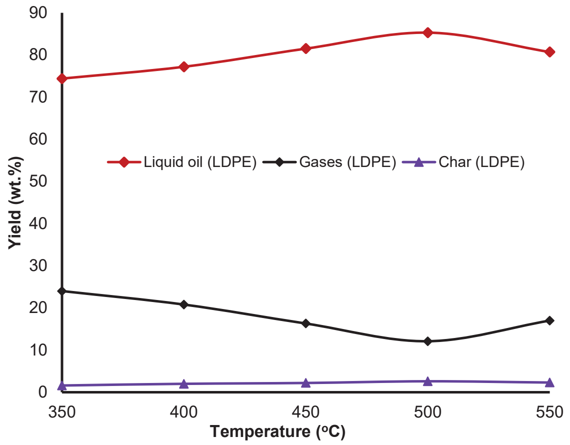

FIGURE 2: Variation of the products yield with temperature at 0.1 catalyst to plastic ratio.

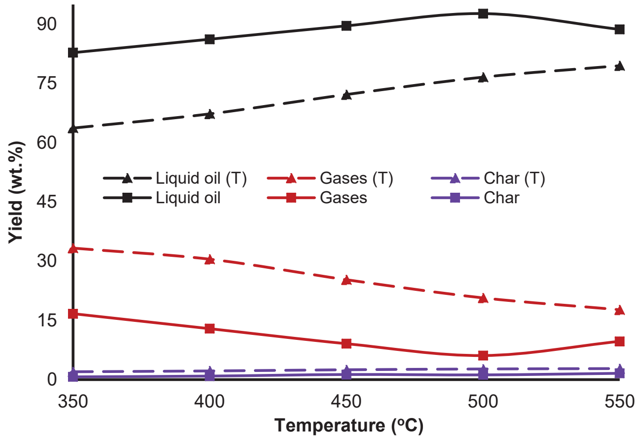

FIGURE 3: Comparing products yield with temperature for thermal and catalytic pyrolysis at 0.2 catalyst to plastic ratio.

the catalyst to plastic ratio increased from 0.1 to 0.25. A similar trend of results was reported by Aisien et al., (2021); Abbas-Abadi et al., (2014); Kyong et al., (2003, 2002); and Sharuddin et al., (2016). They reported a similar effect of spent FCC catalysts on the yield of the pyrolysis products. Therefore, we can state that the FCC catalyst enhanced decomposition or cracking of the LDPE plastic. However, a reverse trend was reported by Kunwar et al., (2016) and Miandad et al., (2016). They stated that catalyst causes an increase in the yield of the gaseous fraction and the amount of char and reduced liquid fraction.

Moreover, Aisien et al., (2021) and Budsaereechai et al., (2019) reported that an increase in the amount of catalyst to a higher concentration did not significantly affect the yield and fuel properties of produced liquid oil. This fact is also confirmed in the degradation of LDPE using FCC catalysts in this study. Hence, it seems unnecessary to use high FCC catalyst loading. Abbas-Abadi et al., (2014) and Shraruddin et al., (2016) reported that the FCC catalyst is one of the best catalysts for the optimization of liquid oil production from plastic pyrolysis and that spent FCC catalyst still has high catalytic performance with produced liquid oil yield above 80 wt.% for all plastics.

3.3 Effect of FCC catalyst on the products yield and residence time

The effect of FCC catalyst on the products yield and residence time from the pyrolysis of LDPE plastic at 350°C is shown in Table 1. We observed that the liquid oil yield from LDPE thermal degradation was lower than catalytic degradation. Besides, the gas and char yields from thermal degradation of LDPE were higher than catalytic degradation. The residence time for the thermal degradation of LDPE was higher than that of catalytic degradation. There is a significant difference ($p<0.05$) in the reduction in residence time between thermal and catalytic degradation of LDPE plastic. This shows that the spent FCC catalyst is very effective in accelerating the LDPE degradation reaction rate. Also, the results showed no significant difference ($p>0.05$) in residence time of LDPE degradation with an increase in catalyst to plastic ratio.

However, the residence time of catalytic degradation

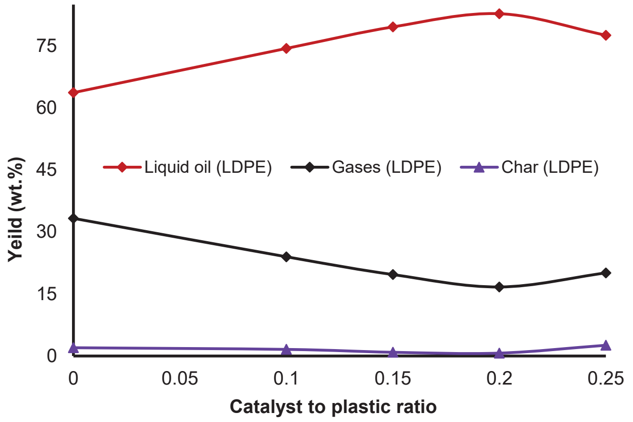

FIGURE 4: The variation pyrolysis products yield with catalyst to plastic ratio at degradation temperature of 350°C.

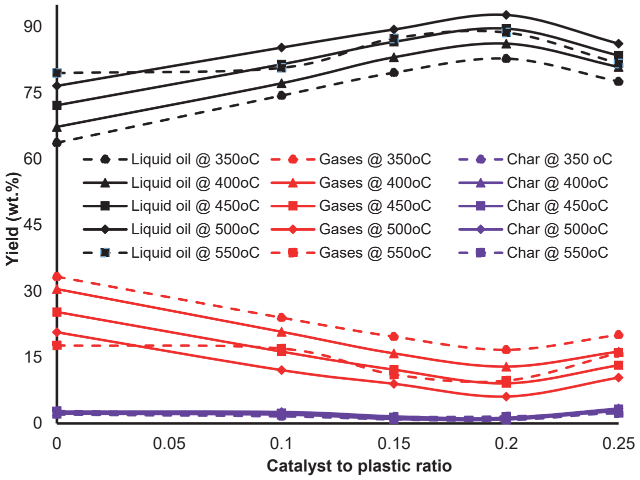

FIGURE 5: Comparing degradation products yield with catalyst to plastic ratio at different temperatures.

of LDPE reduced with an increased catalyst to plastic ratio. This might be due to the high surface area and acidity of the spent FCC catalyst. These results agree with other studies (Aisien et al., 2021; Abbas-Abadi et al., 2014; Kyong et al., 2003; Jung et al., 2010; Demirbas, 2004; Ahmad et al., 2014; Sakata et al., 1999; Costa et al., 2010).

3.4 The characteristics of produced liquid oil from LDPE degradation

3.4.1 Physical properties

Table 2 summarized the fuel properties (viscosity, flash point, API gravity, density, pour point, ash content, and the calorific value) of the liquid oil produced from LDPE degradation at different catalyst to plastic ratios. Table 2 also compares the LDPE produced liquid oil fuel properties with that of commercial fuels (gasoline, diesel, and kerosene). We characterized the produced liquid oil to determine its improved quality and suitability for energy generation and heating purposes. The results for the produced liquid oil from catalyst to plastic ratio of 0.1 are 2.43 cSt; 0.86 $g.cm^3$; 32.52°C; 43°F; -6°C; 0.02%; 0.62% and 41.8 $MJkg^{-1}$ for viscosity, density, flash point, API gravity, pour point, ash content, carbon residue, and calorific value respectively. A similar trend was observed for other catalysts to plastic ratios. Also, the results show that the fuel properties of the produced liquid oil from LDPE catalytic degradation were higher and of better quality than that from thermal degradation. Besides, the fuel properties of the produced liquid oil from LDPE catalytic degradation displayed irregular variation with an increase in catalyst to plastic ratio. Moreover, there is no significant difference (p>0.05) in the fuel properties of the produced liquid oil from LDPE with an increase in catalyst to plastic ratio. The fuel properties of the liquid oil from LDPE compare favorably with that of commercial fuels.

The summary of the produced liquid oil from LDPE with temperature variation is shown in Table 3. The results of produced liquid oil from LDPE at 350°C for viscosity, density, flash point, API gravity, pour point, ash content, carbon residue and calorific value are 2.47 cSt; 0.88 $g.cm^{-3}$; 31.8°C; 42°F; -7°C; 0.015%; 0.59% and 40.7 $MJkg^{-1}$ respectively. We observed a similar trend for other temperatures, and the fuel properties of the produced liquid oil from LDPE catalytic degradation show irregular variation with increased temperature. Besides, there is no significant difference (p>0.05) in the fuel properties of the produced liquid oil from LDPE with an increase in temperature. The results in Tables 2 and 3 were either close to or within the range reported for commercial fuels (Ahmad et al., 2014; Shakirullah et al., 2010) Saptoadi and Pratama, (2015). Hence, the values of the fuel properties of LDPE liquid oil determined are strongly comparable to that of commercial fuels. Shah and Jan (2014) explained that the fuel properties of the produced liquid oil compare favorably with the commercial fuels because the liquid oil contains some aromatic hydrocarbons. Santaweesuk and Janyalertadun (2017) reported that the small ash content values of the liquid oil show it is free from metal contamination and high molecular weight soot.

Therefore, we concluded that the fuel properties of the liquid oil from LDPE were independent of temperature or catalyst to plastic ratio. These results agree with some researchers' results (Bozbas, 2008; Gaurh and Pramanik 2018).

3.4.2 Chemical properties of the produced liquid oil from LDPE degradation

Figure 6 shows the comparison between the produced liquid oil from LDPE degradation at 400°C and commer-

TABLE 1: Products yield distribution and retention time for thermal and catalytic degradation of LDPE plastics at 350oC using a different catalyst to plastic ratio.

Catalyst to plastic ratio	Liquid oil (wt.%)	Gases (wt.%)	Char (wt.%)	Residence time (min.)
0	63.7	33.3	2	83
0.10	74.4	24	1.6	47
0.15	79.6	19.7	0.9	44
0.20	82.8	16.7	0.7	41
0.25	77.6	20.1	2.6	39

TABLE 2: Comparison of the physical parameters of the produced LDPE liquid oil at different catalyst to plastic ratio.

LDPE plastic at different catalyst to plastic ratio	Viscosity @ 40°C (cSt)	Density (g/cm^3)	Flash point (°C)	API gravity @ 40°F	Pour point (°C)	Ash (wt.%)	Carbon residue (wt.%)	Calorific residue (MJ/kg)
0.00	1.53	0.89	27.23	41	-7	0.01	0.58	41.4
0.10	2.43	0.86	32.52	43	-6	0.02	0.62	41.8
0.15	2.32	0.88	32.4	45	-6	0.01	0.61	42.5
0.20	2.47	0.87	31.8	42	-7	0.015	0.59	40.7
0.25	2.38	0.83	30.9	38	-5	0.03	0.60	41.3
Gasoline	0.775 -1.03	0.72 - 0.736	20.8-42	63.24-65.03	6	0.01	0.62	46.86
Diesel	2.0-5.3	0.83 - 0.85	55-60	34.97-38.98	-	-	0.6	42.51
Kerosene	0.9-2.2	0.78 - 0.82	50-55	41.06-49.91				45.46

TABLE 3: Comparison of the physical parameters of produced LDPE liquid oil at different temperature.

LDPE plastic @ different temperature (°C)	Viscosity @ 40°C (cSt)	Density (g/cm³)	Flash point (°C)	API gravity @ 40°F	Pour point (°C)	Ash (wt.%)	Carbon residue (wt.%)	Calorific residue (MJ/kg)
350	2.47	0.88	31.8	42	-7	0.015	0.59	40.7
400	2.50	0.89	32.2	43	-6	0.01	0.60	39.8
450	2.48	0.91	31.3	41	-7	0.01	0.61	39. 2
500	2.46	0.90	30.2	40	-5	0.00	0.60	38.1
Gasoline	0.775 -1.03	0.72-0.736	20.8-42	63.24- 65.03	-	-		46.86
Diesel	2.0-5.3	0.83-0.85	55-60	34.97-38.98	6	0.01		42.51
Kerosene	0.9- 2.2	0.78-0.82	50-55	41.06-49.91	-	-		45.46

cial fuels. The results show that the boiling point range for LDPE liquid oil lies between diesel and gasoline for 40-70% distillate recovery (Figure 5). This further affirms the suitability of LDPE liquid oil for energy production. These results agree with that of Graurh and Pramanik, (2018), who characterized pyrolysis liquid oil from polyethylene. They stated that the boiling point range gives information on the fuel's composition, properties, and behavior.

3.4.3 Chemical composition of the produced liquid oil from LDPE degradation

We determined the chemical composition of the produced liquid oil by GC-MS characterization (Shimadzu. 2011). The GC-MS results are represented in Table 4 and Figures 7 and 8. The results show that the liquid oil from LDPE degradation is complex hydrocarbons with carbon atoms ranging from C_7 to C_{29}. We observed that the LDPE produced liquid oil comprise paraffin (alkanes), olefins (alkene), naphthenes (cycloalkane), and aromatics classes. As shown in Figure 7, the results are paraffin (27.5%), olefins (50%), naphthenes (12.5%), and aromatics (10%) for produced LDPE liquid oil. Also, results shown that C_7-C_{10}, (37.5%); C_{11}-C_{15}, (35.5%); C_{16}-C_{20}, (12.5%) and >C_{20}, (12.5%) were got from LDPE liquid oil. Moreover, we observed that the liquid fraction products range as gasoline range (C_4-C_{12}), diesel range (C_{12}-C_{23}), kerosene range (C_{10}-C_{18}), and motor oil range (C_{23}-C_{40}). The results obtained are gasoline, (60%), diesel, (45%), kerosene, (52.5%), and motor oil, (7.5%) for LDPE liquid oil. This report is in agreement with that of Jung et al., (2010). However, from the perspective of high percentage of olefins in the produced liquid oil when compared with commercial oils, the quality of the liquid oil is lower than that of commercial oils.

Figure 8 shows the GC-MS plots of LDPE produced liquid oil at optimal conditions of 500°C and 0.2 catalyst to plastic ratio. The composition of the LDPE-produced liquid oil is in Table 4. We observed that the liquid oil from LDPE degradation consists of many different hydrocarbons. The significant LDPE- produced liquid oil compounds include 1,3 dimethyl Benzene, Toluene, Naphthalene, 1-methyl, Indene 1,12-Tredecadiene, Cis-9-Tetradecadiene, 9-Tricosene, 1,19-Eicosadiene, 4-Undecene, 2-Dodecene, and Cyclododecane. These results agree with reports of some researchers (Abbas-Abadi et al., 2014; Kyong et al., 2003, 2002; Sarker and Rashid, 2013). However, they were a few variations in the primary hydrocarbon compounds from pyrolysis of LDPE, as reported by some other researchers (Sharuddin, 2016; Sarker and Rashid, 2013; Al-Salem, 2017; Mianda et al., 2019) Miandad et al., (2019) explained this difference. They report that plastic types, catalyst types and amount, temperature, and reaction time are essential determinants of the chemical composition of liquid oil from the pyrolysis of plastics.

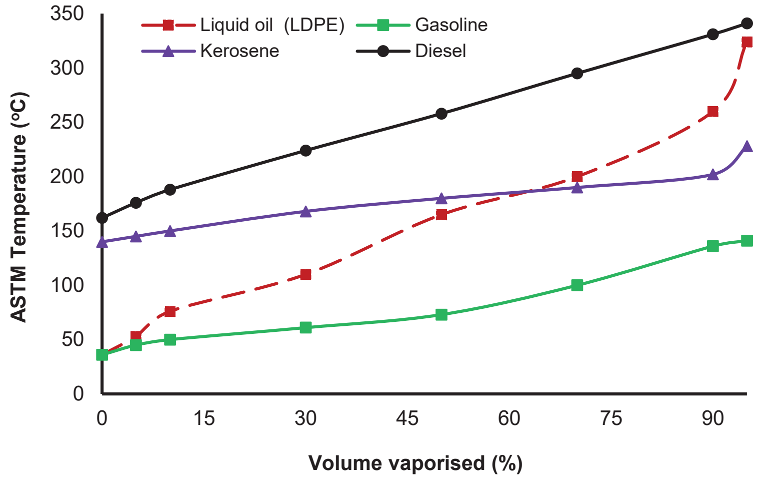

FIGURE 6: Variation of ASTM characteristics of pyrolysis liquids and standard fuels with volume vaporised.

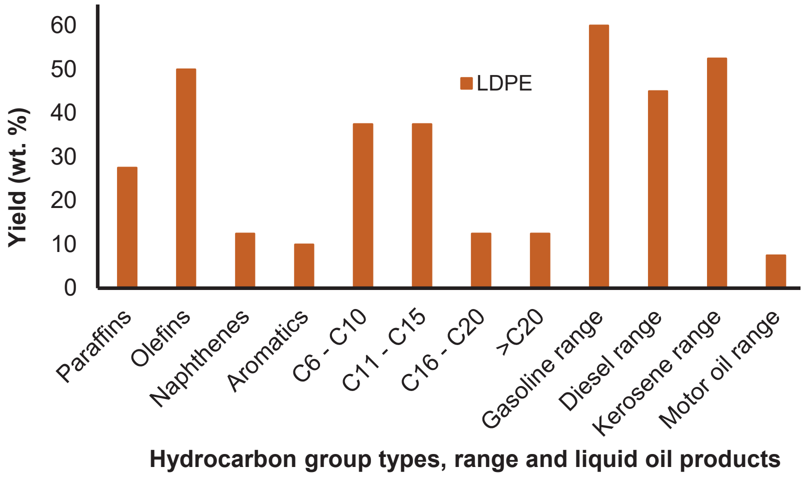

FIGURE 7: GC-MS analysis showing various effects of catalytic degradation on liquid oil from LDPE.

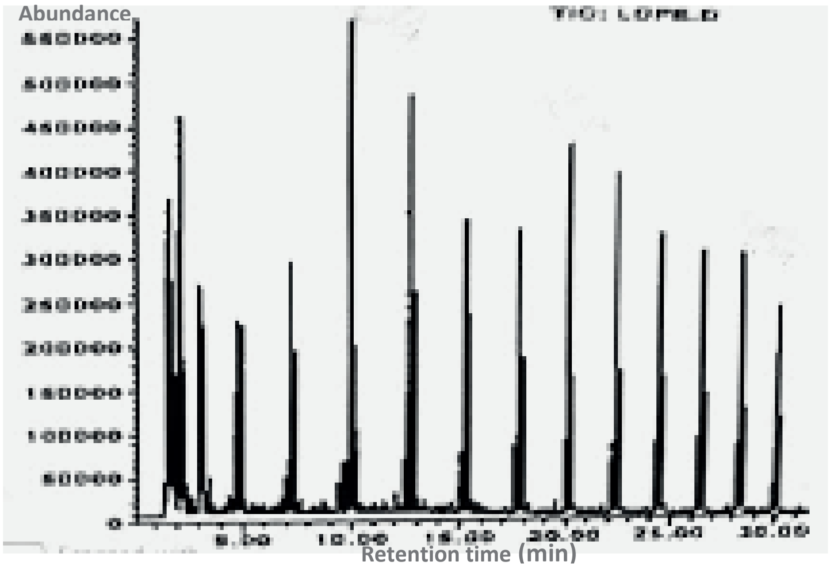

FIGURE 8: GC-MS of the pyrolysis oil from LDPE at 450°C and 0.2 catalyst to plastic ratio.

4. CONCLUSIONS

The following conclusions can be drawn:

1. The temperature variation and the application of spent FCC catalysts significantly impact LDPE degradation's products (liquid oil, gas, and char) yield.
2. The liquid oil yield from thermal degradation of LDPE was lower than that from catalytic degradation. However, the reverse is the case for gas and char yields.
3. The maximum temperature and catalysis to plastic ratio for optimizing LDPE were 500°C and 0.20, respectively.
4. The liquid oil, gas, and char produced at 500°C and catalyst to plastic ratio of 0.20 were 92.7 wt.%, 6.1 wt.%, and 1.2 wt.% respectively.
5. The fuel properties of the produced liquid oil from LDPE compare favorably with those of commercial fuels. The composition and quality of the produced liquid oil from LDPE were similar to gasoline and diesel.
6. The results obtained are gasoline, (60%), diesel, (45%), kerosene, (52.5%), and motor oil, (7.5%) for produced LDPE liquid oil.

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TABLE 4: GC-MS composition of produced oil from LDPE degradation.

S/N	Retention time (min)	Area (%)	Compound	Molecular Formula	Molecular weight
1	3.523	2.11	Toluene	C_7H_8	92.140
2	3.512	1.20	Bicyclo [4.2.0]octa-1,3,5-triene- methyl	C_8H_8	104.149
3	3.670	1.20	Benzene, 1,3-dimethyl-	C_8H_{10}	106.165
4	3.905	1.04	1-(2-propenyl) cyclopentene	C_8H_{12}	108.181
5	4.245	1.12	1,2-dimethyl-1-cyclohexene	C_8H_{14}	110.200
6	4.526	3.14	Indene	C_9H_8	116.160
7	5.435	1.32	Indane	C_9H_{10}	118.176
8	5.563	1.08	Benzene, 1-ethyl-4-methyl-	C_9H_{12}	120.192
9	5.927	1.20	Naphthalene	$C_{11}H_8$	126.171
10	5.956	1.03	4-Nonene	C_9H_{18}	126.239
11	6.051	2.85	2-Methylindene	$C_{10}H_{10}$	130.186
12	6.567	2.34	9-Methylbicyclo [3.3.1] nonane	$C_{10}H_{18}$	138.254
13	6.870	1.06	2-methylpropyl cyclohexene	$C_{10}H_{18}$	138.254
14	6.985	2.72	1-Decene	$C_{10}H_{20}$	140.266
15	7.020	1.43	Trans-3-Decene	$C_{10}H_{20}$	140.266
16	8.123	2.30	Naphthalene, 1-methyl	$C_{11}H_{10}$	142.197
17	8.742	1.26	4-Undecene	$C_{11}H_{22}$	154.292
18	9.347	2.12	Naphthalene, 1.6 dimethyl-	$C_{12}H_{12}$	156.224
19	10.231	1.45	1,6-Dimethyl decahydronaphthalene	$C_{12}H_{22}$	166.303
20	10.542	1.50	2-Dodecene	$C_{12}H_{24}$	168.319
21	10.748	1.10	Cyclododecane	$C_{12}H_{24}$	168.319
22	10.984	2.30	Naphthalene, 2-methyl	$C_{11}H_{10}$	142.197
23	11.307	3.34	1-Tridecene	$C_{13}H_{26}$	182.346
24	11.543	2.51	Tridecane	$C_{13}H_{28}$	184.360
25	11.674	1.57	Naphthalene, 2-methyl	$C_{11}H_{10}$	142.197
26	11.924	3.85	2-Tetradecene	$C_{14}H_{28}$	196.372
27	12.316	1.57	1-Pentadecene	$C_{15}H_{30}$	210.400
28	12.945	2.21	Cis-9-Tetradecadiene	$C_{14}H_{28}O$	212.352
28	13.130	3.52	Pentadecane	$C_{15}H_{32}$	212.410
30	14.643	1.89	3-Heptadecene	$C_{17}H_{34}$	238.452
31	17.401	1.63	1-Octadecene	$C_{18}H_{36}$	252.487
32	18.257	2.23	Octadecane	$C_{18}H_{38}$	254.494
33	18.722	2.87	1-Nonadecene	$C_{19}H_{38}$	266.512
34	22.435	2.16	1,19-Eicosadiene	$C_{20}H_{42}$	282.550
35	24.541	1.78	1-Docosene	$C_{22}H_{44}$	308.585
36	25.467	1.54	9-Tricosene	$C_{23}H_{46}$	322.621
37	26.038	2.46	Cyclotetracosane	$C_{24}H_{48}$	336.638
38	26.542	2.89	Tetracosane	$C_{24}H_{50}$	338.654
39	29.123	1.64	Nonacosane	$C_{29}H_{60}$	408.787
40	30.156	1.35	Cyclohexasiloxane, dodecamethyl-	$C_{12}H_{36}O_6S_{i6}$	444.924

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CISA

FABRICATION OF SAWDUST BRIQUETTES USING LOCAL BANANA PULP AS A BINDER

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ABSTRACT

Sawdust is generated in large quantities in sawmills in Manicaland province in Zimbabwe, posing an environmental threat if incinerated or left to accumulate. This presents an opportunity for valorising the waste into briquettes. To make the briquettes cost effective, a cheap and locally available binder is needed. Hence, the use of banana waste pulp (green banana, ripe banana, pseudo-stem pith and fruit-bunch-stem) as a sawdust briquettes binder was investigated in this study. The quality of fabricated briquettes was evaluated based on relative density, shatter index, ignition time, burning rate, and time taken to boil a constant amount of water. The briquettes showed improved qualities with the increase in binder ratios. The sawdust-binder ratio of 1:4 showed the best briquettes qualities. Factor rating method was done in order to obtain the best scored (scale 0-100) briquettes reflecting weightier factors (scale 0-1). The weightiest factors were time taken to boil water (0.25) and shatter index (0.25), with three other factors weighing 0.5. The best five binder formulations in their order were; banana-pseudo stem-pith (pith) and ripe banana (RB) in the ratio of 3:1 respectively (score weight of 68.75), fruit-bunch-stem, banana-pseudo-stem-pith (pith), green banana (GB) and ripe banana (RB) in the ratio of 2:2:1:1 respectively (score weight of 68.3), banana-pseudo-stem-pith (pith), ripe banana (RB) and green banana (GB) 2:1:1 respectively (score weight of 66.95), banana-pseudo-stem-pith, ripe banana and green banana in the ratios of 2,5:1,5:1 respectively (score weight of 66.05) and banana-pseudo-stem-pith and ripe banana in the ratios of 1:1 (score weight 65.55).

1. INTRODUCTION

The majority of biomass waste is incorrectly disposed of in Zimbabwe and other developing countries, creating an ecological concern in a variety of ways (Charis et al., 2019). Zimbabwe has a significant number of timber plantations. In 2009, plantation forests covered 89, 862 hectares, with about 90% of these plantations located in the Eastern Highlands (Manicaland province) owing to the high altitudes and high rainfalls in the area (Mtisi & Prowse, 2012) . Timber processing into various products generates a lot of waste products, such as tree cut offs, sawdust, and tree bark, and this waste has become prominent due to the high demand for timber and its derived products (Owoyemi et al., 2016). A small percentage is being used in boilers to produce steam used for drying timber mainly (Manyuchi et al., 2018) whilst the rest, especially sawdust, is largely unutilized. Throughout the year, heaps of wood waste accumulate across the whole timber value chain. Although local people and timber processors use some of this waste for fuel, they have been unable to keep up the accumulation of sawdust, reported to be well over 70, 000 tonnes per annum (tpa) (Mtisi & Prowse, 2012). Some of the sawdust is incinerated, while some burns spontaneously as a result of hotspots within the heaps, endangering the environment and its various ecosystems (Charis et al., 2019). The local city council has actually restricted the disposal of sawdust into its dumpsites due to such fire hazards (Manyuchi et al., 2018). There is also a host of other environmental impacts including organic wood leachate and marring of the aesthetic appeal of Manicaland (Arimoro et al., 2006). Similarly, health and environmental issues arise from banana wastes as they frequently harbour disease vectors such as flies and mosquitos (Akala M. B., 2021). Perhaps, the threat to health from such undermanaged wastes is what justifies the call to condescend from the 3R (Reduce, Reuse,

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Recycle) to the 3S (Sanitisation, Subsistence economy and Sustainable landfilling) concepts (Lavagnolo and Grossule, 2018). In this case, there would be an urgent need to address sanitization aspects if the capacity to engage in the 3R is still low, as is characteristic of low income nations. The aim of this project however, was to begin exploring the potential to achieve a circular economy involving locally available sawmill and banana waste, using an affordable recycling method. This also contributes to the achievement of a 'zero waste' society.

Timber waste from sawmills can be valorised using different methods like gasification, pyrolysis, direct combustion, liquefaction, biochemical and densification (Jingura et al., 2013). Whereas the other routes are mostly industrial at medium to high scales of productivity to make economic sense, densification appeals for a wider market from domestic, small scale and commercial/industrial scales. The densified sawdust would then be replacing the traditional use of wood or coal as fuel with the advantage that it is renewable. Indeed, biomass (especially waste) is a more sustainable source of energy than fossil fuels, which are not. The low bulk density (typically 150 to 200 kg/m^3 for woody resources such as wood chips and sawdust), relatively high moisture content and low energy density, of biomass are some of the significant drawbacks on its use as an energy feedstock (Rajaseenivasan et al., 2016). The low bulk density and dusty characteristics of the biomass also cause problems in transportation, handling and storage (Husan et al., 2002) .The application of biomass briquetting i.e. densifying the loose biomass is an effective way to increase the bulk and energy density of the loose chips or sawdust for easier use domestically and industrially (Ogwu et al., 2014). Such briquettes can even be used to fuel power plants for electricity generation. It was on these premises that three potential co-generation projects in Nyanga, Chimanimani and Charter sawmills were proposed by the Department of Energy to produce 3.5 MW, 3 MW and 10 MW of energy respectively from briquettes made from sawdust wastes at those sites (Jingura et al., 2013). These excluded additional wastes from scattered bush mills, Chinese sawmills and dysfunctional sawmill sites. Mechanical compaction of sawdust into briquettes is a costly venture hence the reason why companies prefer to simply dispose of the waste (Nyemba et al., 2018). There is therefore a need for affordable, economic and sustainable ways of making briquettes that are acceptable and meet the needs of both industry and the community (Jingura et al., 2013).

The use of a very cheap, locally available binders for briquettes binding has been found as a good way of reducing costs of making briquettes. A good briquette binder must be economically accessible, strong, environmentally pollution-free and produce strong bond. Therefore, finding a suitable binder in briquetting is an important step (Abdulmalik et al., 2020). Poor quality briquettes may crack and crumble back to the original components when stored, processed or handled (Idah & Mopah, 2013). A suitable binder which addresses these problems is imperative. Furthermore, the density, durability and combustion characteristics of fuel briquettes are influenced by the binder. Traditionally cement, clay, animal dung, bitumen, wood/coal tar, gum Arabic have previously been utilised as binders for making briquettes (Abdulmalik et al., 2020). However, economic viability of these materials is questionable for briquetting due to competition for their usage in other sectors. Moreover, combustibility and calorific values of briquettes based on these binders are low (Abdulmalik et al., 2020). These inorganic binders have good compaction ratio, hydrophobic nature and compressive strength than organic binders but inorganic binders decrease the net heating value of briquettes and increase the burn out temperature and ash content (Manyuchi & Mbohwa, 2018). Several studies have cited organic binders such as starch, protein, fibre, fat/oil, lignosulfonate, modified cellulose, rapeseed flour, coffee meal, orange peels, bananas waste, bark gums, molasses sugar, starch, sulphite liquor, cassava wastewater, microalgae and okra stem gum, pine cones and lignin powder as possible briquettes binders (Abdulmalik et al., 2020; Rajaseenivasan et al., 2016). To that effect some studies have been conducted where several binders have been tested in the production of briquettes from different biomass; rice husk/cassava peel gel, rice husk/banana peel, maize cob/cassava peel gel, maize cob/banana peel, groundnut shell/cassava peel gel, groundnut shell/banana peel, sugarcane bagasse/cassava peel gel, sugarcane bagasse/banana peel with encouraging bulk densities calorific values and other properties. Nevertheless, limited research is available on the use of banana waste (green bananas, ripe bananas, pseudo-stem pith and fruit-bunch-stem) as binders in the fabrication of pine sawdust-based briquettes. Banana waste briquette binders are encouraging since many parts of the banana are rich in starch and vast quantities of these materials are always discarded. Thus, there is ample scope for study on the use this cheap and locally available binder judging by the ever-increasing concerns on the economics of briquettes fabrication and marketing.

This study considered the potential of banana waste as a briquette binder. Bananas (Musaceae) in Zimbabwe are grown in Honde Valley, Burma Valley, Birchnough Bridge, Chimanimani and other areas with a tropical environment that has plenty of humidity, good drainage and fertile soil. In 2017, the Honde Valley in eastern Zimbabwe alone produced over 27 000 tons of bananas. Every month, farmers generate over 1000 kg of bananas (Lacey, 2018). Banana fruit- bunch- stem, pseudo-stem and some green bananas are discarded as banana waste which is thrown away as residue after banana harvesting hence this waste can be collected and put into good use. This study therefore investigated the integration of two wastes into compact wood briquettes, paying special attention to the binder formulation, composition and sawdust to binder ratio.

2. MATERIALS AND METHODS

2.1 Experimental design

A laboratory scale experimental design method was employed for production of briquettes. Sawdust and banana waste binder were prepared and mixed according to the chosen ratios. The resulting briquettes were sun dried and tested for quality. The variables chosen were binder composition and binder ratio. The response variables measured were relative density, shattering test, boiling water test, burning rate test and ignition time test. The binder ratios and the sawdust- binder ratios were as shown in Table 1.

A factor rating method was then done in order to obtain the best briquettes with high quality parameters.

2.2 Briquettes preparation equipment

A digital analytical balance (model KERN 19037) was used for weighing. A blender (model EM BL-1075) was used to make banana waste pulp. A stove (DSP model 4046) was used as a source of heat for boiling the binder solution.

2.3 Material preparation

2.3.1 Feedstock

Pine sawdust was chosen as the feedstock material for this study. Sawdust was solar dried for 3 weeks to achieve a moisture content ~6.50% as achieved previously by Charis et al (2020). The final moisture content of each sample was determined by determining the difference between the original and final sample mass after oven drying for 4 hours, according to the ASTM D4442 method.

2.3.2 Binder

The natural binder used was banana pulp. Banana waste which comprised of green bananas (GB), ripe bananas (RP), pseudo-stem pith (pith) and fruit-bunch-stem (stem) was collected from Burma Valley. These were cleaned to be devoid of foreign matters. 200 g of the waste was cut into 2-3 cm pieces and put in a blender and 100 ml of water was added to the pieces and the blender was set at a speed of 2. The contents where blended to puree form after which 600 ml of water was added to 300 g of the binder. The mixture was then boiled for 1 h under continuous stirring to refine the pulp, which contained sufficient starch for briquettes binding. The procedure was done for banana waste mixtures which were combined using the ratios shown in Table 1, with fruit-bunch-stem and pseudo-stem pith being higher in the ratio since these are found in large quantities as residue compared to discarded bananas.

TABLE 1: Binder and sawdust to binder ratios.

			Sawdust : Binder ratios			
			1:2	**1:3**	**1:4**	**1:5**
Binder ratios	GB: Stem	1:1				
		1:3				
	RB : Stem	1:1				
		1:3				
	GB : RB : Stem	1:1.5:2.5				
		1:1:2				
	GB : Pith	1:1				
		1:3				
	RB : Pith	1:1				
		1:3				
	GB : RB : Pith	1:1.5:2.5				
		1:1:2				
	GB : RB : Stem : Pith	1:1:2:2				

2.3.3 Mixing sawdust with binder

The sawdust-binder ratios used were 1:2, 1:3, 1:4 and 1:5. The mixing of sawdust and binder prior to compaction was done manually. In a 5L bucket using a wooden rod.

2.3.4 Densification process

The sawdust-binder mixture of 50 g mass was put into the cylindrical die of 2.3 cm diameter and 5 cm height. A cloth was used to cover the other end and allow water to pass through. Pressure was applied manually using a wooden rod. The assumption made was that every manual compression stroke was delivered with the same strength. The briquetting process is illustrated in Figure 1.

2.3.5 Briquettes drying

The as-fabricated briquettes were sun dried for 1 week in sunny weather conditions.

2.4 Briquettes quality parameters

2.4.1 Relative density

The relative density of samples was calculated by measuring the volume and weight. It was calculated in accordance to the equation (1) (Manyuchi et al., 2018):

$$Relative\ density\ (kg/m^3) = \frac{mass\ of\ dried\ briquette\ (kg)}{volume\ of\ briquette\ (m^3)} \quad (1)$$

2.4.2 Shatter index

Shatter index is used to assess how hard the briquettes are. The shatter index of the briquettes was determined by dropping 50 g of briquettes put in a plastic bag for five times from a height of 2 metres onto a concrete floor (Manyuchi et al., 2018). The weight of the fragmented briquette was recorded. The shattering index was determined according to ASTM D440-86 of drop shatter developed for coal using the equation (2) (Davies & Davies, 2013):

$$Shattering\ index = \frac{weight\ of\ briquettes\ after\ dropping}{weight\ of\ briquettes\ before\ dropping} \quad (2)$$

2.4.3 Water boiling test

A 50 g mass of each briquette sample was placed in a brazier made from a fabricated can and used as feel to boil 50 ml of water. A stopwatch was used to record the time it took to boil the water in minutes. The mercury dry bulb thermometer was used to pin point the exact temperature at which the water boiled (Thulu et al., 2016).

2.4.4 Ignition time

Each briquette sample was ignited at the base in a drought free corner using matches and the time required for the flame to ignite the briquette was recorded in seconds using a stop watch (Thulu et al., 2016).

2.4.5 Burning rate

A 50 g briquette sample from each blend was burnt and the initial time (ignition time) to the final time of burning (flame extermination) was recorded using a stopwatch. The burning rate was calculated using equation (3) (Hassan et al., 2017).

$$Burning\ rate = \frac{mass\ of\ fuel\ consumed\ (g)}{total\ time\ taken\ (min)} \quad (3)$$

2.5 Factor rating method

Weight was assigned to briquettes quality parameters according to importance as obtained from literature. A scale of 0-1 was used to weigh each factor. The most important factors were the time taken to boil water (weight 0.25), which reflects on the heating value of the fuel; and the shatter index (weight 0.25), which reflects on the impact resistance and strength of briquette (Ige et al., 2020). Each parameter was scored out of a 100, using the factor scale. Scores were multiplied by weights for each factor and these were then summed.

3. RESULTS AND DISCUSSION

3.1 Physical properties of pine sawdust

The raw pine sawdust had been previously tested in work reported by Charis et al. (2020). The results of the proximate and calorific value tests are given in Table 1 (Charis et al., 2020). The ash content of pine sawdust obtained from the study is greater than the ash content value of 0.3-0.4% reported in literature (Garcia et al., 2021; Tillman et al., 2012; Chaula et al., 2014). The fixed carbon value obtained is closer to the value of 18.60% reported by other researchers (Chaula et al., 2014) whilst the value of the volatile matter obtained is inconsistent with the value of 81.03% from literature (Chaula et al., 2014). The moisture content was way above the moisture content value reported by other researchers (Garcia et al., 2021; Tillman et al., 2012; Chaula et al., 2014) and the heating value obtained was slightly lower than the value of 19.98 MJ/kg from literature (Garcia et al., 2021) . The inconsistency of the results with those reported in literature may be attributed to factors like climate conditions, growth conditions and sawdust handling.

3.2 Relative density

3.2.1 Effect of binder ratio on relative density

Figure 2 shows the relationship between binder ratio and relative density. The binder formulations as a ratio to sawdust were averaged and so were the densities of briquettes formed using those sawdust: binder rations. The aim was to see the general trend as the amount of binder increased in the mix.

The lowest relative density was recorded at the sawdust binder levels of 1:2. The density of the briquettes increased as the binder ratio increased up to the sawdust-binder ratio of 1:4. This suggests that briquettes produced with high concentration of binder are denser and have a longer burning time when used for heating while those produced with low concentration of binder are less dense and therefore are likely to ignite much easier when compared with high concentration of binder (Ige et al., 2020). The relative density of the briquette however, decreased from the sawdust-binder ratio of 1:4 to 1:5. The density increase with increase in sawdust-binder ratio could be due to the filling up of air spaces between sawdust particles by the binder (Musa, 2007).

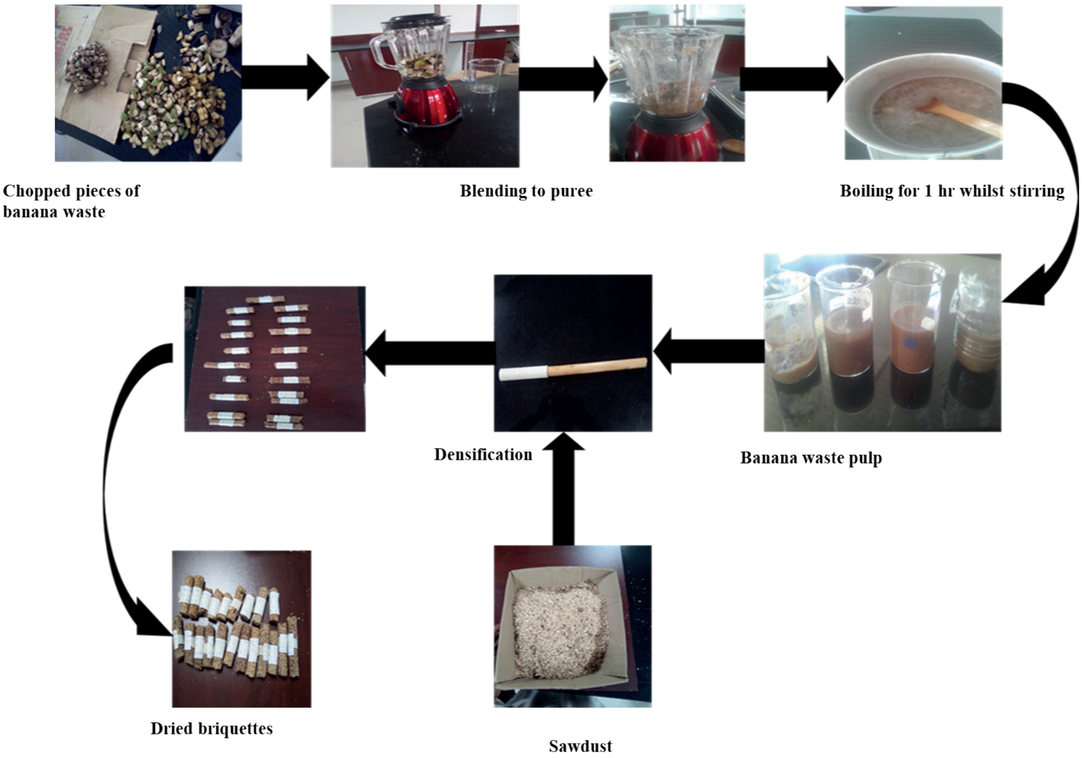

FIGURE 1: Briquetting process.

TABLE 2: Sawdust properties (Charis et al., 2020).

Physical property	Value (% dry basis)
Ash	0.83
Fixed carbon	20.00
Volatile matter	79.16
Moisture content	65.41
HHV (MJ/kg)	17.568

3.2.2 Relative densities of different binder mixtures

Figure 3 shows the relative densities of different binder mixtures of banana waste binders. The highest relative density recorded for a binder mixture with stem was 0.238 g/cm^3 which was a mixture of stem and ripe banana in the ratio of 1:1. The lowest recorded for a stem mixture was 0.18 g/cm^3 for a ratio of 2.5:1.5:1 of stem, green banana and ripe banana, respectively. Banana pith mixture with green banana and ripe banana in the ratio of 2:1:1 exhibited the highest relative density of 0,238 g/cm^3. The results showed that the mixtures of banana pith with green banana in the ratio of 1:1 exhibited the lowest relative density (0.187 g/cm^3) for a binder mixture which included banana-pseudo-stem-pith.

3.3 Shatter index test

3.3.1 Effect of binder ratio on shatter index

Figure 4 shows the relationship between binder ratio and shatter index. The same concept of averaging mentioned in section 3.2.1 was applied to the both the dependent and independent variables. The highest shatter index was at the sawdust-binder ratio of 1:4 (1) and the lowest was recorded at the ratio of 1:2 (0.986). There was an increase in shatter index with an increase in sawdust-binder ratio. The binding agent helps in fusing the biomass particle and thus enhances the durability of the resulting briquette (Saeed et al., 2021). The shatter index is above 95% which falls within the standard pellet quality durability range of 95-100%. Lower durability hinders operation and transportation of briquettes (Saeed et al., 2021).

3.3.2 Shatter index tests for different binder mixtures

Figure 5 shows the shatter index of different binder mixtures. The best shatter index was recorded for the mixtures of banana stem/pith with green banana at the ratio of banana pith/stem to green banana of 1:1. Generally the shattering index for mixtures with banana pith were higher than those with banana stem.

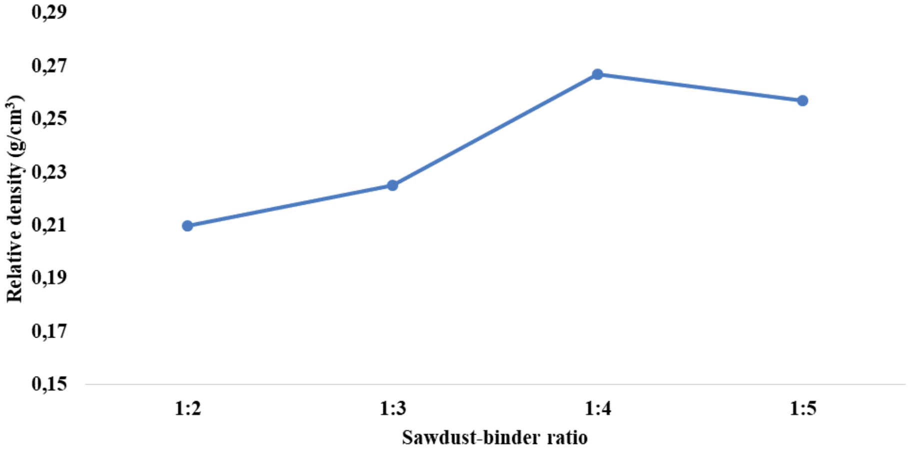

FIGURE 2: Effect of binder ratio on relative density.

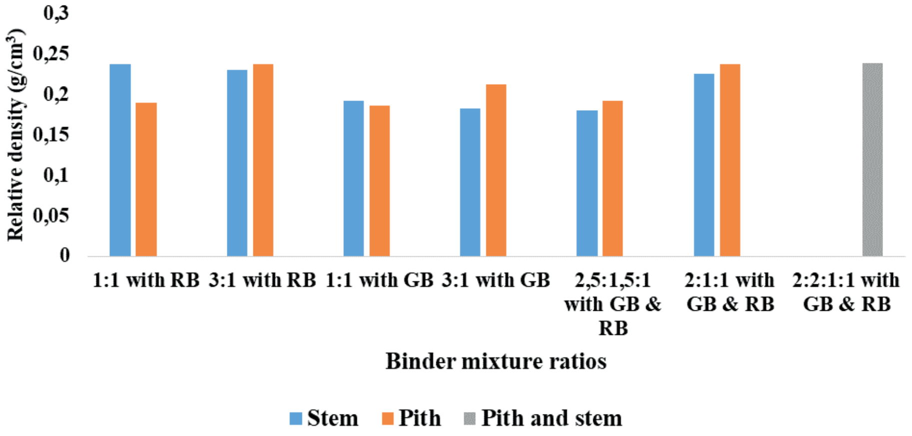

FIGURE 3: Comparisons of relative densities of different binder mixture ratios.

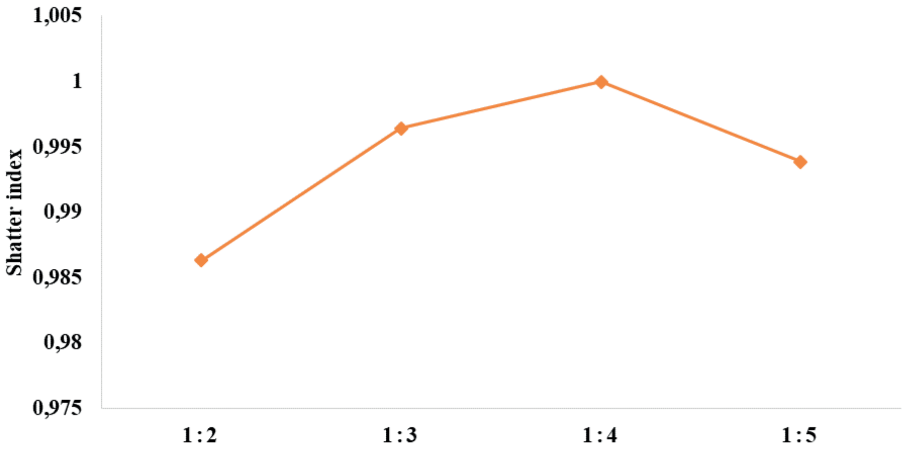

FIGURE 4: Effect of binder ratio on shatter index.

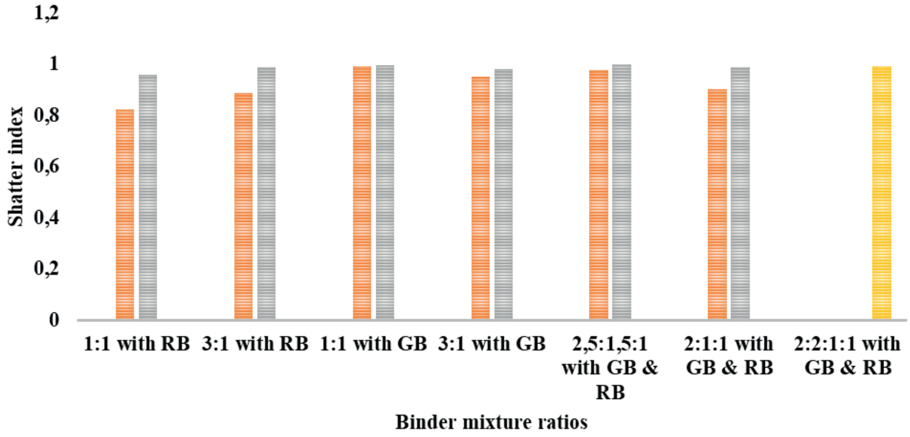

FIGURE 5: Shatter index tests for different binder ratios.

3.4 Ignition time and Water test

3.4.1 Effect of binder ratio on ignition time and time taken to boil water

The relationship between binder ratio and ignition time and time taken to boil water is shown in Figure 6. The same concept of averaging mentioned in section 3.2.1 was applied to the both the dependent and independent variables. The obtained trend indicated that ignition time and time taken for water to boil increased with decrease in binder proportion. This may be due to increase in density. This also indicates that the little the amount of volatile matter the longer the ignition time which is in agreement with the results obtained from studies done by Kimutai and Kimutai (2019) and Ige et al (2020).The briquettes with lower binder ratio burn slowly as a result, lots of the heat released was lost before the water boils.

3.4.2 Ignition time and time taken to boil water for different binder mixtures

Figure 7 and 8 show the ignition time and the time taken to boil water by different binder mixtures. Mixtures containing banana stem had lesser ignition time compared to those with banana pith. Banana stem/pith mixed with green banana in the ratio of 1:1 was the fastest to ignite whereas the mixtures of banana stem/pith with green banana in the ratios of 3:1 respectively were slowest to ignite. Banana stem mixtures took longer to boil water than banana pith mixtures. The longest time taken to boil water was 68 min by green banana with banana stem in the ratio of 1:1. The mixture of banana pith with green banana in the ratio of 3:1 respectively, took the shortest time to boil water.

3.5 Burning rate

3.5.1 Effect of binder ratio on burning rate

Figure 9 shows the relationship between binder ratio and burning rate. The same concept of averaging mentioned in section 3.2.1 was applied to the both the dependent and independent variables. The briquettes burnt fast with increasing binder proportion hence the highest binder ratio burnt for shorter period compared to the lowest. Low volatile matter makes the briquettes to burn slowly and the opposite is true (Ige et al., 2020). This is similar to the findings by Hassan et al (2017), the improvement of fuel ignition time and the burning rate with the increase in binder ratio is due to a higher volatile content (Hassan et al., 2017).

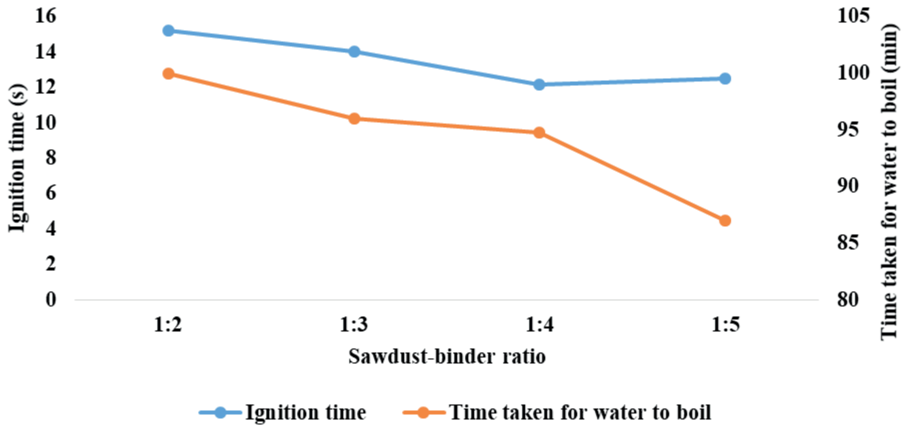

FIGURE 6: Effect of binder ratio on ignition time and time taken to boil water.

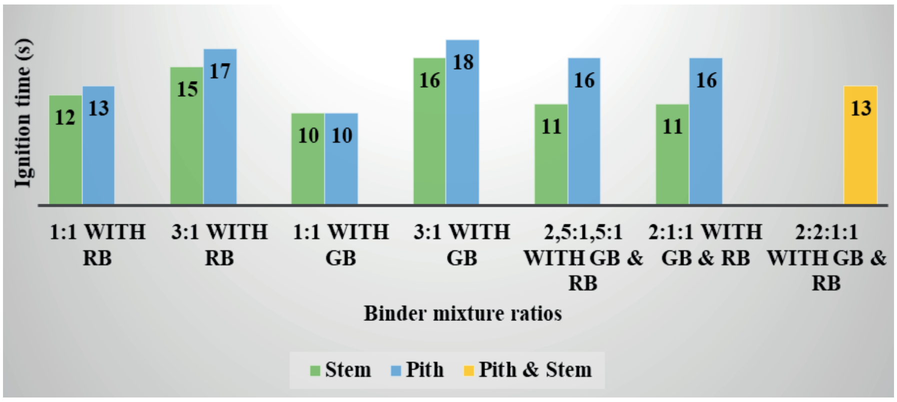

FIGURE 7: Ignition time for different binder mixtures.

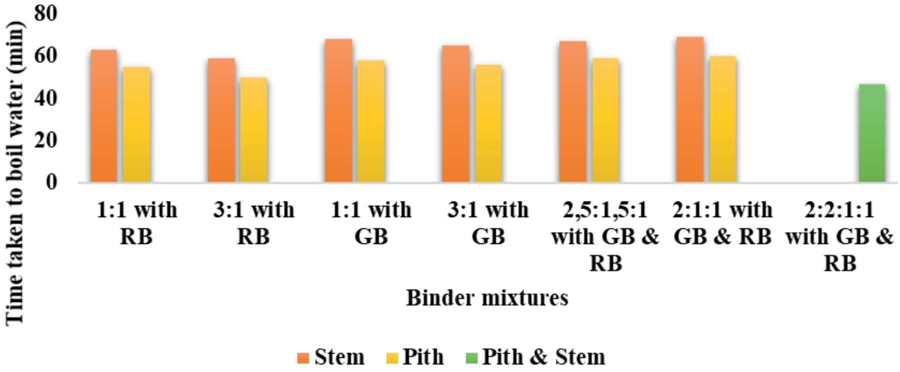

FIGURE 8: Time taken to boil water by different binder mixtures.

3.5.2 Burning rate for different binder mixtures

Figure 10 shows the burning rates for different binder mixtures. The mixture of green banana and ripe banana with pith in the ratios of 1:1:2 respectively, had the highest burning rate of 0.95 g/min whilst the lowest burning rate obtained was from the mixture of green banana with stem in the ratio of 1:1. The mixtures containing stem had the least burning rate compared to those containing banana pith.

This study examined the utilisation of banana waste pulp as a binder for sawdust briquettes. The effects of binder mixing ratio, sawdust moisture content and different binder mixtures on relative density, shattering index, ignition time, burning rate and time taken to boil water were examined. A factor rating method was then used in order to determine which binder mixtures were used to produce high quality briquettes. Tables 3 and 4 show the factor rating method. The factors were given weights considering their relative importance according to literature, with core factors around the physical integrity and thermal value of the briquettes scoring higher.

By using the factor rating method, the best five binder formulations (combinations and ratios) in their order

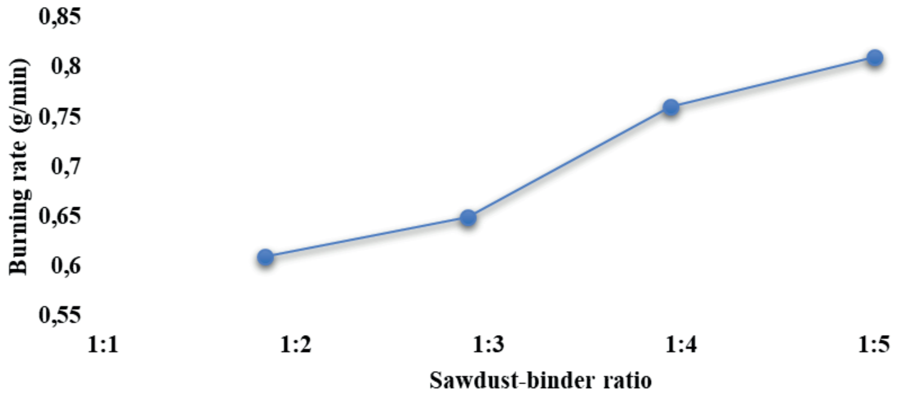

FIGURE 9: Effect of binder ratio on burning rate.

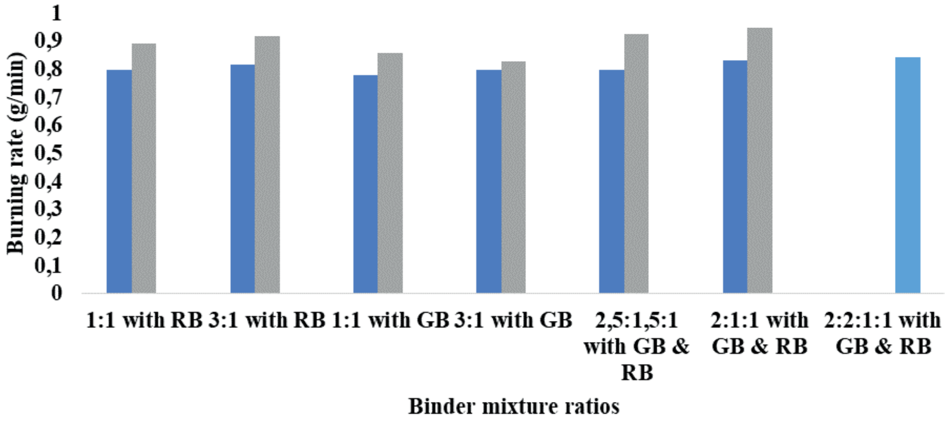

FIGURE 10: Burning rates for different binder mixtures.

were; banana-pseudo stem-pith (pith) and ripe banana (RB) in the ratio of 3:1 respectively (score weight of 68.75), fruit-bunch-stem, banana-pseudo-stem-pith (pith), green banana (GB) and ripe banana (RB) in the ratio of 2:2:1:1 respectively (score weight of 68.3), banana-pseudo-stem-pith (pith), ripe banana (RB) and green banana (GB) 2:1:1 respectively (score weight of 66.95), banana-pseudo-stem-pith, ripe banana and green banana in the ratios of 2,15:1,5:1 respectively (score weight of 66.05) and banana-pseudo-stem-pith and ripe banana in the ratios of 1:1 (score weight 65.55).

4. CONCLUSIONS

This study focused on the fabrication of sawdust briquettes using local banana pulp as a binder. The physical properties of the sawdust briquettes were found to be significantly affected by the binder ratio. The briquette properties improved with an increase in binder ratio and for the optimum sawdust briquette quality on the basis of relaxed density, shatter index and burning properties, a sawdust-binder ratio of 1:4 was obtained. Such cheaply made briquettes have the potential to provide sustainable fuel

TABLE 3: Factor rating for different binder mixture ratios containing stem.

Factor	Weight	1:1 Stem & RB		3:1 Stem& RB		1:1Stem& GB		3:1 Stem & GB		2,15:1,5:1 Stem & RB &GB		2:1:1 Stem & RB& GB		2:2:1:1 Stem & Pith& GB & RB	
		Scores	Weighted scores	Scores	Weighted scores	Scores	Weighted scores	Scores	Weighted scores	Scores	Weighted scores	Scores	Weighted scores	Scores	Weighted scores
Relative density	0.20	24.0	4.8	23	4.6	19	3.8	18	3.6	18	3.6	23	4.6	24	4.8
Shatter index	0.25	82	20.5	89	22.25	99	24.75	95	23.75	98	24.5	90	22.5	99	24.75
Ignition time	0.1	88	8.8	85	8.5	90	9	84	8.4	89	8,.9	89	8.9	87	8.7
Time taken to boil water	0.25	37	9.25	41	10.25	32	8	35	8.75	33	8.25	31	7.75	53	13.25
Burning rate	0.20	80	16	82	16.4	78	15.6	80	16	80	16	83	16.6	84	16.8
Total	1		59.35		62		61.15		60.5		61.25		60.35		68.3

TABLE 4: Factor rating method for different binder mixtures containing pith.

Factor	Weight	1:1 Pith & RB		3:1 Pith & RB		1:1 Pith & GB		3:1 Pith & GB		2,15:1,5:1 Pith & RB &GB		2:1:1 Pith & RB& GB	
		Scores	Weighted scores	Scores	Weighted scores	Scores	Weighted scores	Scores	Weighted scores	Scores	Weighted scores	Scores	Weighted scores
Relative density	0.2	19	3.8	24	4.8	19	3.8	21	4.2	19	3.8	24	4.8
Shatter index	0.25	96	24	99	24.75	100	25	98	24.5	100	25	99	24.75
Ignition time	0.1	87	8.7	83	8.3	90	9	82	8.2	84	8.4	84	8.4
Time taken to boil water	0.25	45	11.25	50	12.5	42	10.5	44	11	41	10.25	40	10
Burning rate	0.2	89	17.8	92	18.4	86	17.2	83	16.6	93	18.6	95	19
Total	1		65.55		68.75		65.5		64.5		66.05		66.95

and power needs for local consumption (Mtisi & Prowse, 2012. Utilisation of sawdust to generate energy could offset the lumber industry's power needs while also potentially injecting a surplus into the national grid. Furthermore, the use of briquettes from waste can curb deforestation for fuelwood. However, a stronger value proposition in this regard could be to make charcoal briquettes which could be more attractive because of their cleaner burning, encouraging users to prefer these to traditional fuelwood. This demonstrates that valorisation of fruit and timber wastes sustainably has great potential in producing convenient energy or fuel sources. As the economy expands and the population increases, Zimbabwe’s energy demands are set to grow (Mtisi & Prowse, 2012). In order to fulfil the fast-expanding energy demand, renewable energy sources must be appropriately utilized. However, sustainable harvest and uses of such waste has to be observed, especially for banana wastes. The waste harvested should leave enough ground cover so that there is continuity of other ecosystems that depend on such waste, including breakdown into humus as a good agricultural process (Charis et al., 2022). At the same time, there should be investigations into alternative socio-economic uses of such waste, so that there is no unhealthy competition in the uptake of both wastes. Charis et al., 2019 already showed that timber sawdusts have minimal uses, however for banana waste, it depends with the part of banana. Although single bananas that fall-out from the bunch are usually counted as waste, there could be arguments stemming from their potential uptake as human or animal feed.

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SOLID MEDICAL HOSPITAL WASTE IN TIMES OF CORONA: INCREASED VOLUME BUT NO INCREASED BIOHAZARD RISK

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ABSTRACT

The effect of the COVID-19 pandemic on medical waste EWC/EURAL code 180103* (infectious medical waste) and 180104 (non-infectious medical waste) was investigated in 6 university hospitals and 6 general hospitals. Data on the number of in-hospital patients and on quantity and volume of waste were obtained during 2019 (control period) and in 2020 up to March 2021 (COVID-19 period) for the hospitals, from the waste managing company, and from the regional destruction facility. The presence of SARS-CoV-2 on the surface of waste recipients was analyzed using RT-PCR. We found that the effect of the pandemic on the total weight of waste is limited during the first wave (March 2020), while during the second wave, the quantity of waste type 180103* increased. The main effect is a nearly doubling of the volume of waste during both waves caused by the use of cardboard hospital boxes with a yellow inner plastic bag. We demonstrated that the average weight of these cardboard boxes generated for the treatment of COVID-19 patients is significantly lower compared to the weight of the waste from non-COVID-19 patients. COVID-19-related health care activities caused a weight increase of the 180103* waste from historical data (0.2-1.4 kg/day/bed) up to 5-8 kg/day/bed. RT-PCR analysis of swabs demonstrated the absence of viral RNA on personal protection materials and on the surface of recipients containing the waste. We conclude that COVID-19-related hospital waste is predominantly of the EWC 180104 type.

1. INTRODUCTION

It is beyond saying that the pandemic caused by severe acute respiratory syndrome coronavirus type 2 (SARS-CoV-2), the virus that leads to coronavirus disease 2019 (COVID-19), has startled society in all aspects since January 2020. Drastic measures such as social isolation, restriction on travel, and economic activity, up to the complete lockdown of cities and countries, were installed and legally secured. The WHO declared the SARS-CoV-2 epidemic officially a pandemic on March 11, 2020 (Cucinotta and Vanelli, 2020). At the time of writing this manuscript in January 2022, nearly 300 million infections are ascertained with nearly 5.5 million deaths throughout the world (Huraimel et al., 2020, WHO, 2021, John Hopkins University, 2022). As a consequence of the lockdown and adjacent measures, other aspects of societal life gained interest. Reports on the effect of climate change on COVID-19-incidence, a significant reduction in air pollution, both in particulate matter and nitrogen oxide, and a reduction of water pollution to name a few, have been published (Sarkodie and Owusu, 2020, Saadat et al., 2020, Hens and Fraeyman, 2021).

Within the health care sector, the focus was evidently on patient care and on the research on antiviral medication and vaccines, the latter being produced at an unprecedented speed (Hebbani et al., 2022, Dubey et al., 2022). Gradually, the management of solid medical waste gained attention because it was suggested that this waste might be a source of viral transmission (Mol & Caldas, 2020). The finding that the virus has extended viability on many inert surfaces indeed warranted a precautionary approach (Wiktorczyk et al., 2021; Van Doremalen et al., 2020). Hence all material used for the treatment or diagnosis of COVID-19 patients was considered dangerous, and the waste was attributed to the EWC/EURAL code 180103*, which is waste with a risk for infection, requiring specific UN3291 certified recipients for collection and transport (Hoseindzadeh et al., 2020; WHO 2020). As a consequence, a massive in-

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crease in waste volume and a shortage in recipients were seen (Lavagnolo, 2020). In Wuhan, China, the volume of waste considered high-risk was in March 2020 about 5 times higher compared to the pre-COVID-19 period, while the waste density (kg/m³) decreased from 120 to 67-85 because of the use of the light-weight single-use personal protection equipment such as aprons, gloves, and masks (Wei, 2021). During that period, rather confusing and conflicting messages were distributed by, among others, the CDC (US Centers for Disease Control and Prevention) and the ECDC (European Centre for Disease Control), who stated that the waste from COVID-19 patients was not different from other patients (CDC 2021, ECDC, 2020), while WHO advised people handling health care waste from COVID-19 patients to wear boots, aprons, long-sleeved gowns, thick gloves, masks and face shields (WHO, 2020). From the point of view of transport legislation (ADR), COVID-19 waste was not taken within list A but considered in list B requiring UN 3291 certified recipients for transport. Needless to say that this was a disturbing and troublesome period for the whole health care sector.

Now, more than 2 years after the start of the pandemic, we want to describe the evolution of the waste management during the first and second wave of infections, based on formally registered data from 6 university hospitals (Vienna, Jena, and 4 university hospitals in Flanders and Brussels in Belgium) and 6 general hospitals in Flanders. First, we addressed a number of questions about the weight and volume and the financial consequences of the COVID-19 waste compared to waste from non-COVID-19 patients. Secondly, we investigated whether the allocation of the waste from COVID-19 patients to the EWC/EURAL 180103* code was justified. We used data from waves 1 and 2 in the year 2020 up to March 2021 and we used similar data from 2019 as an internal control.

2. METHODS

The name and the capacity of the 12 participating hospitals are summarized in Table 1. The number of beds refers to the situation in 2019.

The following data related to bed occupation by in-patients were obtained from the hospital administrations: (1) total number of non-COVID-19 hospitalized patients occupying a bed for at least one day, and (2) number and bed occupancy of hospitalized COVID-19 patients. Data were available on a daily basis, both for 2019 which is considered the control year, and for 2020 up to March 31st, 2021 (approximately the end of the second wave) which is considered the COVID-19 period.

As far as solid medical waste is concerned, we distinguish on the one side, the medical waste that might cause diseases to waste handlers and identified within Europe as waste with code EWC/EURAL 180103* and within the US as regulated medical waste, and waste generated from health care activities but non-infective and comparable to household waste, identified in Europe as EWC/EURAL 180104. The acronyms EWC and EURAL, the latter being frequently used in Belgium, are used interchangeably.

The following data were obtained: (1) quantity in kilograms of waste according to the EWC/EURAL code 180103* and 180104, and (2) the type of recipient the waste was collected in. In this respect, the legal restraints were respected: the risk waste (EWC 180103*) was collected in plastic material bins of 30, 50, or 60 liters or in cardboard containers with a plastic inner bag all according to the UN code 3921. The records of the weight during 2019 and 2020 were as detailed as possible and in most cases, this included the record of the weight of every transport of waste from the hospital to the destruction facility: for EWC/EURAL 180104 on a daily basis; for EWC/EURAL 180103* every 2-3 days.

In one of the university hospitals, the weight of each waste recipient coming from a COVID-19 ward was recorded on a daily basis from December 1st, 2020, until January 31st, 2021, covering a large part of the second wave. As a control, a similar number of recipients from conventional hospital wards were weighted on the same day.

Two additional commercial organizations were contacted and delivered information: the SUEZ group for recovery and waste management and the INDAVER facility in

TABLE 1: Identification of the participating hospitals.

Name and type of hospital	Number of beds
VAMED-KMB University Hospital, Vienna, Austria	1728
University Hospital Jena, Germany	1369
Ghent University Hospital, Ghent, Belgium	1049
University Hospital Saint Luc, Brussels, Belgium	945
Antwerp University Hospital, Antwerp, Belgium	573
University Hospital Brussels, Brussels, Belgium	721
General hospital Sint-Lucas, Bruges, Belgium	422
General city hospital, Aalst, Belgium	344
General hospital Maria Middelares, Ghent, Belgium	542
General hospital Virga Jesse, Hasselt, Belgium	769
General hospital GZA, Antwerp, Belgium	346
General city hospital St Elisabeth, Herentals, Belgium	243

Antwerp for incineration of EWC 180103* type of medical waste in Flanders - Belgium.

Calculations include the comparison between data from COVID-19 and non-COVID-19 patients during the whole study period: (1) estimation of the evolution of the number of patients on a daily basis, (2) estimation of the total waste weight (kg), of the volume and of the weight per day per bed during the control period and during the COVID-19 wave, (3) estimation of the average weight of the recipients.

The presence of SARS-CoV-2 RNA on different surfaces was examined by taking swab samples in one university hospital from the surface of a range of materials. A total of 98 samples were analyzed. These include high-contact materials in the room of 2 patients with proven COVID-19 (mobile phone, food tray, washbasin, door handle of the room and window), surfaces outside the room (floors of corridor and service rooms in the COVID-19 ward, door handles), personal protection materials immediately after disposal (gloves, aprons, masks) and surfaces of all type of waste recipients at the recycling park within the hospital. Samples were obtained by gentle rubbing between 10 and 20 cm² of the surface with a swab moistened in RNA conserving fluid (DNA/RNA Shield, Zymo Research, Irvine, CA). RNA extraction was done with the MagMAX™ Viral/Pathogen II (MVP II) Nucleic Acid Isolation Kit from ThermoFisher (ref: A48383) on KingFisher instruments. The PCR detection was performed using the TaqPathTM COVID-19 CE-IVD RT-qPCR Kit (Thermo Fisher, ref: A28575, targets N-gene; S-gene and ORF1; control MS2) using QuantStudio™ 5 Real-Time PCR Instrument (348-Well Block), Thermo Fisher, Waltham, MA. The results are expressed in Cycle Threshold (CT) values. A CT value lower than 20 is considered a high viral load, between 20-30 intermediate viral load, and more than 30 is the absence of viral load.

The assay included controls, including samples taken in a room of a randomly chosen non-COVID-19 patient and samples taken on the surface of waste recipients originating from non-COVID-19 units.

All data were processed in MS excel. Calculations were done either in excel or in SPSS. Significance was estimated at $p<0.05$. The study was initiated at Ghent University Hospital, Belgium, and the formal approval of the ethical committee was obtained. Not all hospitals could deliver all data, figures are in most cases representative of the general picture.

3. RESULTS

3.1 Evolution of the number of patients, weight, volume, and cost of the medical waste.

Figure 1 shows a typical pattern of the number of COVID-19 patients and non-COVID-19 patients in one university hospital.

During the 2019 control period, a regular sequence of inpatient bed occupations is seen with a drop of about 25% during weekends, a slight reduction in the number of patients during holidays, and a sharp, short-lasting drop during the new year period. This pattern is identical in all hospitals that delivered data. During the first wave in 2020 and in this particular university hospital, the number of non-COVID-19 patients dropped from about 850 to about 500, while the number of COVID-19 patients increased to a maximum of 95. Subsequently, as the number of COVID-19 patients decreased, the number of non-COVID-19 patients gradually increased reaching approximately the same level as before the first wave and just before the second wave. During this second wave, the number of non-COVID-19 patients decreased from 800 to 700 with a parallel increase of COVID-19 patients up to 120. A similar result was found in the other hospitals that delivered the appropriate data (9 out of 14 participating hospitals). The increase in COVID-19 patients was accompanied by a decrease in non-COVID-19 patients and this compensation was more pronounced in the first wave compared to the second.

The effects of the pandemic on the waste volume and weight are shown in Figure 2.

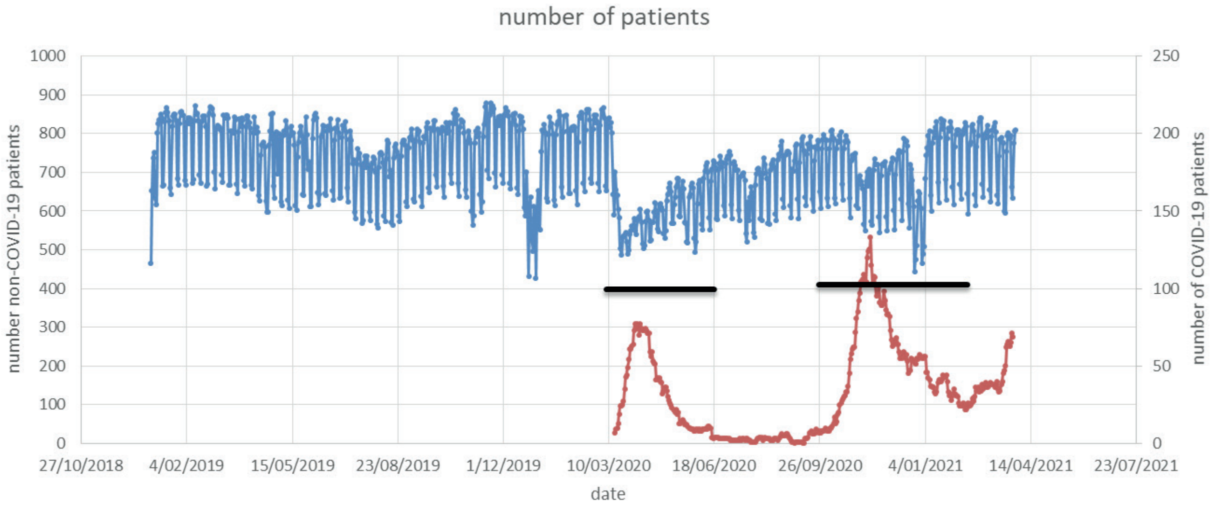

FIGURE 1: Evolution of the number of patients and weight of medical waste. Number of COVID-19 (orange, right y-axis) and non-COVID-19 (blue, left y-axis) patients from 1/1/2019 till 31/12/2020. The black lines refer to wave 1 (1/3/2020 – 20/6/2020) and wave 2 (1/10/2020 – 1/3/2021) of the corona epidemic.

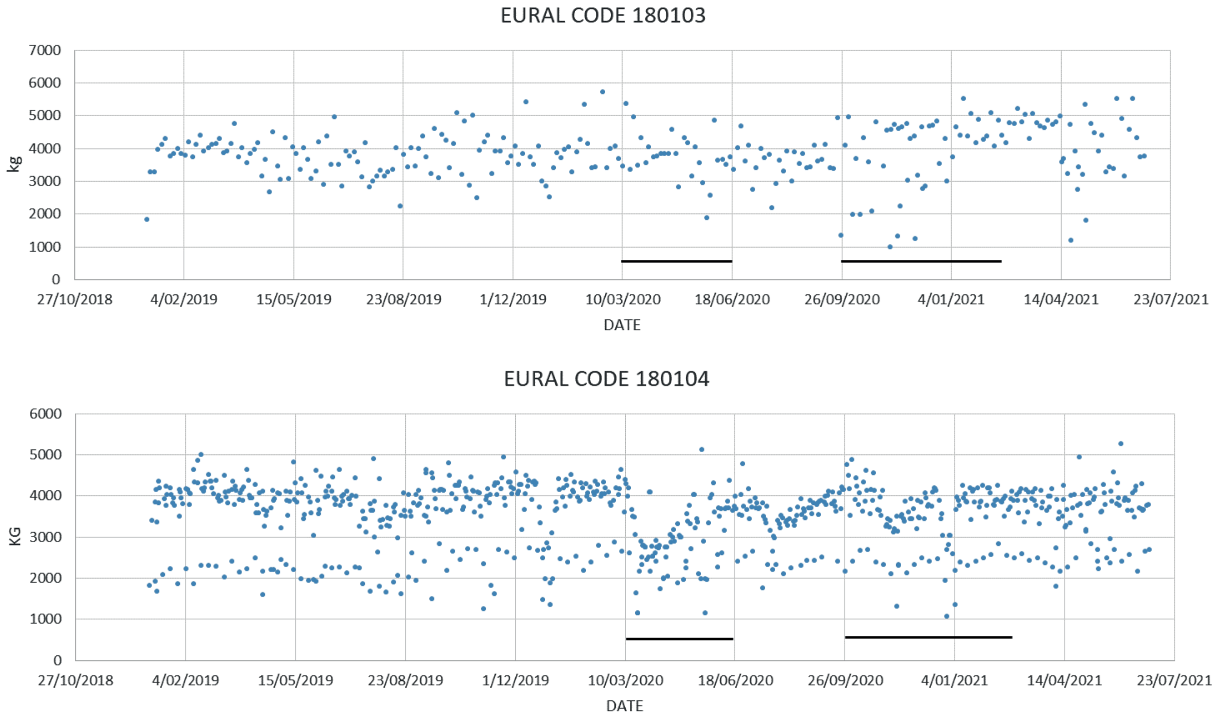

FIGURE 2: Evolution of the number of patients and weight of medical waste. Number of COVID-19 (orange, right y-axis) and non-COVID-19 (blue, left y-axis) patients from 1/1/2019 till 31/12/2020. The black lines refer to wave 1 (1/3/2020 – 20/6/2020) and wave 2 (1/10/2020 – 1/3/2021) of the corona epidemic.

During the first wave, the quantity of the 180104 type of waste shows a tendency to decrease (data from 3 participants) while the quantity of the 180103* waste remained nearly unchanged (data from 9 participants).

The quantitative data on the medical waste EWC 180103* type of waste was further analyzed in more detail in one university hospital; the results are shown in Figures 3 and 4.

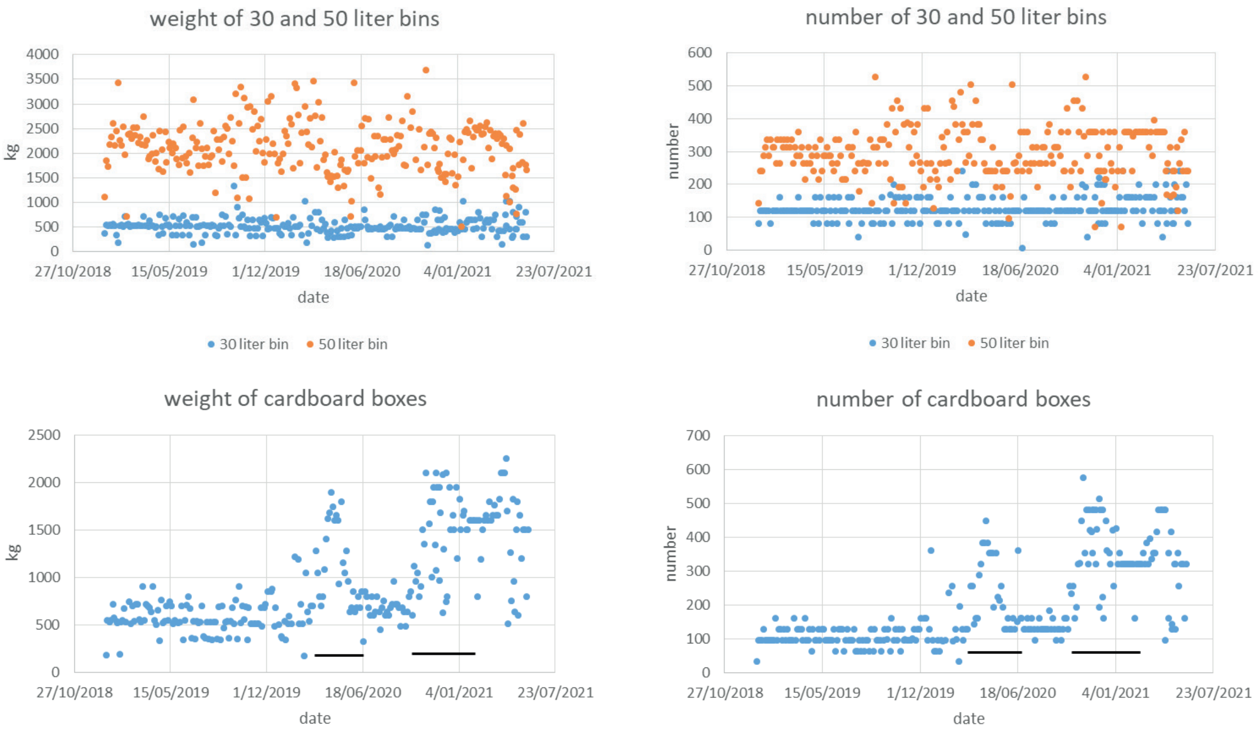

FIGURE 3: Weight (left panel) and number (right panel) of 30 and 50-litre bins (upper panel) and cardboard boxes (lower panel). Each dot represents one transport event from the hospital to the incineration plant. The black lines represent the two COVID-19 waves.

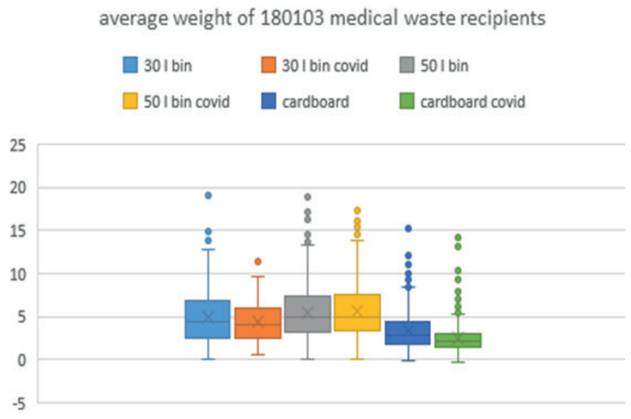

FIGURE 4: B&W plot of the weight of the waste recipients. Significance ($p<0.05$) was obtained for the cardboard containers. The average value of 600-1200 recipients of each type is shown.

The data clearly show that for the 30 and 50-litre bins, the effect of the presence of COVID-19 patients on weight and number of recipients is very limited except for a tendency of the increase in weight of the 50-litre bins during the second wave. The only significant change was a marked increase in the total weight of the cardboard recipient and this increase in weight is mirrored by the increase in the number of recipients leading to a significant increase in total volume. The weight and number of cardboard boxes remain higher in the aftermath of the second wave compared to pre-pandemic figures.

In one hospital, the individual recipients for the 180103* type of waste were weighed during the second wave. The result is shown in Figure 4. The comparison between COVID-19 and non-COVID-19 patients was maintained.

There is no difference in weight between COVID-19 and non-COVID-19 recipients for the solid bins of 30 and 50 litre. The average weight of the cardboard boxes was lower than the weight of the bins and the average weight of the COVID-19-waste containing recipients was significantly lower than the weight of other departments ($p<0.05$).

The observations in individual hospitals are further ascertained by averaging the data on a monthly basis, thereby avoiding the intrinsic variability seen in the first figure, and extrapolating to the total weight of this waste for the whole region of Flanders - Belgium covering about 100 hospitals was analyzed; this is shown in Figure 5.

In 3 university hospitals, the quantity of waste expressed as kg/day/bed was calculated. In 2019 and the years before that, the amount of waste type EWC 180103* was variable depending on the hospital and varied between 0.2 and 1.4. During the subsequent waves of the COVID-19 pandemic, this increased to 5.3, 6.2, and 6.4, respectively. In one hospital, the weight of waste kg/day/bed was monitored during the second wave for two periods of 2 weeks each. On average, 8.5 ± 1.9 and 6.1 ± 2.4 kg/bed/day were found.

The cost of destroying the waste was calculated for 2 partners who delivered sufficient details. The cost during the last 6 months of 2019 was set at 100 %; the results are presented in Figure 6.

This figure is compatible with the information on the quantity of waste. During the first wave, the amount of risk waste was unchanged and consequently, there is no effect on the cost. During the second wave, the quantity of risk waste increased by about 20 % of the control value, and the cost mirrors perfectly this increase to approximately 140 % of the control value.

Additional controls on weight and volume of waste include the registration of residual waste (o.a. packaging materials, plastic bottles, drink cartons….) or green waste. No influence of the pandemic was found (results not shown).

3.2 Is EWC 180103* the correct assignment of the COVID-19-waste?

The results of the PCR assay are shown in Table 2.

Positive samples were found in the room of COVID-19 patients and on materials handled by these patients, such as mobile phones and food trays. Considering the waste recipients, only one weakly positive needle disposal unit was found. Extensive sampling in the waste storage room and at the recycling park on campus yielded no positive results.

4. DISCUSSION

The influence of the SARS-CoV-2 pandemic on societal life is enormous, and long-term negative effects are to be expected (Sarkodie and Owusu, 2020). The initial focus of the biomedical scientific community was evidently on patient care and vaccine development, but it became soon clear that other aspects were affected, in particular environmental and sustainability aspects (Hens and Fraeyman, 2021). Within the broad range of sustainability, interest in medical waste became clear, and alarming messages on the overwhelming and excessive volumes of waste were published both in the scientific and lay press (You et al., 2020, Yang et al., 2021). Over time, this initial panic-like attitude was replaced by a more rational approach (Das et al., 2021) although the available information was still to a large extent contradictory.

The data as presented here are typically highly variable. Furthermore, not all participating hospitals were able to deliver all data for two reasons. The main reason is the overwhelming workload during the COVID-19 waves hindering appropriate engagement to transmit the data. Secondly, it is clear that smaller hospitals in contrast to larger and university hospitals have frequently less developed internal systems for extensive data registration.

The data on the number of both COVID-19 and non-COVID-19 patients are reproducible for all hospitals delivering the data: normal activities were reduced in order to cope with the presumed wave of COVID-19 patients and this reduction is nearly independent of the type of hospital and the number of beds. Consequently, the total number of patients (COVID-19 and non-COVID-19) even decreased during the first wave. During the second wave and in view of the experience gained in the hospitals on how to cope with

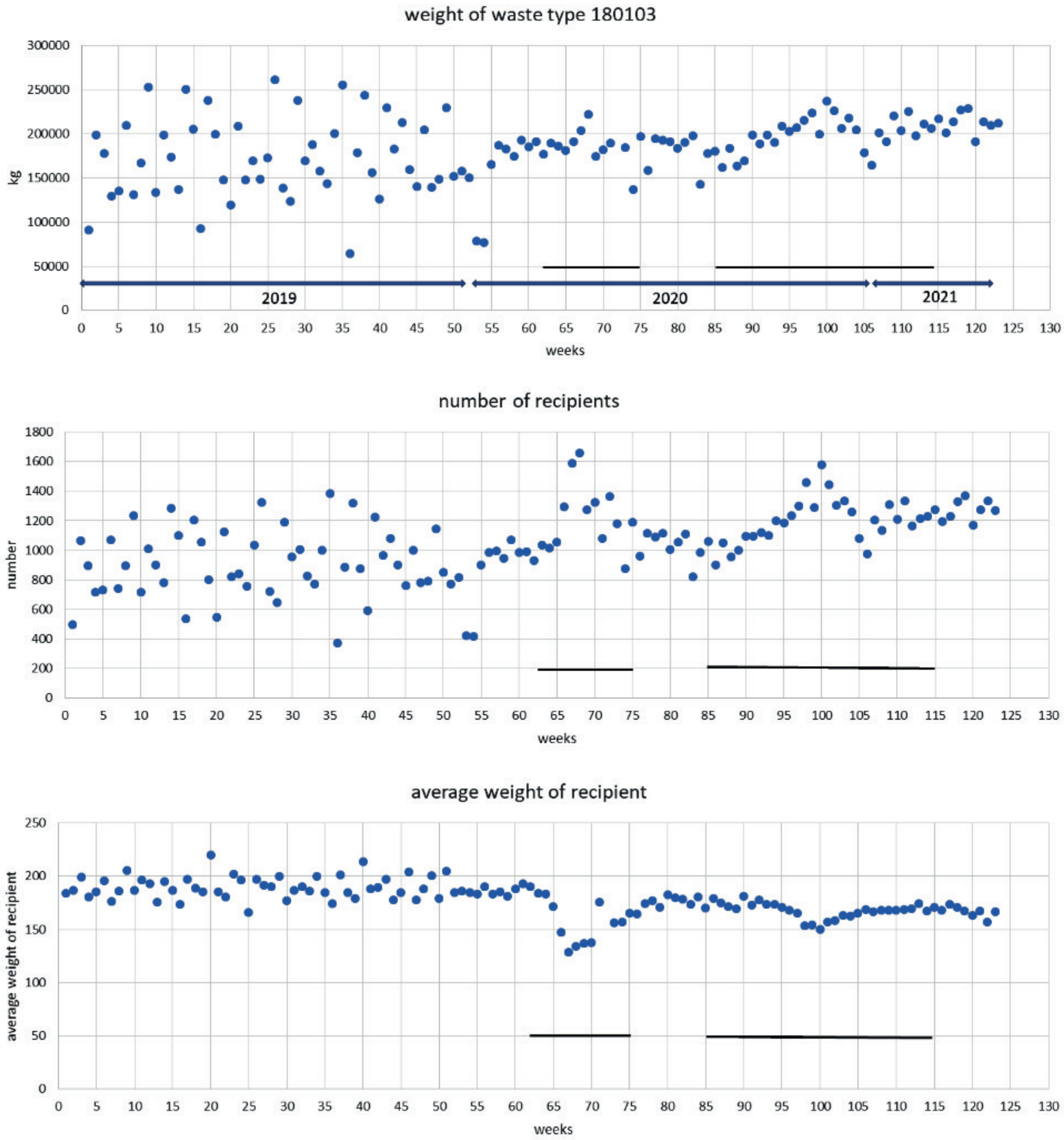

FIGURE 5: Risk waste type EWC 180103* processed at the incineration plant, upper panel: total weight, middle panel: number of recipients, lower panel: average weight of the recipients. X-axis (Time) is expressed in weeks: 2019 from week 1 to 53, 2021 from week 54 to 106, and 2021 from 107 to 123. The black lines represent COVID-19 waves 1 and 2.

the patient flow during the first wave, normal activity was less reduced, resulting in a slightly increased total number of patients. This is highly in contrast to the alarming messages in some countries on the overcrowded hospitals and the pressure on the whole health care system: there is no evidence of an overflow of the hospitals in this study when taking the total number of beds occupied into account. Of note is that at times and particularly during the second wave, the intensive care unit in some hospitals was almost entirely occupied by COVID-19 patients.

The effect of the COVID-19 pandemic on medical waste is more complex than a simple increase in volume and/or weight.

There is a clear difference between the two waves of the COVID-19 pandemic. During the first wave, we found that the total quantity of the EWC 180103* type of waste expressed in kg was virtually unchanged in all hospitals and not different from the situation in 2019. We conclude that the quantity of waste from COVID-19 and non-COVID-19 patients at the level of the individual hospital is in a commu-

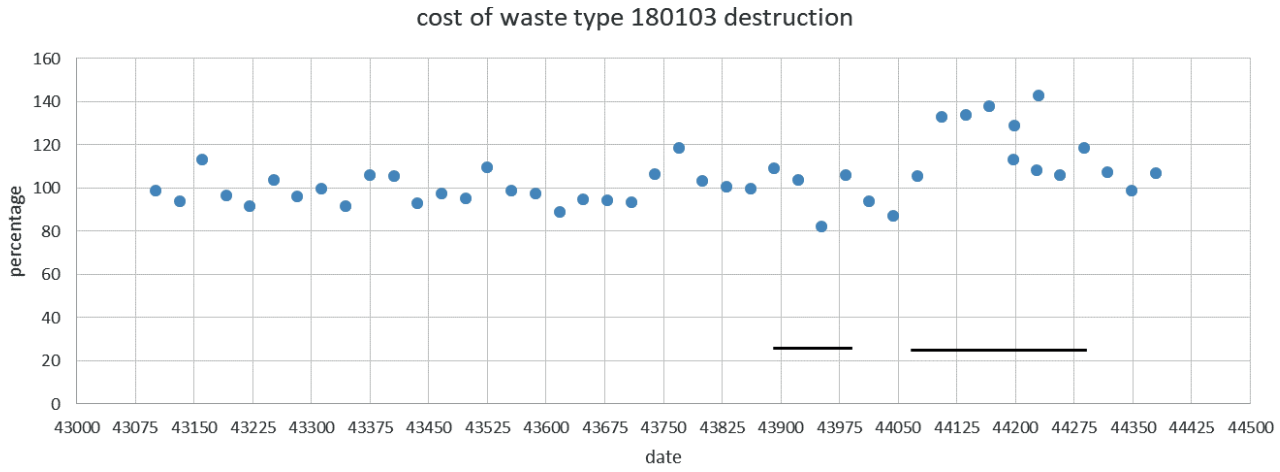

FIGURE 6: Total costs for the destruction of the risk medical waste type EWC 180103*. The average cost for the last 6 months of 2019 was set at 100 %; all other values were calculated as the percentage of the average value. The black lines represent the first and second COVID-19 waves.

nicating vessel system: the reduction of waste because of the reduction of non-COVID-19 patients is compensated by the increase of waste generated by the care for COVID-19 patients. This conclusion is supported by the decrease in weight of non-risk waste (EWC/EURAL 180104) due to the decrease in the number of non-COVID-19 patients which typically produce more non-risk waste, while all the waste from COVID-19 patients was considered as risk-waste.

Although the total quantity of waste within the hospitals remained fairly stable, there is a clear-cut shift in the type of recipient used for transporting the waste showing increased use of cardboard recipients due to the extensive use of personal protection equipment such as gloves, aprons, goggles, and face masks, for treating COVID-19 patients. All of these are lightweight and – mostly not contaminated with blood or bodily excretion products – could be confined to the cardboard recipients. The predominantly lightweight character of COVID-19 waste is convincingly illustrated by the fact that the average weight of the COVID-19 cardboard recipients is significantly lower than that of non-COVID-19 patients. As a consequence, the number of used cardboard recipients increased significantly while the number of 30 and 50-litre bins remained nearly unaltered.

During the second wave, the increase in EWC 180103* type of waste is clear at the level of the region but less obvious for the individual hospitals. The main reason is that many hospitals in the region changed the protocol during or immediately after the first wave because of the growing shortness of the plastic material bins; hence, the waste was no longer considered as category 180103* but as category 180104 eventually including a limited time (72 hours) for quarantine of the waste before destruction. It is clear that this change was less motivated by knowledge of the risks of infection caused by the waste but for logistic reasons. While during the first wave, the numbers of COVID-19 patients and non-COVID-19 patients were in balance as far as the production of waste is concerned, in the second wave, both types of patients were no longer balanced and an in-

TABLE 2: Results of the analysis of SARS-CoV-2 RNA on different inanimate objects. The results are expressed as positive (P) with CT < 20, or negative (N) with CT > 30. CT values between 20 and 30 were considered weakly positive. The number of samples for the control patients and for the COVID-19 patients is 1 for each of the objects mentioned in the table. Otherwise, the number of samples is indicated.

Room, location	samples	Positive (P) or negative (N) PCR test
Room of a non-COVID-19 patient	Mobile phone, food tray, door handle, patient handset, washbasin	N
Room of COVID-19 patient 1	Food tray, washbasin, door handle, table, patient handset, waste recipient, window	N
	Mobile phone, floor	Weakly Positive
Room of COVID-19 patient 2	Patient handset, floor, waste recipients	P
	Mobile phone, food tray, washbasin, door handle, table trapeze bar	Weakly positive
Personal protection material after use	Aprons (n=11), masks (n=6), gloves (n=11)	N
Waste storage room in COVID-19 ward	All filled recipients: plastic bins (n=7), cardboard boxes (n=7), plastic bags (n=14)	N
Hallway and service rooms in COVID-19 ward	Door handle, sharps container (n=1)	N
	Floors (n=5)	P
Waste recycling park in the hospital	All waste recipients from the COVID-19 ward (n=15)	N
	All waste recipients from non-COVID-19 wards (n=9)	N

crease in waste was observed. The cost for waste destruction is compatible with this evolution although most of the increase is due to costs of logistics, sub-optimal transport due to increased volume, costs of extra bins, etc. The cost for incineration in EUR/kg is unchanged.

All data indicate that more waste is generated through the care of COVID-19 patients compared to other patients. This is obvious from the figures obtained from 3 university hospitals and amounts to between 5 and 8 kg/day/bed. This is significantly higher than the normal historical value obtained in control conditions (between 0.2 and 1.4 kg/bed/day). Literature data indicate similar figures : (all in kg/day/bed) 3.4 (Haque et al., 2021), 3.64 and 2.7 (Yang et al., 2021) and 3.95 (Abu-Qdais et al., 2020). The abundant use of personal protection equipment when taking care of COVID-19 patients is obviously the main reason for this difference.

Numerous alarming messages on the possibility that medical waste from COVID-19 patients should be considered as a transmission route for the virus were spread through scientific literature and the lay press. Reports on the survival of the virus on different surfaces gave additional way to concern (Bedrosian et al. 2021, Cheng et al., 2020, Hoseinzadeth et al., 2020, Huraimel et al., 2020, Kampf et al., 2020, Pastorino et al., 2020, Peng et al., 2020, Ren et al., 2020, Van Doremaelen et al., 2020; Wang et al., 2020). Mobile phones were identified as a possible pathway for microbial transmission (Olsen et al., 2020). The dependency on environmental conditions of the infectivity of SARS-CoV-2 and of other coronaviruses present on surfaces was demonstrated (Bueckert et al., 2020). However, most of these studies are done under experimental laboratory conditions with high virus loads and optimal environmental conditions of humidity and temperature. It is to be expected that the real, in-hospital conditions are far from the optimal experimental conditions; hence the reliability of the above-mentioned results is questionable as shown by others (Ben-Shmuel et al., 2020, Cheng et al., 2020, Goldman, 2020, Huang et al., 2020, Hororo et al., 2020, Ong et al., 2021, Meyerowitz al., 2020, Shah et al., 2021, Wei et al., 2020). Our results confirm the latter observations: nearly all surfaces touched by the patients are positive – as expected – but except for one tray in the room of a patient which was slightly positive, all surfaces of waste recipients were negative. During sampling, care was taken to mimic worst-case scenarios i.e. circumstances in which most chance for infection would occur: the patients in the room were diagnosed with COVID-19, the CT-value was low (below 20) suggesting the presence of active virus, samples were taken in different locations in the COVID-19 ward including the waste collection room where waste is stored after providing medical care to the patient. Samples at the recycling park were taken upon arrival at the park of the waste from the COVID-19 ward. We clearly demonstrated that no contamination with SARS-CoV-2 RNA was found on the surface of any of the waste recipients manipulated at the recycling park of the hospital. This indicates that the viral RNA does not survive very long on these surfaces and even the one slightly positive sample found in the room of the patient has apparently no consequences. Even more important, none of the personal protection material, used in close contact with the patients, was positive. A similar finding has been published before (Ong et al., 2020, Huang et al., 2020). We believe that in real-life conditions, these materials are not able to retain any RNA on their surfaces. Two important restrictions are to be made. The RT-PCR test only provides evidence for the presence of SARS-CoV-2 RNA (Ben-Shmuel et al., 2020). Hence, a negative PCR test is meaningful, a positive PCR test reveals the presence of RNA but does not distinguish between viable and non-viable viruses. However, Mondelli et al. (2021) demonstrated that SARS-CoV-2 RNA recovered from a positive swab was unable to grow on Vero E6 cells, demonstrating that these fomites are unable to infect people. Until now and to the best of our knowledge, no case of transmission of SARS-CoV-2 through waste has been documented, which is compatible with our findings. Secondly, our conclusions are valid for the virus-RNA species that were the dominant infectious variants at that time, In theory, extrapolation of our conclusions to other variants e.g. the actual dominant infectious omicron variant should be done cautiously. However, no data are available on the variants which might suspect a difference in virus-surface interactions, As discussed in extenso by Castano et al. (2021), the most important physicochemical characteristic of the viruses in relation to virus-surface interaction is the presence of the viral envelop which is likely to be comparable for all coronaviruses. Hence, it can be expected that the properties of the virus particles from different variants of the SARS-CoV-2 related to survival outside the patients on the surface of innate subjects are less affected compared to e.g. capacity to infect people.

5. CONCLUSIONS

In conclusion, the dependency of the quantity and volume of medical waste on the COVID-19 pandemic is complex and dominated by the balance between the number of COVID-19 and non-COVID-19 patients. The shift from EWC type 180103* to 180104 waste is plausible because we provided additional evidence that SARS-CoV-2 RNA is absent at the surface of all waste recipients investigated, and that waste originating from the care for COVID-19 patients, in particular the personal protection equipment, is likely to be of the EWC/EURAL type 180104.

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SPATIAL VARIABILITY OF GAS COMPOSITION AND FLOW IN A LANDFILL UNDER IN-SITU AERATION

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ABSTRACT

In-situ aeration of landfills accelerates biodegradation of waste organic matter and hence advances waste stabilization. The spatial outreach of aeration greatly affects stabilization efficiency. This study analyzed the spatial variability of gas composition and flow in 230 wells spread over four compartments of a Dutch landfill which is under in situ aeration since 2017, as well as the carbon extraction efficiency, temperature, and settlement. Flow rates and gas composition in the extraction wells varied strongly. The highest variability was observed in the compartment with the highest water tables with submerged filter screens for most wells, with low flow rates, and elevated ratios of CH_4 to CO_2, indicating predominance of anaerobic processes (compartment 11Z). The compartment with the most uniform distribution of gas flow rates, composition and lower ratios of CH_4 to CO_2, suggesting a significant share of aerobic carbon mineralization, also showed higher temperatures, a carbon extraction efficiency, and larger cumulative settlement, all indicative of enhanced microbial activity (compartment 11N). In this compartment, the amount of extracted carbon exceeded the carbon generation predicted from landfill gas modeling by the factor of 2 over the hitherto four years aeration. The effect of water tables on gas flow and the correlation between the flow, and the ratio of CH_4 to CO_2 appeared weak, indicating that also other factors than water tables influence gas concentration and flow. Future work includes stable isotope probing to analyze the significance of microbial respiration and microbial CH_4 oxidation for the composition of the final extracted gas mixture.

1. INTRODUCTION

In-situ aeration is considered a possible method for the stabilization of landfills, reducing waste reactivity and the landfill's emission potential faster than under anaerobic conditions and therefore also reducing the time for monitoring during aftercare (Erses et al., 2008; Grossule & Stegmann, 2020; Ritzkowski et al., 2006). On a field scale, an important factor for the efficacy of treatment by in-situ aeration is the spatial distribution of air and gas throughout the waste body (van Turnhout et al., 2020), controlling (a) the reduction of the remaining methane production potential, (b) the desired increase in organic matter decay by microbial respiration and (c) the extent of methane oxidation within the landfill fulfill. However, homogeneous aeration on a field scale is a challenge not easy to reach (Ritzkowski & Stegmann, 2012), and is mainly limited by zones of water saturation and preferential liquid flow paths (Fellner & Brunner, 2010; Hrad et al., 2013). The spatial distribution of gas flow also depends on the operational conditions of aeration. For example, a model developed to find an optimal aeration strategy in a landfill indicated that air injection reaches a larger volume fraction of waste with higher airflow, but extraction appears to achieve a more homogeneous distribution of oxygen throughout the waste body (van Turnhout et al., 2020). Most of the in-situ aeration strategies use low-pressure aeration, which usually considers pressure within the range of 20-80 mbar (Ritzkowski & Stegmann, 2012). For optimal performance of in-situ aeration, the permeability of the waste body to both gas and water has to be considered (Ritzkowski & Stegmann, 2012; Xu et al., 2020).

Within the Dutch sustainable landfilling program iDS (Sustainable Landfill Foundation, n.d.), four compartments of the landfill Braambergen located near the city of Almere (The Netherlands), have been aerated since September 2017 (Cruz et al., 2021; Lammen et al., 2019, 2021). The waste comprises mainly soil and soil decontamination residues and around 15% of organic waste (more detail in section 2.1). Two different aeration strategies were em-

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ployed: over-extraction and combi-aeration (low-pressure aeration). The over-extraction method creates a suction pressure by extracting more gas than gas produced, thereby causing ambient air to intrude into the landfill. Low-pressure aeration combines air injection and gas extraction considering lower pressure for injected air than extracted gas (Vereniging Afvalbedrijven, 2015). This paper researches the spatial variability of landfill gas composition and flow under the operational conditions of combi-aeration. It was hypothesized that based on the variability of the water tables detected in the injection-extraction wells (Gebert et al., this issue), neither the flow of the extracted gas nor the flow of the injected air is distributed uniformly. Consequently, the enhancement of biodegradation would differ spatially, and this would be reflected in the landfill gas (LFG) composition. To better understand which factors cause this spatial variability, data on gas composition was combined with data on gas flow rates and water columns in all compartments. The ratio of CH_4 to CO_2 was used to assess the spatial variability of the impact of aerobic and anaerobic processes on the measured gas composition. Further, this study analyzes the temporal variability of carbon extraction efficiency, temperature, and settlement for the period 2017-2020. It was hypothesized that compartments with higher aeration efficiency would show a higher carbon extraction efficiency, higher temperatures in relation to higher biodegradation rates, and higher settlement rates.

2. MATERIAL AND METHODS

2.1 Description of site and aeration system

The Braambergen landfill is located near the city of Almere in the northern part of the Netherlands, with the four pilot compartments 11North (11N) and South (11Z) and 12 East (12O) and West (12W) in operation from 1999 to 2008 on a surface area of approximately 10 ha. The pilot compartments contain around 1,200,000 tons of waste, mainly composed of soil and soil decontamination residues (80.6%) (Cruz et al., 2021; Lammen et al., 2019). Figure 1 shows a timeline of landfilling considering the different compartments and the main waste components, including soil and soil decontamination residues, construction and demolition waste, commercial waste, shredder, street cleansing waste, coarse household waste, sludge, and household waste. Compartments 12O and 11Z comprise mainly soil and soil decontamination residues (95.8%) and are approximately the same age. The oldest compartment is 12W and has the lowest percentage of soil and soil decontamination residues. This compartment together with compartment 11N are the ones that were filled with commercial waste and household waste (2.7% for compartment 11N). Compartment 11N is also the one with the longest active landfilling period.

Landfill stabilization through in-situ aeration is carried out since September 2017. A network of 230 wells spaced at 15 to 20 m distance over the four compartments (Figure 2, left) can be operated in an over-extraction or combi-aeration mode (Figure 2, lower right). From north to south, each aeration line is denoted by a letter (A to W), and from west to east, each well by a number (1 to 8-11) (Figure 2, top right). All wells are deep filtered with the filter screen over a height of 1.8 m from the bottom of the well, which has been inserted to a total depth of up to 10-12 m below the landfill surface, corresponding to approximately 2 m below the Amsterdam Ordnance Datum (NAP) into the waste body.

2.2 Data acquisition, selection and processing

Flow velocity and temperature (multifunctional handheld unit, Höntzsch U426, TA-10 probe), gas composition (Geotech Gas Analyzer GA2000; detection limit 0.1%), and the pressure (pressure gauge by Blue line S4600) were measured manually on a monthly interval. At the gas blower station, landfill gas (LFG) flow rate (Proline Prosonic Flow B 200 Ultrasonic flowmeter for gas extraction, and Proline 65i T-mass flowmeter for air injection; both Endress+Hauser),

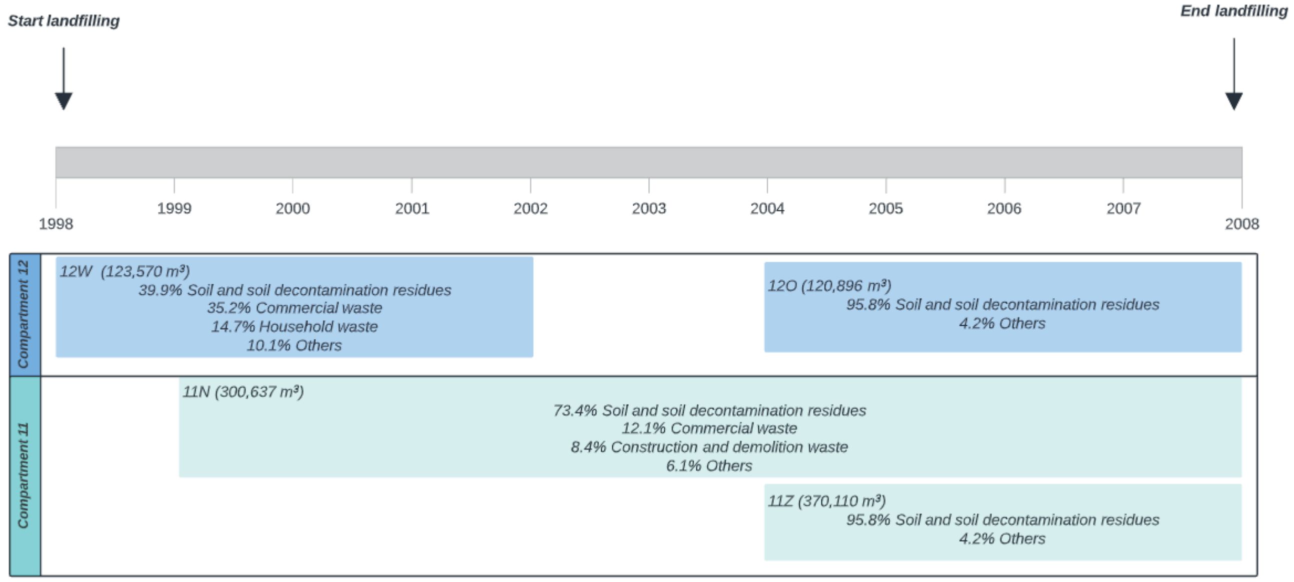

FIGURE 1: Timeline of waste landfilling per compartment.

pressure (PMC51-R8K2/0; Endress+Hausser), gas composition (CH_4, CO_2, and O_2, Biotech Gas Analyzer 3000), and temperature (iTEMP TMT181; Endress+Hausser) were continuously measured and recorded every fifteen minutes. The data from the gas blower station with the same collection time as the individual well sampling was selected for analysis. To obtain the flow rate for each well, the normalized flow (at standard temperature and pressure) from the gas extraction blower were averaged over the period in which the manual measurements were carried out, then divided by the individual wells in proportion to the velocity measured at each well. This study presents data from March 2020.

Using Python, MS Excel, and Origin software packages, the spatial variability of gas composition, flow, and water columns were visualized and analyzed. Based on the gas composition and the ratio of CH_4 to CO_2, the percentage of anaerobic activity (PAA) was calculated for each well (Yazdani et al., 2010) as follows:

$$PAA = \frac{2[CH_4]}{2[CH_4]+([CO_2]-[CH_4])}\ x\ 100 \tag{1}$$

where $[CH_4]$ and $[CO_2]$ are the measured concentrations (% v/v) of CH_4 and CO_2. PAA was then normalized to the maximum observed value to obtain the normalized anaerobic activity. The difference of 1 was considered as the normalized aerobic activity (NAA, Equation 2):

$$NAA = 1 - \frac{\mathrm{PAA}}{PAA_{max}} \tag{2}$$

where PAA_{max} is the highest percentage of anaerobic activity.

The coefficient of variation for gas composition and flow was calculated by dividing the standard deviation and the mean for all the datasets in each compartment. The spatial variability was analyzed using the Rijks-Driehoek (RD) coordinate system from the Dutch Geographical service in EPSG projection 28992 (Amersfoort datum). The relationship between the individual parameters was tested using Pearson's coefficient r. The number of observations, the variability of the parameters, and the confidence level determined the significance of this correlation.

Using the ideal gas law, the gas concentration, flow rate, pressure, and temperature, the extracted carbon in kilograms of carbon per hour was calculated. The carbon extraction efficiency was calculated using the amount of extracted carbon normalized to the number of wells and tons of waste in each compartment.

The temperature in compartments 11N and 11Z was obtained using a Silixa Ultima XT-DTS distributed temperature sensor which performed a double-ended measurement every 0.5 m using a long section of glass fiber distributed over 12 wells in each compartment (11N and 11Z) between -2 m and 9 m with respect to the NAP. For data configuration and collection, Silixa software was used. The height of the landfill surface and hence landfill settlement was measured twice per year on a network of 43 settlement beacons distributed over the four compartments using a TRIMBLE R8-2 rover GNSS Receiver.

3. RESULTS AND DISCUSSION

The following sections present the spatial variability of CH_4 concentrations, the ratios between CO_2 and CH_4, methane and total gas flow rates, and temperature in the individual wells. Gas-related parameters are only taken into account for the 110 wells that are operating in extraction mode during simultaneous air injection and gas extraction on alternative wells (combi-aeration) in March 2020.

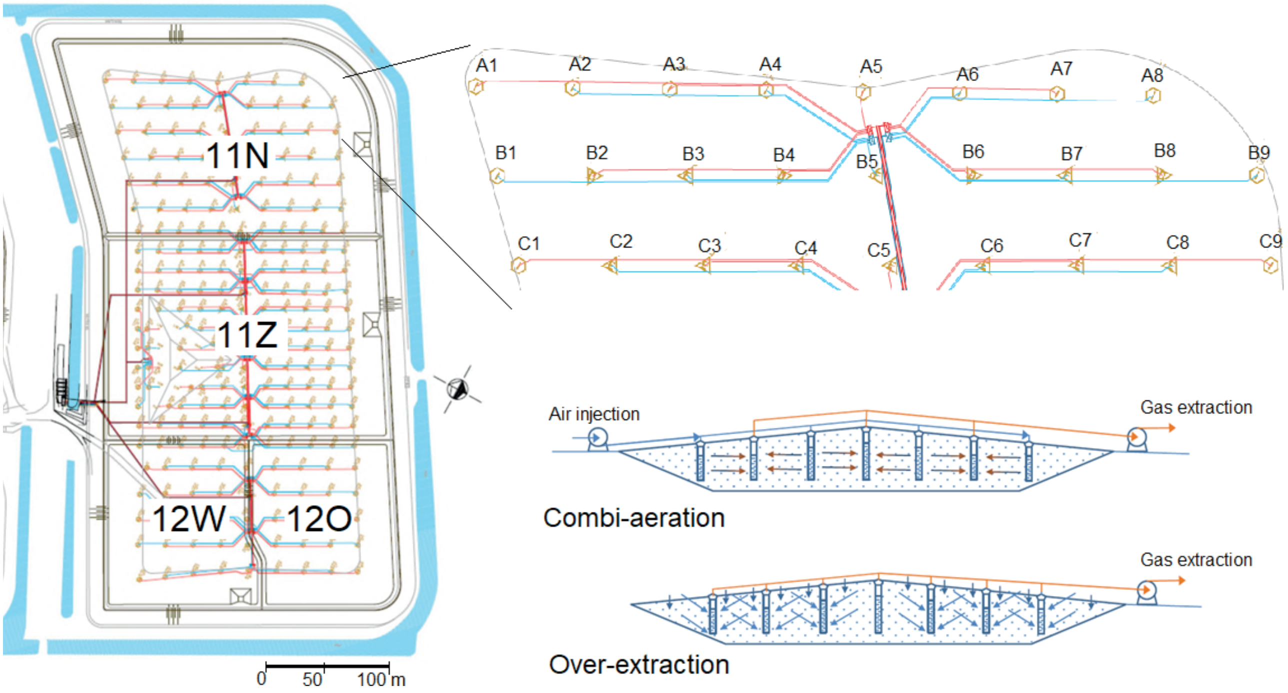

FIGURE 2: Network of aeration wells, systematics of well labeling and a schematic representation of combi-aeration and over-extraction.

3.1 Variability of gas composition

The CH_4 concentration in aeration wells of the compartments 11N, 11Z, 12W, and 12O of Braambergen landfill in March 2020 was not spatially uniform, with the largest range observed in compartments 11N, 11Z, and 12O (Figure 3). In those three compartments, CH_4 concentration varied between close to zero and higher than 40%. High CH_4 concentrations are related to anaerobic waste degradation processes, whereas low CH_4 concentrations are related to CH_4 oxidation processes, as well as to non-existent or reduced (anaerobic) landfill gas formation, for example as a result of aeration. Little information is available from the literature regarding the variability of CH_4 concentrations in waste bodies. However, high variability in CH_4 concentrations in 90 cm depth of an old municipal solid waste landfill (Röwer et al., 2011) have been reported, and the high variability of surface CH_4 emissions is well known (Giani et al., 2002; Mønster et al., 2019; Rachor et al., 2013; Spokas et al., 2003). As the spatial patterns in surface CH_4 emissions are not only impacted by the heterogeneity of waste composition and flow paths within the waste, but also by the variability of cover soil permeability. The comparability between the dynamics of the waste body's gas phase and surface emission is limited.

The ratio of CH_4 to CO_2 and the normalized aerobic activity (NAA) in the extracted gas (Figure 4) provides information about the share of aerobic to anaerobic processes occurring in the waste body. In compartment 11N most of the wells produced more CO_2 than CH_4 (ratios CH_4:CO_2 below 1), indicating the dominance of aerobic activity in most of its wells compared to the other compartments. Aeration in this compartment is likely to be more efficient due to the lower water tables (Gebert et al., this issue), allowing for an increased level of aeration and therefore increased CO_2 production from both, respiration of waste organic matter and CH_4 oxidation. For an old municipal solid waste landfill, it showed that the onset of aeration led to a decline of the ratio of CH_4 to CO_2 to values below 1 in most of the control wells. However, in many of the monitoring wells, especially those intercepting the deeper layers, this ratio increased again after the three year aeration period was terminated, suggesting that the effect of aeration on waste stabilization was limited by the vertical distribution of the injected air and that therefore stabilization had not been completed (Hrad & Huber-Humer, 2017).

Compartment 11Z had the highest water tables and the largest spread with the ratios of CH_4 to CO_2 ranging between 0 and 7, and the NAA varied between 0 and 0.8, as was also observed in compartment 12O.

In compartment 12W most wells produced the same or higher concentration of CH_4 than CO_2, indicating a predominance of anaerobic waste degradation. This was consistent with the low aerobic activity found in 8 out of 12 wells (NAA<0.5, Figure 4 right). Overall aeration appeared to be most effective and uniform (Figure 5) in compartment 11N.

The spatial distribution of CO_2 follows a similar behavior, with the highest variability in compartment 11Z (Figure 5). The fraction of CO_2 increases under conditions of aerobic waste degradation (respiration) compared to anaerobic conditions when also CH_4 is produced besides CO_2. On the other hand, the O_2 concentration in compartment 11Z was more homogeneous than other compartments, with a coefficient of variability of 0.3. Higher O_2 concentrations were found in compartment 11Z, with most of the values close to 21%, reflecting near-atmospheric concentrations. The fact that the extracted gas has a similar percentage of O_2 as the injected air suggests short-circuiting of atmospheric air along with the wells and consequently a reduced extent of aeration of the waste body in the area of influence of the well, likely maintaining the anaerobic conditions. In general, the N_2 concentration varies the least, in line with the fact that N_2 is non-reactive and therefore only affected by variability in transport processes. The observed spatial variability in the gas composition is likely to be closely related to water content and water tables in the landfill, increasing the resistivity to the airflow, hence limiting the aeration efficiency (Hrad et al., 2013).

Although the CH_4-CO_2 ratio and the calculated share of aerobic activity (NAA) give an estimation of aerobic activity, the contribution of the individual processes (respiration, CH_4 oxidation) to the final value is unclear and shall be further investigated using stable isotope probing that could elucidate the significance of anaerobic and aerobic processes for gas composition in the wells. Those wells producing a mixture significantly impacted by CH_4 oxidation should show enrichment in ^{13}C (Cabral et al., 2010; Chanton et al., 2008; Gebert & Streese-Kleeberg, 2017) whereas wells with low gas generation, or in wells where LFG is diluted by air short-circuiting, the isotopic signature of CH_4 should be similar to that of the original landfill gas.

3.2 Variability of gas flow

Compartment 11N showed both higher total flow rates and higher CH_4 flow rates (Figure 6). Gas flow rates in com-

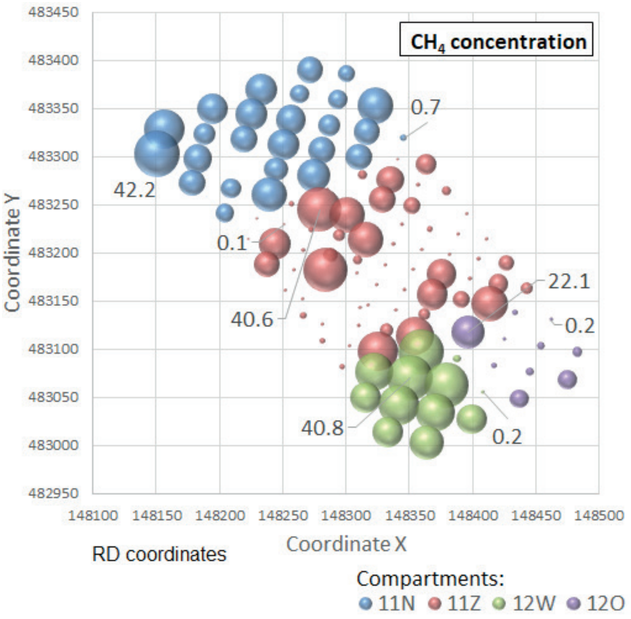

FIGURE 3: Spatial distribution of CH_4 concentration (vol.%) in the extraction wells - March 2020. The size of the symbol is indicative of CH_4 concentration.

partment 11Z were significantly lower, with 35 out of 64 wells showing no flow, presumably in connection with high water tables (Gebert et al., this issue). Compartments 12W and 12O seem to be more homogeneous despite the outliers in wells W2, W4, and S8. The spatial pattern of total flow rate and CH_4 flow rate is similar for compartments 11N and 11Z, but similarities are less for compartments 12W and 12O, in which some wells produce a high total flow rate at low CH_4 concentration and vice versa.

The variability of CH_4 flow rates was similar to the gas concentration variability in each compartment, i.e. compartments 11N and 12W showing a lower variability and compartments 11Z and 12O a higher variability (Figure 5). Considering the waste composition and age, the similarity in gas composition and flow between 11N and 12W and between 12O and 11Z was expected. The highest values for the variability of CH_4 and CO_2 flow rates and concentrations were found in compartment 11Z.

To analyze possible correlations between the flow rate (for conditions of flow > 0) and the ratio of CH_4 to CO_2 in the extraction wells, Pearson's coefficient was calculated. In the case of airflow limiting the aerobic processes, an inverse correlation should be visible, i.e. higher flow rates correlating with higher CO_2 concentrations and therefore a lower ratio of CH_4 to CO_2 in extracted gas. On a confidence level of 99.95%, the flow and the ratio of CH_4 to CO_2 for the total dataset (n=73) were indeed negatively correlated (-0.23), albeit on a low level. With some variation between the compartments (12W: n=12, -0.74; 12O: n=10, 0.01; 11N: n=24, -0.29; 11Z: n=27, -0.10) indicating that the relation-

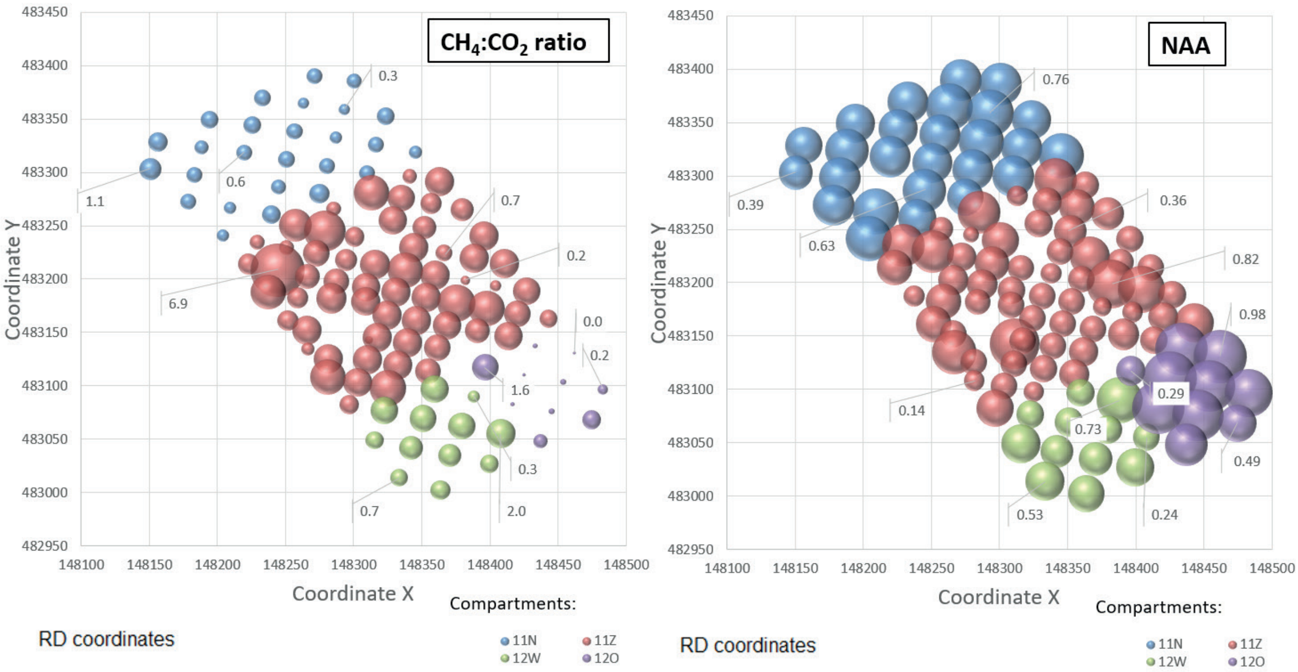

FIGURE 4: Spatial distribution of the ratio CH_4 to CO_2 in the extraction wells (left) and percent of anaerobic activity (right) - March 2020. The size of the symbol is indicative of the ratio CH_4 to CO_2.

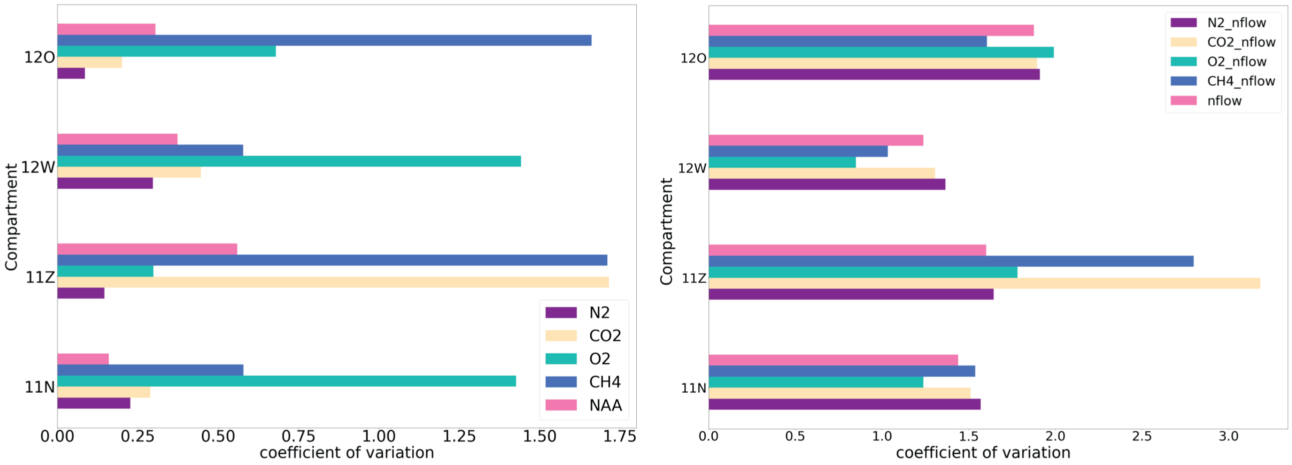

FIGURE 5: Coefficient of variation for the concentration of CH_4, CO_2, O_2, and N_2, and normalized aerobic activity (NAA) (left) and for flow rate, and flow rates of CH_4, CO_2, O_2, and N_2 in the gas extraction wells - March 2020.

ship between the two parameters is confounded, for example, by short-circuiting or near-well differences in waste permeability and hence efficiency of aeration.

The gas prognosis for the aeration pilot (total of all four compartments), carried out with the Afvalzorg Multiphase Model (available from https://www.afvalzorg.com/landfill-gas/lfg-models), estimated the total carbon generation (CH_4-C + CO_2-C) of ~1010 t C for the years 2017 to 2020 if the compartments would not have been aerated (Table 1). Overall compartments, the extracted C exceeded this estimate by a factor of 1.5. A more detailed look at predicted C generation versus realized C extraction reveals that for the wells aerated in compartment 11N, showed the highest C extraction efficiency and highest cumulative settlement, 436 t C more were extracted than predicted to be generated, increasing the factor to > 2, while C extraction in the other compartments remained more or less in the order of magnitude of predicted (anaerobic) C generation. C generation was only modeled for compartment 12 as a whole; hence it is possible that the higher C extraction efficiency calculated for 12W was masked by the less efficient compartment 12O.

While the measured C extraction is deemed accurate, gas generation modeling is subject to high uncertainty (e.g., Scharff & Jacobs, 2006). For example, due to often insufficient data on the waste composition and/or model assumptions on the degradable fraction or kinetic parameters that do not match the deposited waste. The comparison of absolute differences between predicted C generation and extracted C should thus be undertaken with caution. Further, the extracted carbon relates strictly to the gas phase and does not include carbon released from waste biodegradation that leaves the waste body as dissolved organic or inorganic carbon with the leachate. Adding this fraction would increase the share of carbon released in comparison to the predicted carbon generation (landfill gas modeling).

3.2.1 Effect of water tables on gas flow

Analysis of the spatial distribution of water tables over the four compartments showed that most of the wells in compartment 11Z have water tables over the screened part of the aeration well (1.8 m; Gebert et al., this issue). Compartment 11Z is also the one with lower flow rates, higher variability in CH_4 concentrations, and higher ratios of CH_4 to CO_2 (Figures 3 to 6), the latter two suggesting a higher share of anaerobic processes.

The water tables in the waste impede landfill gas and airflow through the wells. It is expected to have a decrease in the flow while the water table gets closer to the top of the screening part of the aeration well. Figure 7 on the far right shows the theoretical relationship between gas flow and height of the water table in the wells, given the location of the filter screen in the lower 1.8 m. For all compartments, the outer boundary of the data approximates this relationship. However, in all compartments also lower gas flow rates were measured than would have been expected from the observed water table, indicating that other factors than the height of the water table, such as the (variable) permeability of the surrounding waste body (Gebert et al., this issue; Xu et al., 2020) also impact aeration efficiency. This was especially pronounced for compartment 11Z in which a high number (98 out of 132 wells) of 'no flow' wells were detected. It can also be seen that the magnitude of gas flow varied per compartment, with at comparable pressures compartment 11N achieving the highest gas velocities.

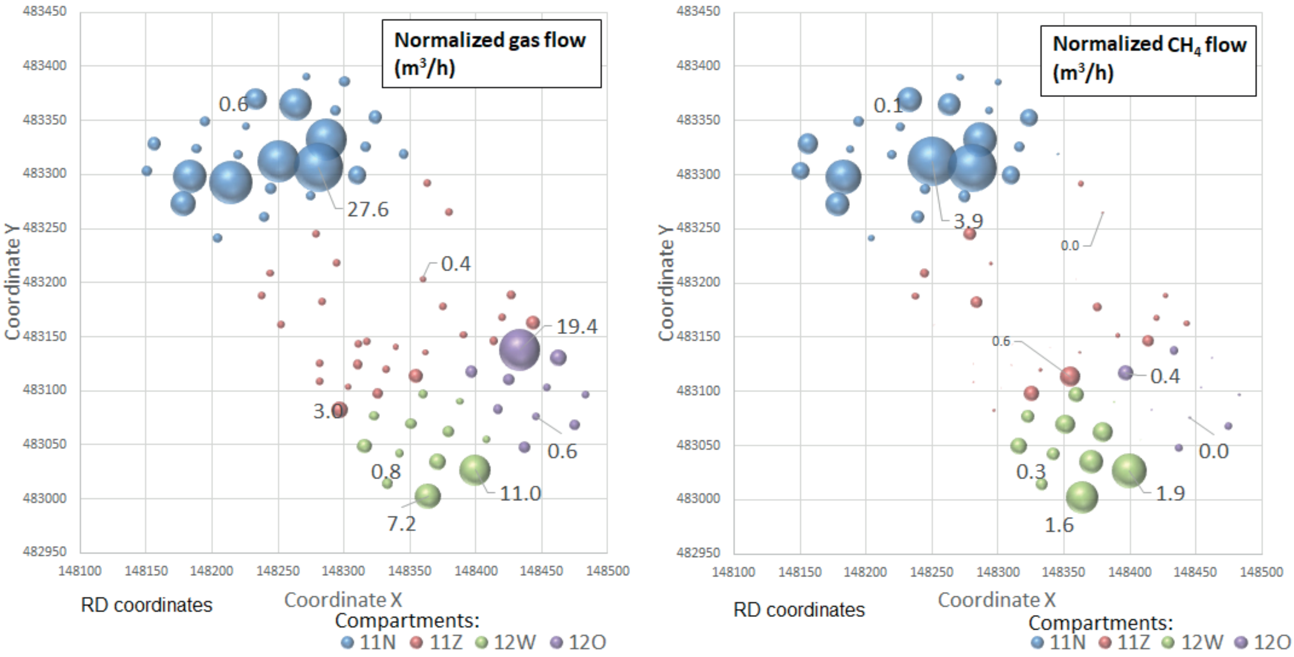

FIGURE 6: Spatial distribution of normalized flow rates (m_3/h) (left) and CH4 flow rates (right) - March 2020. The size of the symbol is indicative of (CH_4) flow rate. Data in graph = exemplary flow rates and CH_4 flow rates.

TABLE 1: Predicted average landfill gas generation and measured average landfill gas extraction for 2017-2020.

Compartment	Predicted cumulative carbon generation (CH_4-C + CO_2-C) 2017-2020 (tons C)	Measured cumulative carbon extraction (CH_4-C + CO_2-C) 2017-2020 (tons C)
11N	382.36	817.82
11Z	193.01	291.60
12W + 12O	435.12	398.87
Total	1010.48	1508.29

3.3 Temporal variability of carbon extraction efficiency, temperature and settlement

As seen from Figure 6 (data for March 2020), compartment 11N showed higher carbon flow rates than the other three compartments. To analyze whether this enhanced performance is consistent over time, the carbon extraction efficiency (carbon flux per compartment normalized to the number of wells and waste volume), waste temperature, and cumulative settlement were analyzed. The carbon extraction efficiency was calculated for the four compartments from the beginning of the aeration in 2017 to 2020 (Figure 8). It is seen that, plausibly, the compartment with the higher aeration efficiency, i.e. the higher share of aerobic processes (NAA, Figure 4) also showed by far the highest carbon extraction efficiency. Compartment 12W showed the second highest carbon extraction efficiency, followed by 11Z and 12O, reflecting the order already seen from the CH_4 flow (Figure 6, right). The differences between the compartments were consistent over the four years since the onset of aeration in 2017.

Figure 9 shows the temperature in compartments 11N and 11Z at +9 m, +6 m, +2 m, and -2 m with respect to the NAP. The landfill surface is between +8 m to +10 m NAP, hence the temperature at +9 m NAP reflects near-surface effects in the landfill, showing the expected seasonal variability with lower temperatures at the beginning of the year and higher temperatures during summer (Figure 9, top left). In the underlying layers, the temperature is influenced by processes within the waste body. The higher aeration efficiency and hence the higher carbon removal efficiency in compartment 11N, as discussed above, reflects clearly the higher waste temperatures due to enhanced biodegradation rates, releasing more heat. Although both com-

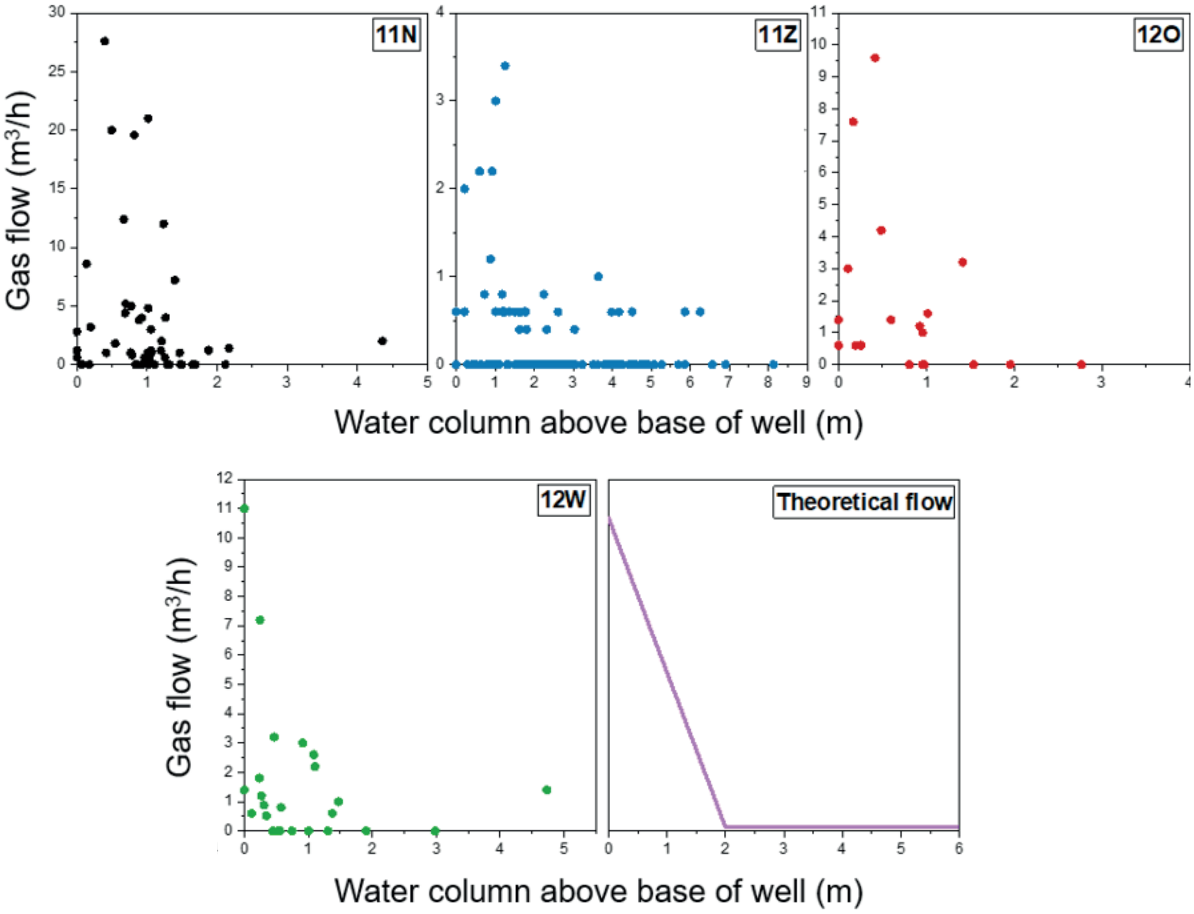

FIGURE 7: Measured normalized gas flow in relation to the height of the water table above the base of the well for compartments 11N, 11Z, 12O, and 12W, expected relationship (far right).

partments are filled with approximately the same amount of waste (11N: 300,637 m^3, 11Z: 370,110 m^3), they differ slightly with respect to waste composition: 11N contains almost 20% of waste with potential organic material in it, compared to the only 2.93% in 11Z. This waste body includes commercial waste (11N: 12.1%; 11Z: 2.3%), shredded waste (11N: 2.2%; 11Z: 0.33%), street cleansing waste (11N: 0.2%; 11Z: 0.0%), coarse household waste (11N: 0.5%; 11Z: 0.28%), sludge (11N: 0.4%; 11Z: 0.02%) and household waste (11N: 2.7%; ; 11Z: 0.0%), suggesting a higher potential for microbial degradation of waste organic matter. It is likely, however, that the difference in aeration efficiency and therefore the difference in temperature is due to the differences in water tables (Figure 7), limiting aeration efficiency in compartment 11Z.

The cumulative settlement with respect to the first measurement (2017) until 2020 is shown in Figure 10. In line with the increased extent of aeration and increased carbon extraction efficiency, compartment 11N stood out with the highest cumulative settlement of on average 0.3 m in the period 2017-2020. Also, the range of cumulative settlement in 11N increased over time, indicating that individual areas are subject to higher biodegradation rates than others.

The higher carbon extraction efficiency, temperature, and cumulative settlement suggest a higher microbial activity in compartment 11N, which corroborates the better performance of the aeration system, evidenced by a higher share of CO_2 in the extracted gas compared to CH_4, higher flow rates, lower water tables and higher amount of organic waste.

4. CONCLUSIONS AND OUTLOOK

The dense network of wells installed for in-situ aeration of four compartments of the Braambergen landfill provided a unique opportunity to study the small-scale spatial variability of gas flow, composition, and water columns in the wells. So far, the following conclusions can be drawn:

Gas composition and gas flow rates are subject to high spatial variability, both within one compartment and between compartments.

Particularly in compartment 11Z, considerable perched water tables, impede gas flow and hence aeration efficiency, as also from high ratios of CH_4 to CO_2, indicating predominantly anaerobic conditions. This is consistent with the estimated low aerobic activity. However, high water columns alone can not explain the difference in flow rates. Other factors need also to be considered such as the spatial variability of the gas permeability within the waste body.

The detected short-circuiting of atmospheric air along wells will limit the efficient aeration of the zone of influence of the respective well.

The highest difference between the measured cumulative C extraction and the predicted cumulative C generation (by landfill gas modeling) between 2017 and 2020 was found in compartment 11N, with more than double the extracted carbon with respect to the predicted value.

Higher aeration efficiency (11N) enables higher organic matter degradation, evidenced by higher carbon extraction efficiency, higher temperature, and higher cumulative settlement. Future investigations will include liquid and gas tracer tests to analyze the spatial variability of permeability within the waste package and the sphere of influence of the aeration wells. Further, stable isotope probing of the gas in the aeration wells will be carried out to identify wells influenced by processes of CH_4 oxidation, respiration, anaerobic landfill gas production, and dilution.

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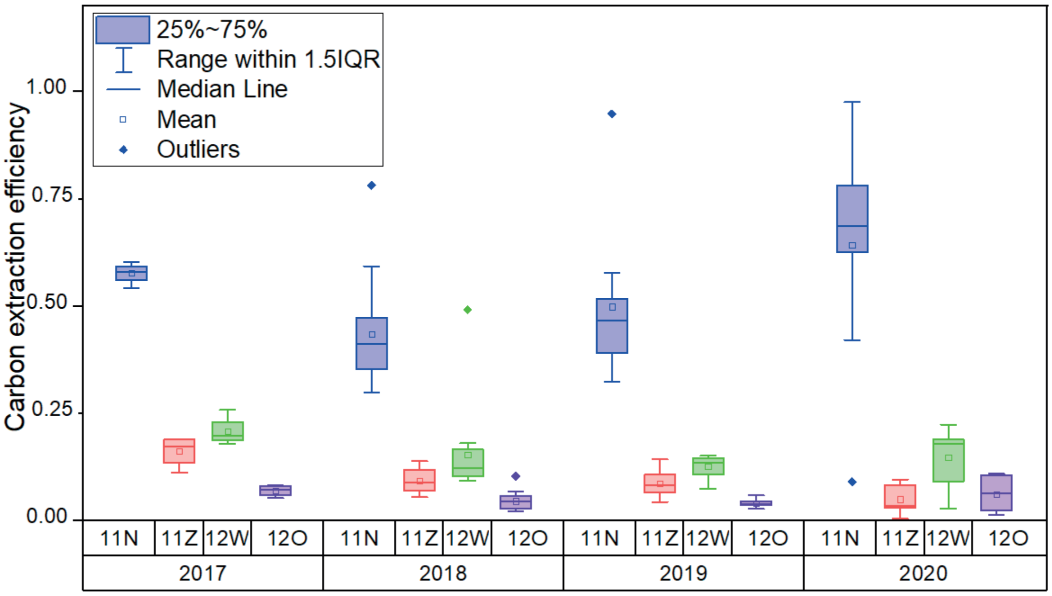

FIGURE 8: Carbon extraction efficiency for compartments 11N, 11Z, 12O, and 12W over the years 2017- 2020.

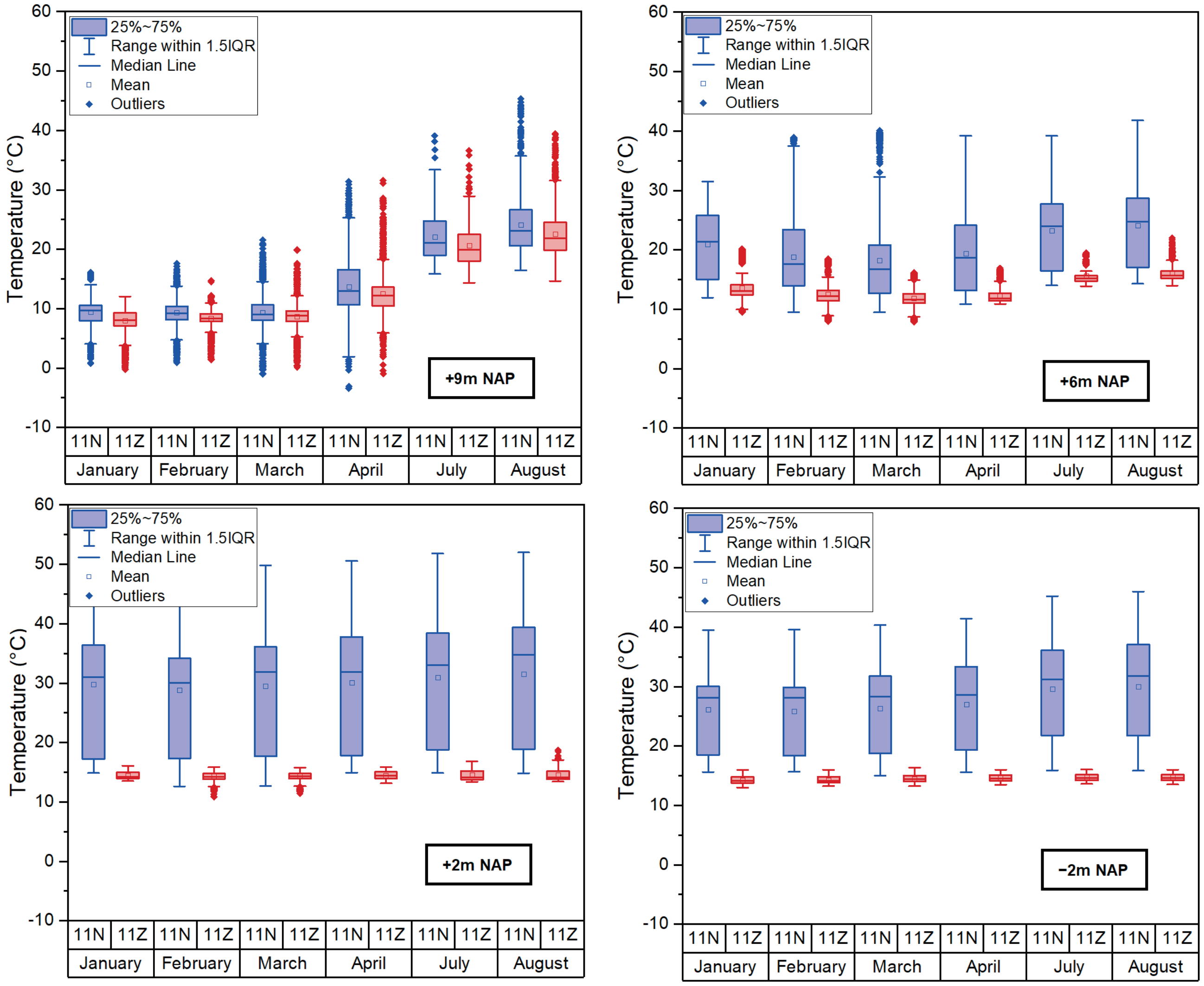

FIGURE 9: Temporal variability in temperature at +9m NAP (top left), +6m NAP (top right), +2m NAP (down left), -2m NAP (down right) – January-August 2020.

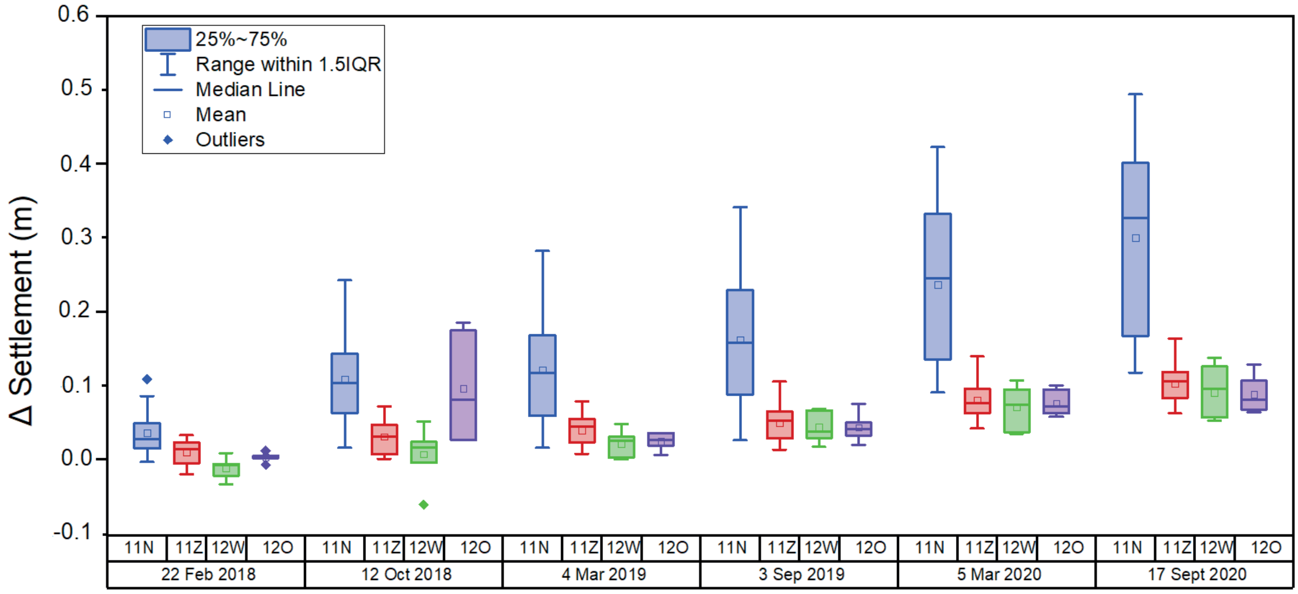

FIGURE 10: Cumulative settlement for compartments 11N, 11Z, 12O, and 12W over the years 2017 to 2020.

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SPATIAL VARIABILITY OF LEACHATE TABLES, LEACHATE COMPOSITION AND HYDRAULIC CONDUCTIVITY IN A LANDFILL STABILIZED BY IN-SITU AERATION

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ABSTRACT

Within the framework of the Dutch sustainable landfill project iDS, four compartments of the Dutch landfill Braambergen have been treated by in-situ aeration since 2017. The aeration infrastructure comprises 230 wells with a spacing of 15 to 20 m, distributed over an area of around 10 ha, intercepting a waste body of 1.2 × 10^6 t of contaminated soils, soil treatment residues, bottom ashes and construction and demolition waste. The wells, used in an alternating fashion for air injection and gas extraction, can also be used to monitor water tables within the waste body. In order to describe the spatial variability of waste hydraulics, design a larger scale leachate pumping test and, eventually, support model predictions of the site's water balance and emission potential, analyses of leachate composition and pumping tests on individual wells have been conducted. The spatial variability of leachate quality and water tables is very high with no geospatial relationship between the sampling points. Each sampling point is representative of itself only. Large differences prevail not only between and across the compartments, but also between directly neighbouring wells. Both the small scale differences in leachate tables as well as in leachate quality indicate a spatial pattern of zones with low horizontal connectivity within the waste body. Recovery rates of drawdown in the wells yielded preliminary estimates of horizontal waste hydraulic conductivity in the order of $1×10^{-7}$ to $6×10^{-4}$ m/s.

1. INTRODUCTION

A novel approach to landfill after-care has seen a focus on the accelerated stabilization of wastes under controlled conditions within in the time frame of current liability using in-situ techniques such as aeration and/or (re-)infiltration of water or leachate. Both techniques aim to decrease a landfill's emission potential by accelerating the decay of reactive organic matter to expedite the attainment of stable conditions in the waste (Kosson et al., 2002; Scharff et al., 2011; van der Sloot et al. 2017) and environmentally acceptable residual emissions (Brandstätter et al., 2015a,b, Heyer et al., 2005; Oonk et al., 2013; Reinhart et al., 1996, 2002; Ritzkowkski et al., 2006). These are reached when the water leaving the landfill complies with soil and groundwater quality criteria and can therefore be safely received by soil, ground- and surface waters. Heterogeneity and anisotropy of waste properties challenge the desired homogeneity of aeration (Ritzkowksi & Stegmann, 2012). The extent of the (preferably homogeneous) spatial outreach of air flow depends on well spacing, aeration/extraction pressure, waste permeability (Xu et al., 2020) and the amount of water in the waste, obstructing air and gas flow (Hrad et al., 2013). The typical, construction-related layered structure in waste bodies, the differential response of the heterogeneous waste inventory to vertical overburden, impermeable waste constituents such as plastic sheets, and entrapped landfill gas (Woodman et al., 2014) can lead to the development of perched water tables and preferential flow paths for water and gas. In such a system, significant volumes of the contaminated material are isolated from water flow, so that transport of contaminants can only take place through (slow) diffusion rather than (fast) advection (Bendz et al., 1998; Rosqvist & Bendz, 1999; Rosqvist et al., 2005). Preferential flow paths therefore pose a major rate-limiting constraint on both in-situ treatment and emissions.

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Within the framework of the Dutch sustainable landfill project iDS (https://duurzaamstortbeheer.nl), two landfills have been treated by in-situ aeration since 2017 (Lammen et al., 2019, 2021; Cruz Osorio et al., 2021). In some of the four aerated compartments of the landfill Braambergen, high leachate tables are observed. It is hypothesized that these are responsible for reducing the efficacy of in-situ stabilization. The aeration infrastructure can also be used to monitor and sample leachate tables and provided a unique opportunity to study the variability in hydraulic behavior and leachate properties within the waste body. In order to design a larger scale leachate pumping test and, eventually, support model predictions of the site's water balance and emission potential, analyses of leachate levels, composition and pumping tests on individual wells were conducted.

2. DESCRIPTION OF SITE AND AERATION SYSTEM

Braambergen landfill is located near the city of Almere in the northern part of the Netherlands, with four pilot compartments 11N/Z and 12O/W in operation from 1999 to 2008 with a surface area of approximately 10 ha. The pilot compartments contain around 1,200,000 tons of waste, mainly composed of contaminated soils, soil residues, bottom ashes, and construction and demolition waste (Lammen et al., 2019) and are covered with at least one meter of soil of varying origin. In-situ aeration is carried out since September 2017. A network of 230 wells spaced at 15 to 20 m distance over the four compartments can be operated in an over-extraction (until October 2019) or low-pressure aeration mode (combi-aeration, since November 2019) (Figure 1). All wells are deep filtered with the filter screen over a height of 1.8 m from the bottom of the well, which have been inserted to a total depth of approximately 2 m below Amsterdam ordnance datum (NAP, Normal Amsterdams Peil) into the waste body, equivalent to a depth of 10 to 12 m from the surface, depending on the specific location. The total extracted flow from the four compartments since three months after the beginning of aeration in 2017 varies between 200 and 400 Nm^3/h, with CH_4 concentrations between 5 and 15% and CO_2 concentrations between 15 and 22% (Cruz-Osorio et al., 2021).

3. METHODS

3.1 Leachate tables and leachate composition

Water levels in the 230 wells of the complete aeration/injection network of all four compartments were measured in March 2020 with a dip meter and, in the 132 wells of compartment 11Z, additionally with CTD Divers (Model DI271) in combination with a Baro Diver (Model DI500, both van Essen instruments). The CTD divers also delivered data on electrical conductivity (EC) of the leachate. In compartment 11Z (132 wells), leachate was also sampled for standard parameters in December 2020 using manually operated inertial pumps (Type SKU, In-Situ). These included pH, EC, redox potential, concentration of NH_4^+ (LCK303 ammonia cuvette test kit, Hach-Lange DR6000 UV-VIS spectrophotometer), dissolved organic carbon (DOC), measured as absorbance at 254 nm on a Hach-Lange DR6000 UV-VIS spectrophotometer and translation to DOC using a calibration function established for landfill leachate.

3.2 Pumping tests

Pumping tests were carried out in compartment 11Z using inertial pumps in combination with the CTD and Baro

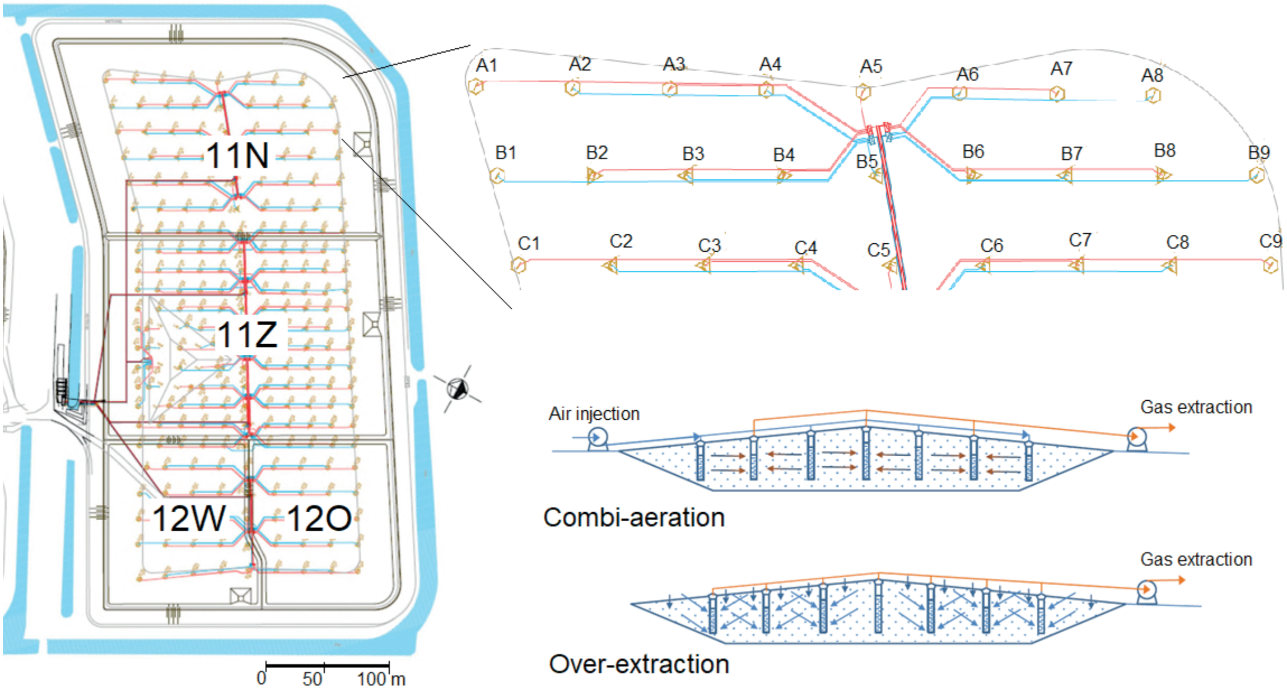

FIGURE 1: Network of aeration wells, systematics of well labeling and a schematic representation of combi-aeration and over-extraction.

divers, where the water head in the well was lowered and the recovery of the head was monitored using high-frequency logging by the CTD divers (Figure 2). Some of the 132 wells were too dry or broken and could not be investigated. In addition to the rate at which the water level recovers, indicative of the permeability of the surrounding waste, the electrical conductivity data informs whether the water present in the well at the start corresponds to the leachate in the waste body or is, for example, influenced by 'short-circuited' precipitation, reaching the wells through preferential flow channels less or not affected by contact with the waste. Equation 1 was used to estimate the volume of water influenced by one well:

$$V = \frac{Q \cdot t_{III}}{WC_V - WC_{II}} \cdot (WC_{II} + d) \tag{1}$$

where V is the total volume of the water body [L^3], Q is the pumping rate [L^3T^{-1}], t_{III} is the duration of phase III [T, Figure 2], WC_{II} is the water column during phase II [L], WC_V is the water column in phase V [L] and d the distance between the bottom of the well and the bottom of the water body.

As the distance d is not known, the radius of the water body was chosen to describe the water body that surrounds the well rather than volume. If it is assumed that the water body has a cylindrical shape, then the radius of the water body is obtained by Equation 2:

$$r = \sqrt{\frac{Q \cdot t_{III}}{(WC_V - WC_{II}) \cdot \pi} \cdot \frac{1}{\varphi}} \tag{2}$$

where r is the radius of the cylindrical water body [L] and φ the porosity of the waste [-].

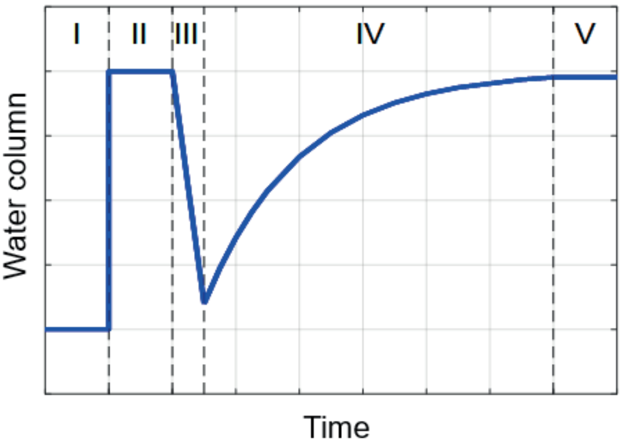

FIGURE 2: Representation of water level change during pumping test . Phase I: Diver not lowered into well yet, Phase II: Diver inserted in well, measuring full water column, Phase III: Draw-down due to pumping, Phase IV: Recovery of water level after pumping, Phase V: Recovery of water column completed.

4. RESULTS AND DISCUSSION

4.1 Variability of leachate levels and response to pumping

Significant levels of stagnant water tables were found at Braambergen landfill (Figure 3 left), with particularly high levels exceeding the height of the screened part of the aeration wells found for 83 out of 132 wells in compartment 11Z (Figure 3, right). Compartment 11Z also showed low gas flow rates, and higher ratios of CH_4 to CO_2 (Meza et al., this issue), suggesting a higher share of anaerobic processes and hence reduced aeration efficiency. Brandstätter et al. (2020) also reported impeded aeration and higher CH_4 concentrations after water-blockage of aeration pipes. For all compartments, the envelope of the scatter plot relating well flow rates to height of water column described the expected inverse relationship (Meza et al., this issue). However, water tables alone could not explain the variability in the detected flow rate. This was especially pronounced for compartment 11Z in which a high number of 'no flow' wells were detected.

Spatial variability of leachate levels, investigated in more detail for compartment 11Z, also varied in time (Figure 4), with differences mostly in the order of a few meters between March and November 2020, but also a change of 8 m was observed. Geospatial analysis of the leachate levels indicated that there is little or no connec-

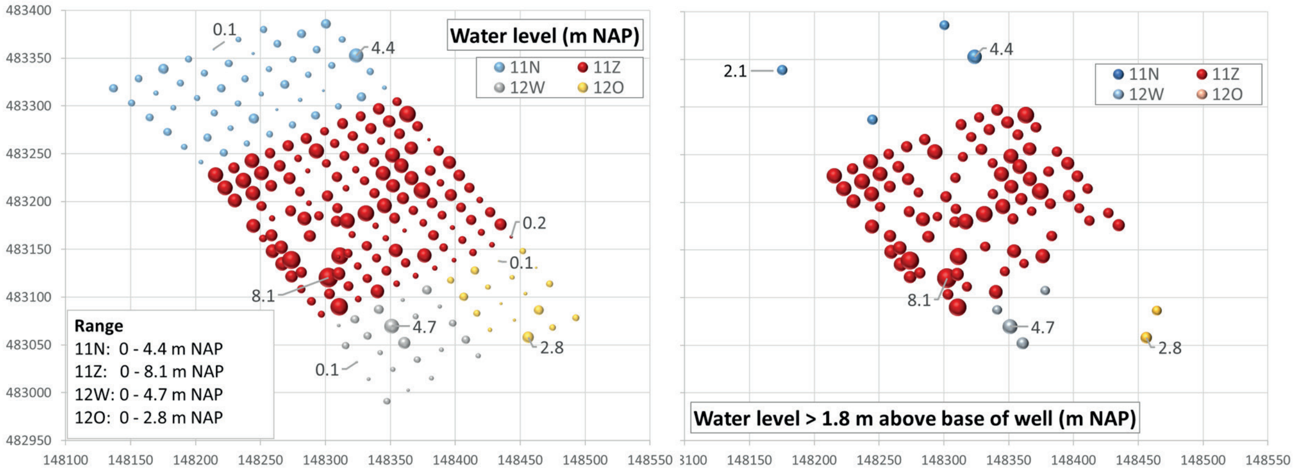

FIGURE 3: Water levels in compartments 11N, 11Z, 12O and 12W normalized to m +NAP (Normaal Amsterdams Peil, Amsterdam Ordnance Datum) March 2020 (left), levels exceeding the height of the filter screen (right). The size of the symbol is indicative of water level. Coordinates given in EPSG:28992 (Amersfoort datum).

tion between the water bodies surrounding neighbouring wells (as seen also from Figure 5 left), supported by the small apparent radii of the water body surrounding individual wells as estimated from Equation 2 (Figure 5, middle). While it has been acknowledged that the hydraulic conductivity within the waste body is heterogeneous in the vertical dimension, thus creating prefential vertical leachate pathways (Fellner & Bruner 2010), models describing leachate flow through landfills ignore heterogeneity in the horizontal plane (e.g., Slimani et al., 2017). It appears, however, that in the investigated landfill the water bodies surrounding the individual wells are stagnant zones that are hydraulically separated from each other in the horizontal plane, as a result of which differences of several meters can be sustained in stagnant zones that are less than 15 m apart. The water within the waste can be viewed as a series of disconnected ponded zones of variable vertical and horizontal permeability, overflowing when specific leachate levels are exceeded.

The hydraulic response to pumping differed significantly between wells. For 13 wells it was possible to calculate k values from the recovery of the head (Figure 5, right). Values spanned almost four orders of magnitude, 1×10^{-7} to 6×10^{-4} m/s, and are typical for the range that can be expected for silty soils to coarser sands. Given that compartment 11Z mainly harbours soils and soil treatment residues, the range appears plausible and may represent the range of horizontal hydraulic conductivities to be expected in landfill Braambergen.

Recovery behavior was grouped into the six classes (Figure 6), including a class for dry wells and for broken wells. 'Instant recovery' relates to a behavior where the recovery rate equalled the pumping rate. Wells were classified as 'no recovery' wells when the water level did not show any signs of recovery within 10 minutes after being pumped empty, intermediate behavior was classified as 'slow recovery'. In comparison to leachate properties (section 4.2), a higher level of spatial clustering could be found for the response

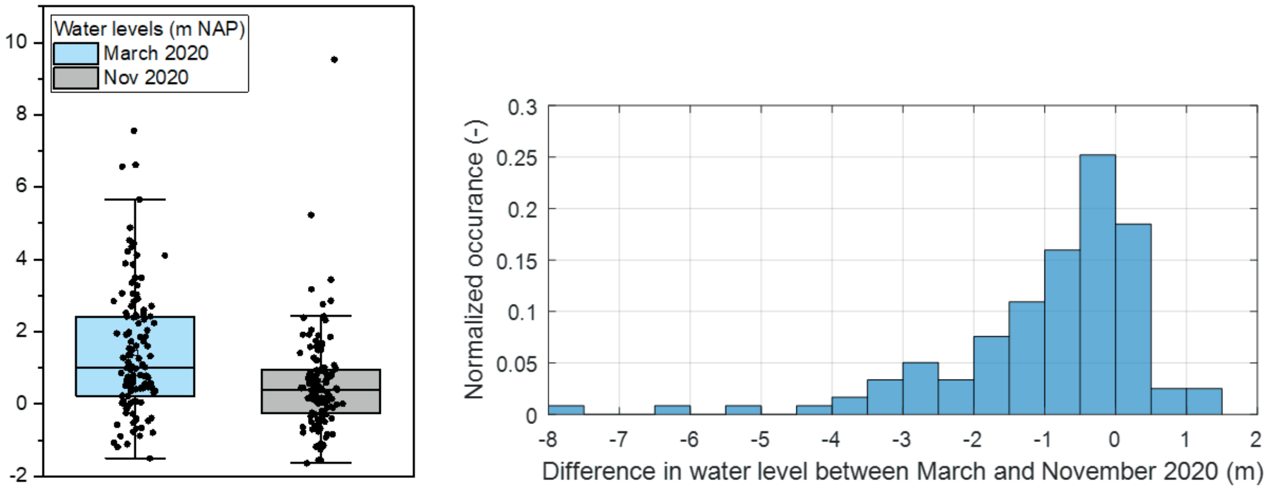

FIGURE 4: Water levels normalized to m NAP (Normaal Amsterdams Peil, Amsterdam Ordnance Datum) in compartment 11Z in March and November 2020 (left), frequency distribution of the difference between the two dates (right). Box = 25th/75th percentile, whiskers = 10th/90th percentile, line = median, closed symbols = data points.

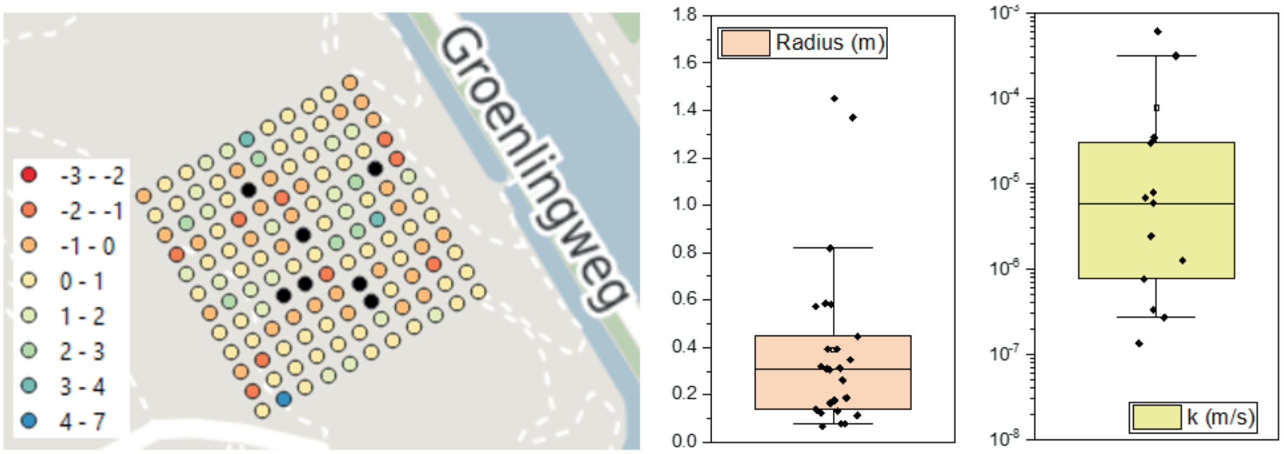

FIGURE 5: Spatial variability of water levels normalized to m NAP (Normaal Amsterdams Peil, Amsterdam Ordnance Datum) in compartment 11Z in November 2020 (left) and variability of water body radii around wells as calculated by Eq. 2 (middle) and hydraulic conductivity (right). Box = 25th/75th percentile, whiskers = 10th/90th percentile, line = median, closed symbols = data points.

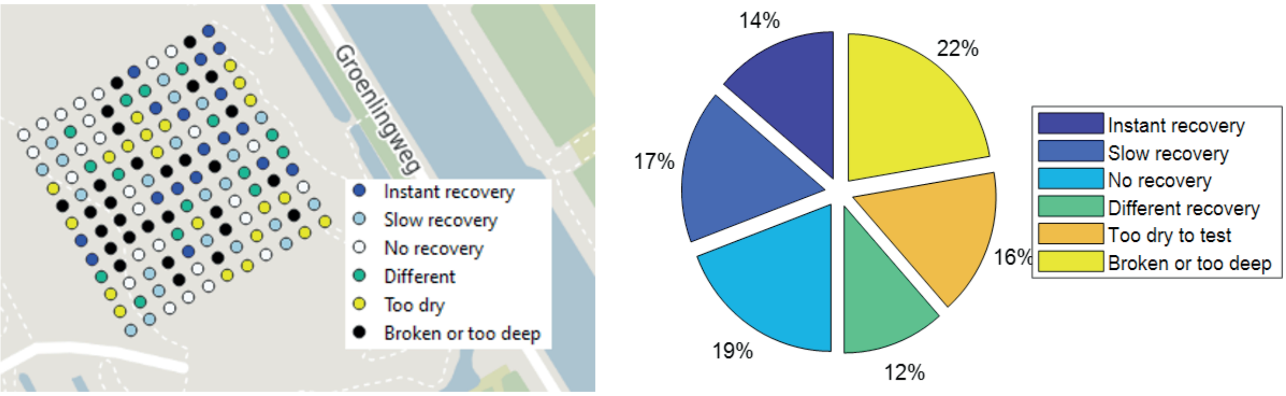

FIGURE 6: Spatial variability of response to pumping (left) in compartment 11Z, share of response classes (right).

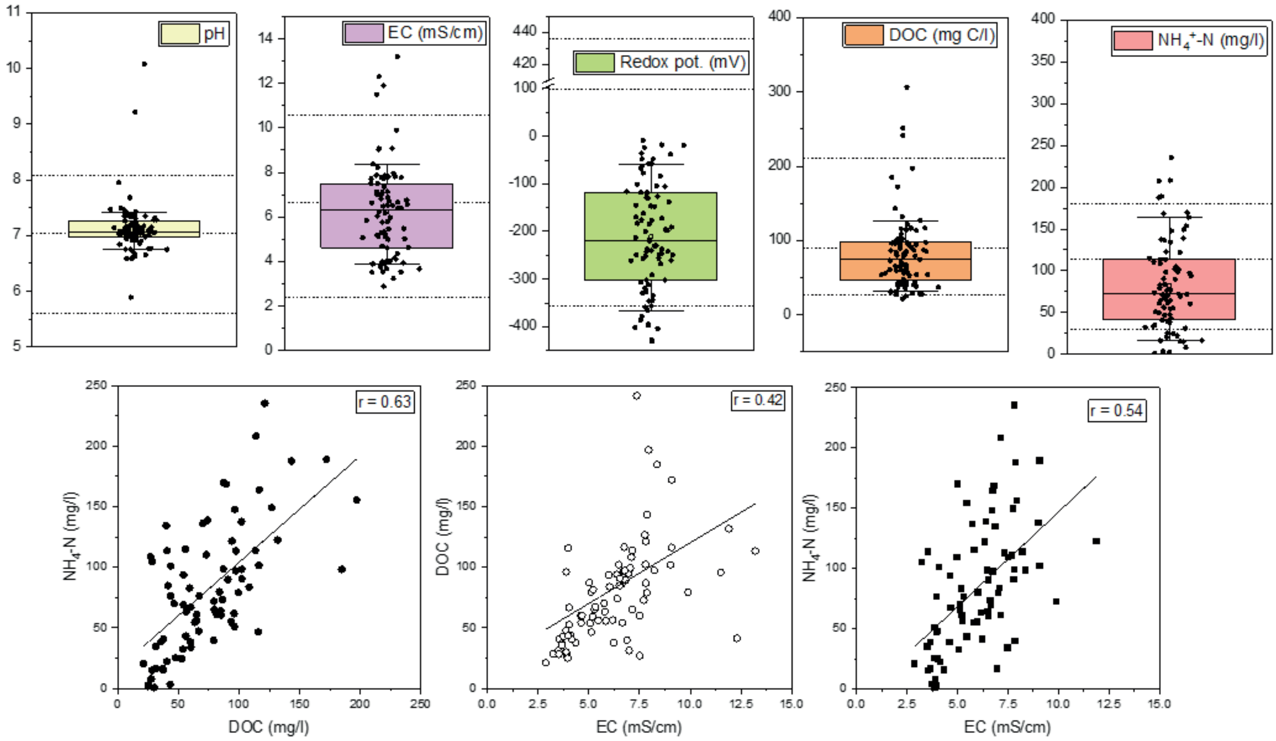

FIGURE 7: Variability of selected leachate parameters in compartment 11Z m December 2020 (top) and correlation between selected parameters (bottom). EC = electrical conductivity, DOC = dissolved organic carbon. Box = 25th/75th percentile, whiskers = 10th/90th percentile, line = median, open square = mean, closed symbols = data points. Dashed lines = maximum, minimum and average value in bulk leachate from compartment 11Z in months November, December and January from 2013-2019.

to pumping. However, also here wells showing instant recovery were directly neighbouring wells that were too dry to carry out the pumping test, corroborating the concept of discrete zones of hydraulically disconnected water bodies. All response classes were approximately equally present, with the class of ‘no recovery’ assuming the largest share of the wells where pumping tests could be carried out.

4.2 Variability of leachate quality

Leachate quality within compartment 11Z varied significantly, particularly with respect to electrical conductivity (EC), dissolved organic carbon (DOC) and ammonium (Figure 7). The variability can be caused by both the heterogeneity of the deposited waste, and the heterogeneity of waste permeability, leading to variable mixtures of original waste water and percolating precipitation. Rainwater EC is generally lower than 10 µS/cm (Zdeb et al., 2018), while EC of landfill leachate frequently ranges between 2,500 and 35,000 µS/cm (Christensen et al., 2001). The data cluster within the range that was measured in the winter months 2020-2021 in the bulk leachate, indicated by dashed lines. However, redox potential, DOC and ammonium concentrations were in the lower range of values observed for the bulk leachate, reflecting the fact that the properties of the bulk leachate are also determined by waste volumes in the unsaturated, better aerated and therefore more stabilized part of the landfill body, yielding leachate with lower ammonium and lower DOC concentrations and a lower redox potential.

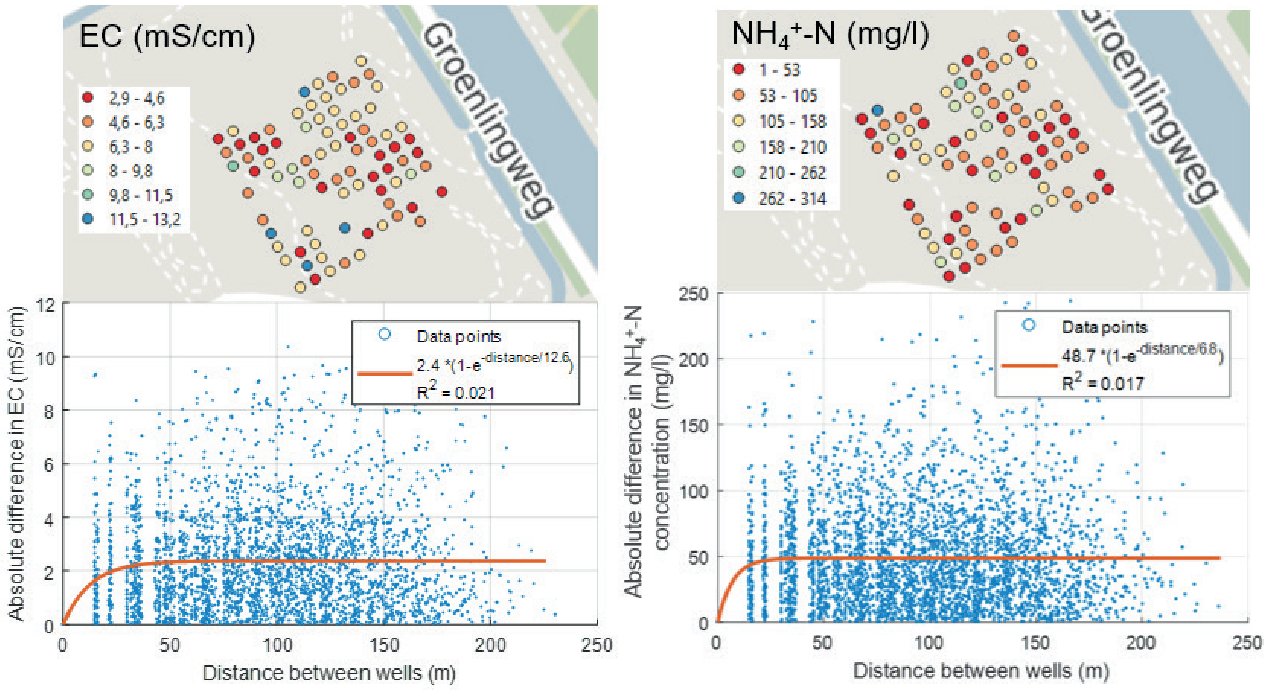

FIGURE 8: Top: Spatial variability of EC (left) and NH_4-N concentration (right), measured in aeration wells in compartment 11Z of Braambergen landfill. Bottom: Semivariogram of EC (left) and NH_4-N (right) with the concentration of each well plotted against the concentration of all other wells.

The high variability of leachate properties did not cluster spatially. Directly neighbouring wells showed large variations, as seen in Figure 8 for the concentration of solutes (EC) and ammonium. The absence of any geospatial relation of leachate properties in neighbouring wells was confirmed by a semivariogram analysis, plotting the difference in a given leachate parameter (Figure 8, lower panel). As to be expected, concentrations of dissolved organic carbon, ammonium, and electrical conductivity correlated positively (Figure 7, lower panel), highlighting the relationship between organic matter (OM) degradation, producing DOC, ammonium as the terminal nitrogeneous compound of anaerobic OM degradation and the concomitant increase in total solute concentration benchmarked by the bulk parameter of electrical conductivity. The latter is also determined by non-degradable waste components, explaining the lower level of correlation to the parameters related to biological degradation of waste organic matter. No correlation could be found for water levels or the response to pumping and leachate composition, indicating that waste hydraulics and waste biogeochemical processes.

5. CONCLUSIONS AND OUTLOOK

The aeration system on landfill Braambergen with 230 wells in four compartments and 132 wells in the focus compartment 11Z alone, at a 15 m spacing, provided a unique opportunity to study spatial variability of waste hydraulics and leachate properties. The data allow for the following conclusions:

Large hydraulic gradients persist within the waste body in compartment 11Z, which are also stable over time. This suggests zones of very low hydraulic conductivity in the horizontal plane, effectively isolating zones of ponded water from each other. Under these conditions, the spatial outreach and hence efficiency of aeration are low.

For the investigated compartment 11Z and waste bodies exhibiting similar hydraulic properties, water storage within the waste is a significant term in water balance modeling.

The well spacing of 15 m is larger than the extent of similarity of the measured properties within the waste, both with respect to hydraulic properties and leachate composition.

The radius of the saturated zone ('free' water) within the sphere of influence of each well is very small, often less than one meter. The estimates for the horizontal hydraulic conductivities found with the pumping test range from 1×10^{-7} to 6×10^{-4} m/s, however, these estimates only hold in the close proximity of the aeration wells in which they were measured.

Given the horizontal no-flow zones, pumping from one or several wells may not be effective to reduce the water table within compartment 11Z.

Solute composition and concentrations varied greatly, indicating variable extent of aeration and/or a high level of waste heterogeneity as well as the share of precipitation and original leachate in the final aqueous mixture. The properties of the bulk leachate from the entire compartment covers the range of properties found for the individual wells.

The absence of any relation between water levels or response to pumping, and leachate composition suggests that waste hydraulics and waste biogeochemistry in compartment 11Z are either not coupled or that any relation is masked by high variability in both sets of parameters.

All results combined support the concept of a 'bucket model' within the landfill, with the individual buckets demarcated by zones of low vertical and horizontal permeability. Once a bucket is full, adding more water will not increase the water level further as the bucket will overflow. The relatively low permeability zones do not have to be fully impermeable; therefore in some zones a decrease in water table is observed when the fresh accumulation of leachate is low, for example under conditions of reduced precipitation and/or high evapotranspiration from the soil cover.

Future infiltration tests as well as gas tracer tests shall further elucidate the permeability of the waste to water and gas flow and provide more insight into the wells' radius of influence. Further, it shall be investigated how DOC fractions can be used to characterize the waste organic matter, the dynamics of its degradation and the impact of anaerobic and aerobic conditions on DOC patterns.

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Extra contents

COLUMNS AND SPECIAL CONTENTS

This section comprises columns and special contents not subjected to peer-review

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Info from the global world

FROM CIRCULAR TO LINEAR SYSTEMS: HOW THE REFILLABLE GLASS BOTTLE BECAME SINGLE-USE PACKAGING

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The purpose of this note is to analyse why the refillable glass bottles were once introduced, how single use beverage packaging was phased in and why the refillable bottles were finally phased out. This note thus describes a reversed transition; from the circular to the linear. Materials have been collected from Swedish governmental reports, brewery magazines and newspaper articles.

Introducing the refillable glass bottle (1886-1954)

The first deposit-refund system was introduced in Sweden 1886 through an agreement between the Swedish brewers (Bring, 1935; 322). Previously, to buy beverages, customers typically brought their own jars to the breweries. But with the new and popular lager beer, the barrels needed to be emptied quickly, without interruption. Furthermore, the glass bottles were blown by hand, which made them expensive. Reversed logistics became the solution, where consumers merely borrowed the bottles. A deposit fee was paid at the purchase of the bottles, which was refunded when the bottles were returned. The breweries then inspected, washed and refilled the returned bottles for resale.

Establishing a deposit-refund system for glass bottles coincided with two other historical events in Sweden. The national deposit-refund system required cooperation rather than competition in the brewing industry. Thus, the deposit-refund system contributed to establishing Sweden's first business association; the Swedish Brewers' Association (Bring, 1935:217). To facilitate handling, sorting and refilling in the reversed system, all breweries were required to use the same bottles. The Brewers' Association adopted therefore in 1886 the world's first standardized glass bottle, the Stockholm bottle, a 33 centilitres bottle (Bring, 1935:322).

The refillable standardized system was so successful that it was gradually introduced for virtually all forms of beverages during the 20th century in Sweden. For example, Vin & Sprit, the Swedish company with monopoly on the production and import of strong alcoholic beverages, built a centre in 1956 for bottling wine in Stockholm. The wine was transported by boats in large containers from abroad to be bottled in Sweden in standardized refillable glass bottles.

Most popular, however, was the 33 cl Stockholm bottle that was used for beer, soft drink and carbonized water. The bottles were typically bought in trays, which were also subject to a deposit fee. The design of the standardized 33 cl bottle has not changed much since 1886 (Figure 1). Machine blowing of the bottles was introduced in the 1910s and a cap replaced the cork in 1932 (Bring, 1935: 321). All bottles were initially coloured brown to reduce the effect from the sun on the content and were to at least be recirculated 100 times.

However, the Stockholm bottle was controversial. During the 1940s, the bottle, rather than the negative social impact of alcohol, was accused of causing the bad repetition of beer (Henriksson 2019:15). The chairman of the breweries' standardization committee stated that "the brown colour and the clumsy shape [of the bottle] contributes to, consciously or unconsciously, [the beer's] declassification" (Bryggeritidskrift, 1949).

Phasing in single-use packaging (1955-1997)

To improve the beer's reputation, new types of packaging were launched. In 1955, the beer can was released on the Swedish market (Ministry of Social Affairs, 1969: 29). From viewing the refillable bottle as a problem in the brewing industry, the steel can brought the packaging to the

FIGURE 1: The development of the standardized 33 cl Stockholm bottle over 130 year. The two left bottles were blown by hand (1880s, 1901) and the three bottles on the left (1880s, 1901, 1930) were sealed by a cork. The others were blown by a machine and capped (1937, 1959, 1968, 2013). Image used with permission from Henriksson (2019).

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forefront of the breweries' marketing. For example, when one of Sweden's largest breweries, Pripps, advertised their new beer, Pripps Blå, in 1961, the brilliance of the packaging was emphasized rather than the content, as the can required "no deposit and no empty bottles to bring home" (Figure 2). The advertisement ends with "an extra tip" to emphasize the simplicity of disposable packaging for the sailing trip; "Make a hole in the bottom of the empty can and it will sink faster." (Göteborgs Kungliga Segelsällskap, 1961).

The convenience of single-use packaging was thus market as a perfect match with the emerging leisure life. To meet the increased competition from the can manufacturers, the glassworks launched the single-use glass bottle a few years later, in 1959 (Ministry of Social Affairs 1969: 29).

The share of beer sold in refillable glass bottles started thus to decrease, from 94% in 1963 to 87% in 1967 (Bryggeritidskrift, 1968). But with the increased use of disposable materials, the problem of littering in Sweden grew during the 1960s. Single use packaging became a problem that required action. However, the Social Democratic government was negative to ban disposable packaging (Ministry of Social Affairs, 1969: 143). So to meet the protests, the measures focused on the effects of the use of the single-use packaging, rather than the source of the problem, i.e. the disposable packaging.

Pripps nya 45 cl engångsförpackningar för öl är verkligen idealiska för segelturen. De tar liten plats, väger mindre, är lätta att kyla och sist men inte minst — ingen pant och inga tomflaskor med hem. Nu finns både Pripps Pilsner och Pripps Special på 45 cl glaskrus och 45 cl burk.
Ett litet extra tips, gör ett hål i botten på den tomma burken så sjunker den fortare.

FIGURE 2: A Swedish beer advertisement from 1961 that emphasizes the simplicity of the single-use can and how easily it can sink through “a hole in the bottom”. The text is partly translated in the body text above. Used with permission from Göteborgs Kungliga Segelsällskap (1961).

Hence, a ban and fines on minor littering in both nature and urban areas were introduced. The bans were combined with massive anti-littering campaigns to change people's behaviour. In 1970, the Swedish Government launched the nationwide campaign "Keep Sweden Tidy" against littering. The municipality was assigned responsibility for sanitation and began to place infrastructure in the form of large bins all over. To cover the costs, a packaging fee was introduced.

But these measures against littering had only a marginal impact. For example, the police rarely prioritized littering offenses, while littering of single-use packages, especially cans, continued to increase during the 1970s (Table 1).

When the production of cans shifted material from steel to aluminium in 1981, the centre-right government demanded breweries to introduce a deposit-refund system for cans in Sweden (Swedish Government, 1981). But unlike the glass bottles, the cans and especially its sealing, the pop tab were originally designed for single use. Refilling was thus not discussed, instead the cans were to be recycled after collection, which was promoted by the high scrap value of aluminium (Swedish Government, 1981: 9).

The first plastic bottle made of PVC, Rigello, was introduced to the Swedish market 1970. But it was not until the introduction of the PET bottle in 1983 that the plastic bottle was popularized. The possibilities of resealing made it possible to sell larger volumes per unit. In 1990, 30% of all soft drinks in Sweden were sold in PET bottles, corresponding to 100 million bottles (Swedish Government, 1990:5).

Like previously successful introductions of single-use packaging, the PET bottle was also debated, and action was required. But unlike the previous governmental assessments of disposables, the consumers were no longer in focus. Instead, the manufacturing of the PET-bottle was primarily problematized:

"The manufacture of single-use packaging of PET for beverages does not correspond to the requirements on packaging of the future or their significance for conserving energy and resources" (Swedish Government, 1990: 6).

The Social Democratic government therefore banned single use plastic for beverages in 1991 (Swedish Government, 1990). Just like the refillable glass system, PET

TABLE 1: Measurements of littering along Swedish roads between 1972 and 1975, divided into different beverage packaging (Ministry of Agriculture, 1974: 81).

		1972	1973	1974	1975
Glass	Refillables	11 %	7 %	6 %	6 %
	Single use	29 %	26 %	22 %	20 %
Cans		42 %	45 %	55 %	57 %
PVC Plastic bottles		18 %	22 %	17 %	17 %
The share of beverage in relation to all packaging litter		10,2 %	10,7 %	11,6 %	12,6%

bottles were instead to be refilled through a deposit-refund system, managed by the breweries. The sealing of the bottles with a screw cap opened up possibilities for refilling. But in order to be reusable, a standardized, thicker PET bottle was introduced (Figure 3).

The system with refillable plastic bottles were nonetheless discontinued after a few years. This since the brewers' association claimed that small breweries lacked finances to establish dishwashing facilities. In addition, PET-bottles proved less suitable for industrial reuse, compared to glass bottles. The repeated cleaning resulted in discoloring and scratching the PET-bottles. Therefore, the ban on single-use PET bottles was reformulated by the centre-right government the year after its implementation, in 1992 so that the deposit-refund systems for plastic bottles would be based on single-use and recycling, just as for the can (Swedish Government, 1992).

Phasing out the refillable glass bottles (1998-)

Although the refill system for plastic bottles failed, the political decision to introduce such a system needs to be understood against the background that the refillable glass system was still running smoothly 100 years after its introduction. In 1987, about 50% of all beverages were still sold in refillable glass bottles (Bryggeritidningen, 1988). In 1986, 250 million refillable glass bottles circulated on the Swedish market. This is to be compared with 53 million in 1937 (Henriksson, 2019: 23), when refillable glass bottles were the sole packaging for beverages. The refillable glass system was thus resilient and single-use packaging did not primarily replaced the glasses, but rather expanded the market.

However, in the end of the 1990s, the refillable glass system started to be phased out. This process was not primarily driven by consumers' purchasing priorities. Nor was it because single-use packaging was cheaper for the breweries. In fact, single-use packaging was about 20% more expensive (Friedel, 2014: 510). Instead, it was an active decision taken by the breweries, since the switch to single-use packaging opened up possibilities to increase profits and markets.

Vin & Sprit, which had a monopoly on the import and production of alcohol in Sweden, abandoned the refill system in 1998 because "single use bottles are much more profitable for us" (Svensson, 1997) according to their CEO. The trays with the standardized 33 cl. bottles are here a prime example. They had become a catchpenny for the grocery stores in the 1970s and 80s: buy food for 30 euros, and buy a tray with 20 bottles for 3 euros!

FIGURE 3: The refillable PET-bottle (Vänermuseet, 2015). Licensed under creative commons.

Hence, customers were not willing to spend money on beverages sold in refillable bottles. The refillable bottles had simply become a symbol of cheap beverages and the margins in all stages were limited. A representative of Sweden's breweries association justified the phase out of the refillable glass system by saying that "no one makes any money from them. The price for the consumer is simply too low" (Gustafsson, 2013). However, restaurants could charge higher prices for the bottles, where it is thus still possible to buy the refillable Stockholm bottle.

By filling the beverage in new packaging, for example 50 cl. bottles of plastic that were placed in refrigerators, stores were able to charge higher price per product and litre, which increased the margins. A study showed that the prices of soda increased by 100% per litre after the switch from refillable glass bottles to single-use PET-bottles (Edman, 2013).

Furthermore, the dishing process at each brewery required space. A space that was justifiable when packaging materials were expensive, but less rational when the price of glass, plastic and aluminium was marginalized compared to other production costs. The trays with the standardized bottles were also a problem for some of the grocery stores since they were heavy, bulky and exposed the products poorly. By removing the trays, more units could be placed in the same space, with improved visibility.

In addition to these practical reasons, the globalization and liberalization of the Swedish beverage market contributed also to the phase out of the refillable glass system. Or as Vin & Sprit's CEO argues, the refillable bottles "worked well during the monopoly era, but not in today's free market" (Svensson, 1997). When Sweden became a member of the European single market in 1995, Vin & Sprit lost their monopoly on import. Single-use bottles began to flow into the Swedish market from all around the world.

With increased competition, a growing supply of different beverages and as the consumers' purchasing decisions moved to the stores, the Swedish breweries' interest in using the packaging to expose their products increased. Profiling the beverages only through the labelling, which were the case for standardized bottles, were no longer sufficient.

When the Swedish Minister of the Environment, Birgitta Dahl, banned single-use plastic bottles in 1991 and tried to introduce standardized refillable PET bottles, the American soda producer, Coca Cola, refused to adapt its curvy bottles. In a letter to the Swedish Prime Minister, the head office in Atlanta described how the distinctive shape of the bottles was a central part of the company's profile. Therefore, the soda giant could not accept a standardized bottle, which could according the company affect the possibilities of supplying Sweden with the specific beverage (Broberg, 1992). The specific bottle was thus exempted from the standardization.

Conclusions

This study demonstrates that the concerns over single-use packaging have gone in circles. How the problem of disposables is understood will influence the choice of measures. For example, if littering is defined as the problem, the focus of the measures may fall on the effects of consumption. Littering can thereby be tackled through bans, fines, infrastructure, bins, campaigns and deposit-refund systems. But such measures generates also acceptance for increased production of single-use packaging. Furthermore, if only one type of single-use material is problematized, the production of disposables typically continue, but only of a different material, with new problems as a result.

In order to establish large-scale reuse systems, product standardizations seem to be central. This since standards create the necessary predictability for those who shall receive the used products to adapt the circulation processes accordingly. However, it remains to be seen how standards for circulation can be introduced when free product development, packaging design and exposure have become cornerstones of the market economy.

Finally, transitions in consumption and production systems are the result of at least two different but related processes. How the linear systems are phased in or out, and how the circular systems are phased in or out. For many decades, the refillable systems and disposable systems were running side by side in Sweden, before an active decision was made to phase out the refillable bottles.

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Environmental Forensic

THE ROLE OF THE PRECAUTIONARY PRINCIPLE IN THE AGRICULTURAL REUSE OF SEWAGE SLUDGE FROM URBAN WASTEWATER TREATMENT PLANTS

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The precautionary principle

Society is constantly striving to achieve a high enough level of protection for human health and the environment, including animal and plant health (COM, 2000). In most cases, policies making it possible to achieve this high level of protection can be determined on a satisfactory and acceptable scientific and technical basis. However, when there are reasonable causes for concern that potential hazards may directly or indirectly affect ecosystems and, at the same time, the scientific information is insufficient, inconclusive, or uncertain, the precautionary principle has been politically accepted as a risk management strategy in the EU and USA (Silva and Jenkins-Smith, 2007).

The precautionary principle is now manifesting as the founding rule of the "law of uncertain science" (Rosario, 2020): uncertainty, relativization and the intrinsic nature of scientific acquisitions, combined with the incessant (and even sometimes "out of control") evolution of technologies, has led to an increase in the instances in which the application of the precautionary principle is seen as the only solution to analyze the problem.

At the international level, the precautionary principle is recognized and enshrined in a number of legal acts, including the Rio Declaration, the Convention of Biological Diversity (CBD), and the Climate Convention (UNFCCC) (Lavrik, 2022). At the EU level, the importance of applying the precautionary principle is also emphasized to uphold a high level of environmental protection (Art. 191 of Treaty on the Functioning of the European Union, TFEU), and the principle is explicitly expressed in most of the environmental Directives. Guidance as to when, i.e., in what situations, the precautionary principle is applicable is provided by the European Commission: "Recourse to the precautionary principle presupposes that potentially dangerous effects deriving from a phenomenon, product or process have been identified, and that scientific evaluation does not allow the risk to be determined with scientific certainty" (COM, 2000).

In the current column, the precautionary principle is analyzed in relation to the very "hot topic" of the management of sewage sludge.

The precautionary principle and the environmental forensic

There is a strict connection between the precautionary principle and the environmental forensic approach. This is particularly evident for the 'Shifting the burden of proof' concept, a central component of the precautionary principle that is perhaps the most important in regard to legal redress of environmental issues and consequently for environmental forensics (Kriebel et al., 2001). Following the European Commission document, the precautionary approach put up to the producer or user "to demonstrate the nature of a danger and the level of risk of a product or process" (COM (2000) 1 final).

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Consider the case of sludge use in agriculture. The decision of an environmental agency to use sewage sludge for agricultural activities may invite objections from the neighborhood that may eventually lead to parties approaching courts for redress. Where sewage application on agricultural fields is practiced, residents in the vicinity may raise complaints of health issues that they attribute to sewage spreading. During the legal procedure, the burden of proof, which otherwise would have been with the person(s) filing the complaint, shifts to the agency that changed the status quo if the precautionary principle is applied. In other words, the legal stand of “innocent until proven guilty” changes to “guilty until proven innocent" (Van den Belt and Gremmen, 2002). This shift in the burden of proof is important for the forensic investigation carried out for the case in many respects. Apart from establishing if legal limits of pollutants are exceeded, it may also become necessary to identify and establish the uncertainties associated with facts in question for the application of the precautionary principle. Application of the principle may also imply a shift in the responsibility of investigation.

Basic principles of sewage sludge management in the EU

Sewage sludge consists of residues collected at different stages of the wastewater treatment process. It is a kind of “biological-organic cocktail” containing large amounts of organic material and nutrients, such as phosphorus and nitrogen, as well as possible residual concentrations of pollutants, including heavy metals (HMs), organic pollutants and pathogens (Fijalkowski et al., 2017).

In particular, the focus of the current column is on sewage sludge originating from the treatment of domestic or urban wastewater treatment plants (WWTPs) and from the treatment of other wastewater with a similar composition. Industrial sludge is not considered here. However, it should be noted that the WWTPs in some countries also allow the entry of a considerable proportion of industrial wastewater, which may lead to sewage sludge contaminated by high concentrations of pollutants (Collivignarelli et al., 2019b; Feng et al., 2015).

The legal nature of sewage sludge is ambiguously used in the scientific community because both the terms “byproduct” and “waste” are used. However, following Directive 2008/98/EC (WFD 2008), these are two distinct concepts: a “waste” is any substance or object, which the holder discards or intends or is required to discard (Article 3(1)); and a “byproduct” is a production residue that fulfils the specific conditions (Article 5(1)) and can be commercialized.

According to the European Waste Catalog (EWC) (CEC, 2001), sludge from urban WWTPs is identified by the code 190805 “Sludge from treatment of urban wastewater” under the subchapter 19 08 “wastes from wastewaters treatment plants not otherwise specified”.

In the EU, the use of sewage sludge as a fertilizer is separately regulated by the Sludge Directive (86/278/EEC). The Directive sets rules for the use of sewage sludge as fertilizer to prevent harm to human and environmental health by “ensuring that the nutrient needs of the plants are considered and that the quality of the soil and of the surface and ground water is not impaired.” However, the Sludge Directive is now over 30 years old and has only been updated a few times since its adoption (last, by Regulation (EU) 2019/1010).

In summary, EU regulation indicates the following principles regarding the use of sewage sludge in agriculture (see also Figure 1):

1. Compliance with quality limit values for sewage sludge. The Sludge Directive only sets limits for some HMs (Cd, Cu, Hg, Ni, Pb and Zn). Several European countries have adopted more stringent requirements and limit values for concentrations of other heavy metals (Cr, As), synthetic organic compounds (PCB, AOX, LAS, DEHP, NP/NPEPAH, PCDD/F) and microbial contaminants (pathogens such as Clostridium perfringens, Enterobacteria, Enterococci, Enterovirus, Escherichia coli, Fecal streptococci, Helminths eggs, Salmonella, Thermotolerant coliforms).
2. Compliance with quality limit values for soil where biosolids are used. In addition to restrictions regarding chemicals in biosolids, EU law sets limit values for HMs in soil to avoid long-term accumulation. In most Member States, these limit values are set lower than those required by the Sludge Directive.
3. Compliance with the maximum amount of biosolids that can be spread on land per year and per unit of surface. This quantity is not directly prescribed in the Sludge Directive, but it states that it is necessary to limit the amount of HMs added to cultivated soil; therefore, many countries have adopted specific rules for this point.
4. Compliance with specific conditions for the agricultural land in which the sludge is used. The Sludge Directive (article 7) provides restrictions regarding the spreading of biosolids on grazing and pastureland and on land on which vegetables and fruits are grown. These provisions have been transposed by Member States, which often have introduced additional requirements for land spreading (for example, restrictions in the cases of sloping land, wet land, or after heavy rain).
5. A ban on using untreated sludge. According to the Sludge Directive, sewage sludge must, as a main rule, be treated before its use in agriculture. The use of untreated sludge can, however, be allowed in the case of injection or if the sludge is worked into the soil. While most Member States have prohibited all use of untreated sludge, France, Ireland, and the UK are exceptions (Collivignarelli et al., 2019). It is also important to note that the Sludge Directive does not specify what treatment technology has to be used.

There are relevant studies that compare and analyze how different EU countries have regulated the use of biosolids on agricultural land (Collivignarelli et al., 2019a; Hudcová, et al., 2019; Mininni et al., 2015; Kelessidis and Stasinakis, 2012).

In some countries, such as Ireland, the United Kingdom and Spain, a large proportion of sewage sludge is used in

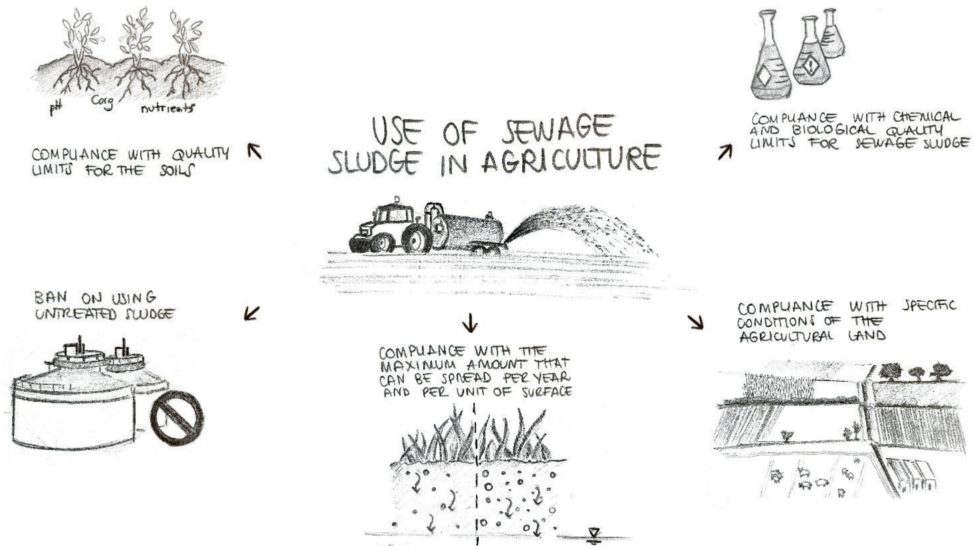

FIGURE 1: Conditions for the use of sewage sludge in agriculture.

agriculture (> 70%), while in others, such as Italy and France, it ranges between 20 and 40%, and still others, such as Germany, where less than 20% of the sludge is used in agriculture (Eurostat, 2022). Finally, there are countries (e.g., Switzerland and, more recently, the US state of Maine) in which the use of sludge in agriculture is completely banned (Collivignarelli et al., 2018; Guardian, 2022).

Although in general soil needs exogenous organic matter to maintain its functionality (and Mediterranean one need more addition of organic matter than those of central and northern Europe), it is reasonable to assume that different physical and chemical characteristics of the soil, i.e., pH, organic matter content, texture, redox potential, etc., can contribute to determining different environmental risks.

The term “biosolid” means sewage sludge that is treated for reducing human and environmental pollution risks and producing more stabilized residues. In fact, biological sewage sludge from WWTPs is usually a liquid or semi-solid liquid that typically contains 3% solids (97% liquids), whereas biosolids are typically 15-90% solids. The characteristics of biosolids vary depending on their origin and the treatment process (Collivignarelli et al., 2019b).

A wide range of sludge treatment technologies is used in the EU Member States (MS), defining its further “indirect” or “direct” agricultural use. The most common stabilization method intended for the “indirect” use of sewage sludge in agriculture is anaerobic and aerobic digestion, pursuing the recovery operation R3 “Recycling/reclamation of organic substances which are not used as solvents (including composting and other biological transformation processes)” (WFD, 2008). Sewage sludge is commonly biologically treated with other biowastes, and additional requirements may be set, including the maximum amount of sewage sludge in the cocomposting feedstock.

Methods such as mechanical sludge dewatering, drying beds, and thermal drying are widely implemented in many countries and can be considered “light” technologies to treat sewage sludge before its “direct” use in agriculture, in compliance with “R10 Land treatment resulting in benefit to agriculture or ecological improvement” (WFD, 2008).

Further treatment methods, such as lime stabilization, chemical treatment, and chemical disinfection processes (i.e., ozonation and Fenton oxidation), thermal treatment and innovative treatments are outside of the scope of this column.

Strategies for the reuse of sewage sludge in agriculture

There is a strong heterogeneity in the application of the abovementioned rules regarding sewage sludge use in EU Member States, which indicates a difference in the interpretation of the precautionary principle. It is important to note that different interpretations of this principle do not necessarily imply a lesser or greater risk for the environment. For example, with neutral-basic and/or calcareous soils, more “permissive” concentrations of some metals can be adopted when compared with acid soils without increasing the risk. Increasing or more in-depth knowledge of the specific problem thus calls for different applications of the precautionary principle.

In regard to the use of sewage sludge on agricultural land, two rather extreme alternatives concerning the role of the precautionary principle can be discerned:

a) A complete ban of all spreading of sludge in agriculture, combined with a demand for recycling in the form of material recycling where the phosphorus is extracted from the sludge. Very small facilities or sludge with low phosphorous content may be exempted from the recycling requirement. Therefore, under this alternative, the possibility of recovering sewage sludge after cocomposting and/or codigestion with biowaste (biological treatments) is banned.
b) The possibility of spreading or/and injecting sewage sludge, which meets the previously mentioned legal requirements, on agricultural land.

Both these alternatives are founded on the precautionary principle but in different ways. With regard to alternative (a), the risk of serious health and environmental consequences as a result of the spread and/or injection of sludge is not considered controllable by limit values or quality demands, and the ban is based on an real proven risk. Under alternative (b), the role of the precautionary principle is instead to direct decisions or exceptions based on quality demands.

It is important that the scientific and technical community improve the knowledge on this topic without being a sort of "supporter of a soccer team" for one or another strategy.

Following the European Commission Guidelines, the precautionary principle measures should be proportional to the desired level of protection; nondiscriminatory and consistent with similar measures taken in similar situations; based on an examination of the potential benefits and costs of action or lack of action (including where appropriate and feasible, an economic cost/benefit analysis); subject to review, in light of new scientific data; capable of assigning responsibility for producing the scientific evidence necessary for a more comprehensive risk assessment (COM, 2000). Risk can rarely be reduced to zero, but incomplete risk assessments may greatly reduce the range of options available to risk managers (COM, 2000). With this in mind, the proportionality test is highly relevant. All European Constitutional Courts have developed expertise in such a test (Butti 2007; Lang 2020; Butti and Toniolo 2018).

Any evaluation should start with as complete as possible scientific analysis of the evidence and, where possible, with a comparison of the different points of view. In Table 1, technical and scientific evidence supporting options "a" and "b" are listed.

TABLE 1: Technical and scientific evidences supporting options "a" (a complete ban of all sewage sludge in agriculture) and "b" (the possibility of spreading or/and injecting of sewage sludge).

Technical and scientific evidences supporting option "a"	Technical and scientific evidences supporting option "b"
Legal aspects	
The requirements and limits established in the Sewage Sludge Directive (86/278/EEC) are based on an old toxicological body of knowledge, i.e., not updated to the most recent research outcomes, with particular reference to the so-called Emerging Contaminants (e.g., perfluorinated compounds, polycyclic aromatic hydrocarbons -PAHs-, personal care products, pharmaceuticals, antibiotic resistance genes, microplastics, pathogens, etc.). In fact conventional WWTPs are not designed to remove these emerging contaminants that can be present in wastewater effluents (for example the antibiotics range from ng/L to µg/L) and accumulate in sewage sludge and biosolids (Silva et al., 2021; Grenni, 2022).	The main aim of the Sludge Directive (86/278/EEC) is to regulate the use of sewage sludge in agriculture preventing harmful effects while encouraging its correct use (Art. 1). Therefore, a complete ban of the use of sewage sludge would not meet the aims of the EU law framework, at least as formulated in the Sewage Sludge Directive. The current challenge should be the improvement of the overall quality of sewage sludge by means of several measures: improving the quality of wastewater optimizing the dedicated infrastructures (i.e., differentiated sewage systems), improving the efficiency of treatment systems of sewage sludge; promoting environmental education among the population to minimize the release of specific substances to the sewage system (e.g., pharmaceuticals, etc.)
Sewage sludges could contain a "cocktail" of many regulated and still not regulated substances. In this context, operators shall be aware of possible environmental crimes related with their agricultural reuse. This can be exacerbated by the continuous technological development of analytical tools capable of monitoring an increasing number of substances, also at very low concentrations.	
The general compliance with concentration limits is based on a limited set of parameters (i.e., HMs). However, the portion of sludges suitable to agricultural reuse decreases when more restrictive limits are imposed or new substances are included in the checklist for land spreading (e.g., by national or regional regulations).	Chemical characterization of sewage sludge is generally compliant with legal limit values established for the agricultural reuse.
Circularity	
For a better inclusion into the most updated regulation framework of circular economy, the implementation of the end-of-waste procedure for sewage sludge should be assured, including criteria derived from the expertise of the agronomic sector. However, the discussion by the European Joint Research Center (JRC) in Seville regarding the application of end-of-waste criteria for human organic residues (compost, digestate, biosolids) has produced a final document (IPTS 2014) where sludge was excluded from the organic wastes admitted for producing an end-of-waste.	The banned use of sewage sludge is not in agreement of the so-called "circular economy" action plan. Because the Circular Economy Action Plan (2020) requires the commission to consider revising the Sewage Sludge Directive, EU lunched in 2021 a public consultation on Sewage sludge use in farming (European Commission, 2021.).
The current prescriptions established for agricultural reuse does not just requires compliance control of the treated substrates. The current regulation includes further requirements expressed in terms of maximum amount of sludge applied and specific conditions for lands. This regulatory framework could hinder the large-scale agricultural use of sewage sludge.	
Supply of organic matter	
Soil improvers derived from biowaste and agricultural residues (i.e., compost and digestates without addition of sewage sludge) should be preferred to sewage sludge in terms of meeting the demand of organic matter to agricultural soils. Moreover, biochars from thermal treatment can be considered as candidate for this purpose; however, further research shall be performed to avoid the occurrence of contaminants such asPAH. Regarding HMs, quite strong evidence support biochar as a mean of HMs immobilisation in contaminated soils (Arabi et al., 2021), reducing HMs plant-uptake and related risks (El Naggar et al., 2021, 2022).	Direct or indirect agricultural use of sewage sludge represents a very convenient and sustainable solution to fulfil the supply of organic matter required by agricultural soil. The application of biosolids has been identified as a promising strategy to increase C sequestration in soils, directly by increasing Soil Organic Carbon (SOC) from their residual C, and indirectly by improving soil health and thereby increasing biomass production (Wijesekara et al., 2021).

Technical and scientific evidences supporting option “a”	Technical and scientific evidences supporting option “b”
Supply of nutrients	
Recovery of nutrients from sewage sludge can be achieved without their direct or indirect land reuse. Several conventional and innovative technologies of nutrients recovery from sewage sludge are available in the market or under development (e.g., Struvite crystallization technologies, P-recovery technologies from the ashes and thermal oxidation materials) (Gianico et al., 2021; Kirchmann et al., 2017)	Direct or indirect agricultural use of sewage sludge is the primary solution to fulfil the supply of nutrients required by agricultural soil, especially nitrogen and phosphorus.
Potential accumulation of pollutant in the soil	
In addition to general compliance with regulation limits, the agricultural application of sewage sludge could lead to a potential long-term accumulation of toxic elements in the soil ecosystem. In particular, occurred accumulation of HMs in sewage sludge amended soil has already been thoroughly reported by scientific literature (Charlton A., et al., 2016a, 2016b, Black A., et al., 2011). Further evidence was recently discussed about the accumulation of emerging contaminants, mainly related with pharmaceutical active products, such as antibiotics and nonsteroidal anti-inflammatory drugs, nanomaterials and microplastics (Grenni et al., 2022; Buta et al., 2021; Collivignarelli et al., 2021; Sorinolu et al., 2021; Fu et al., 2016; Kim et al., 2017; Wang et al., 2018. Yaseen et al., 2022). Residual concentrations of antibiotics in soils receiving biosolids may contribute to the antibiotic resistant pathogen abundance (Zhang et al., 2022; Kaviani Rad et al., 2022). In fact, the residual antibiotic concentrations in soil receiving biosolids can exceed the ecotoxicity effect trigger value (100 μg/kg; Yang et al., 2018).	Regarding the problem of potential accumulation of pollutants in the soil, compliance to regulation limits guarantees a fair level of safety. Lowering the current limits or broadening the list of regulated compounds could determine unsustainable characterization and managements costs.
Potential transfer of pollutants from soil to plants	
Land application of sewage sludge could lead to contaminants uptake by crop plants cultivated on amended soils. Thus far, experimental evidence of occurred transfer to plants from sludge amended soils are already available in the scientific literature, regarding both regulated (as HMs) and nonregulated substances (Buta et al., 2021, Sorinolu et al., 2021). In particular, antibiotics and their related resistance genes may enter the human food chain through the consumption of harvested vegetables (Sorinolu et al., 2021). Whether the absorbed quantities could determine a risk for human health (e.g., through the “contamination” of the food chain), it should still be further investigated (Madikizela et al., 2022; Wang et al., 2022).	Regarding the problem of the potential transfer of pollutants from soil to plants, compliance with regulation limits guarantees a fair level of safety. Lowering the current limits or broadening the list of regulated compounds could determine unsustainable characterization and managements costs.
Time quality trend of sewage sludge	
Some exceptions to the general reduction over time of the quality of produced sewage sludge can be found for specific pollutants: • In the study of Liu et al. (2021), the mean Cu concentration has remained relatively static and contemporary concentrations are similar to those observed in the early 1990s. • In the study of Olofsson et al. (2012) some emerging contaminants in sewage sludge such as alternative flame retardants showed an increasing trend; this could be due to nonefficient environmental policies to reduce the emission of these compound in the environment.	The quality of produced sewage sludge has generally improved over time (Kirchmann et al., 2017). • Liu et al. (2021) reported data from 75 wastewater treatment plants (WWTPs) in the United Kingdom, for the period 1989–2017. Only inorganic compounds (heavy metals mostly) were statically analysed: Zn, Cu, Ni, Pb, Cd, Hg, Cr, Mo, As, Se and F. The study showed that trace element concentrations decreased significantly in response to declining pollutant emissions, demonstrating the environmental benefits of effective source control and cleaner technologies. • Olofsson et al. (2012) reported data for the period 2004-2010 in Sweden. Metals, POPs, pharmaceuticals and personal care products (PPCPs), and other organic compounds in sludge were analysed. The study indicated that many of the sludge contaminants (75% of the contaminants for which statistically significant trends were found) followed a decreasing trend due to regulatory actions. • Kirchmann et al. (2017) reported data for the period 1970-2010 in Sweden. Only metals were analysed: Ag, Cd, Hg,Pb, Cu, Zn. The study showed that the quality of sewage in terms of heavy metal content has greatly improved over the past 20 years.
Land application techniques	
The current regulatory framework does not provide sufficient requirements for the modality of land application (spreading, spraying, injection, incorporation, etc.)	Relevant negative impacts (odors, ammonia emission, etc.) due to unsuitable spreading techniques (ex. splash-plate spreading) can be easily minimized by available agronomic best practices (i.e., soil injection).
The role of thermal treatments and landfills	
The management of sewage sludge should not be achieved through agricultural reuse. To do so, the capacity of the thermal treatment sector shall be increased exploiting both conventional and innovative processes. In any case, landfill disposal shall be guaranteed for residual sludges that cannot be treated alternatively.	Considering the ongoing minimization of landfill disposal (established by regulations) and the current under capacity of the thermal treatment sector, agricultural reuse is fundamental to achieve the efficient management of sewage sludge.
Competitive treated organic residues (compost, digestate, biosolids, etc.)	
Composting plants can provide a more rigorous quality control of input feedstocks than what can be achieved by a wastewater treatment plant. Moreover, the law framework related to compost production and use is more up-to-date and generally stricter (e.g., in terms of lower concentration limits of contaminants).	In the past years, composts from biowaste and agricultural residues were deemed by the agricultural sector as not suitable or convenient soil improvers. Conversely, composts are currently considered as safe, sustainable and high-quality amendments for agricultural use.

Technical and scientific evidences supporting option “a”	Technical and scientific evidences supporting option “b”
Currently, the quantitative value expressing the real amount of organic carbon needed by agricultural soil is generally not addressed by waste management plans. This information is very important to support decisions on the agriculture reuse of different treated organic residues (compost, digestate, biosolids, etc.).	The amount of produced digestates and composts is not enough to cover the soil requirements for organic carbon in most of the area.
The “zero risk” concept	
The search for a high level of health and safety and environmental and consumer protection belongs in the framework of the single market, which is a cornerstone of the Community.	Zero risk for the agricultural reuse of human organic residues (compost, digestate, biosolids) in reality cannot be achievable even if the residues are treated. Moreover, the reuse with zero risk is unfeasible with sustainable costs.
LCA studies	
Several authors used the LCA methodology (and its limits) to show that it is preferable to avoid use of sewage sludge in agriculture (Harder et al., 2016; ten Hoeve, 2018; Siegert et al., 2020; Teodosiu et al., 2016; Heimersson et al., 2016; Avadi, 2020). The main raisons to support this point-of-view are related to potential effects of the nutrients (nitrogen and phosphorous compounds) on environmental categories (as climate change, abiotic resources depletion and freshwater eutrophication), pollutants responsible for toxicity impacts (such as heavy metals), and the damage associated with specific pharmaceuticals (as hormones and anticancer drugs). Other motivations to support this perspective are the uncertainty of environmental effects related to long-term damages in ecosystem, the behavior of metabolites in sewage treatment plants, and the inability of LCA models to quantify the adverse effects of pathogens on human health.	Several LCA studies reported better environmental impacts for the agriculture use of treated sewage sludge respect chemical fertilizers (Herrera et al., 2022; Di Maria et al., 2016; Chiew et al., 2015; Tidåker et al., 2006; Suh and Rousseaux, 2002; Hareder et al., 2017; Hospido et al., 2005; Brockmann et al., 2018; Muñoz et al., 2017; Chiu et al., 2016). From these studies the combination of anaerobic digestion and agricultural land application appears the most environmentally convenient option thanks to less emissions and less consumption of energy, by avoided mineral fertilizers. Environmental criticalities related to heavy metals released seem to be negligible contribute to the impacts on human toxicity and ecotoxicity. Better wastewater management strategies can be developed combining different treatment methods. Any new risks associated with chemicals in wastewater can be minimized by improving the quality of the data with which LCA studies are performed.
Monitoring the use of sewage sludge	
A sustainable and reliable planning requires, as fundamental input data, the true amount of sewage sludge used in agriculture. However, at the moment in EU there are not harmonized criteria to evaluate this information. The EU database (Eurostat, 2022) shows a high level of missing information on this topic. Moreover, the accurate amount of sewage sludges biological treated (composting, anaerobic digestion) in agriculture is difficult to quantify because: the declaration of the type of soil improvement is not mandatory; the majority of farmers do not like to report the real application amount of sewage sludge even though they are qualified.	

Conclusions

The issue of the role of the precautionary principle in the management of sewage sludge in the European Union is giving rise to much debate, and the arguments put forward are often contradictory. In addition, the process in which the precautionary principle is applied is characterized by a need to balance the social and economic impacts of bans or restrictions, with the necessity of reducing the risk of adverse effects on the environment, including human, animal and plant health.

Based on the premise of the precautionary principle - to guide decisions in case of uncertainty - some points can be highlighted. One is the need for knowledge. With more knowledge, uncertainty decreases, and thus the need to apply the precautionary principle; in such cases, it may be sufficient to prescribe suitable precautionary measures that reduce the negative effects of the activity. More knowledge may, however, not always be achievable; a certain degree of uncertainty is likely to prevail even for less complex issues than the effects of the use of sewage sludge in agriculture. If that is the case, a recourse to the precautionary principle is an appropriate way to assess whether the individual activity, in that context, entails risks that it should not be permitted.

However, more information as a (partial) answer to the question of what role the precautionary principle should play also points to the need to both acquire and disseminate this important knowledge. This in turn requires not only research and resources but also dialog, both between different scientific areas and between science and practice.

The scientific community has yet to reach a shared position on this topic, and in the current column, two extreme alternatives have been discussed. No joint message can therefore be conveyed to authorities, farmers, or the public regarding the risks of using sewage sludge as fertilizer.

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DETRITUS & ART / A personal point of view on Environment and Art

by Rainer Stegmann

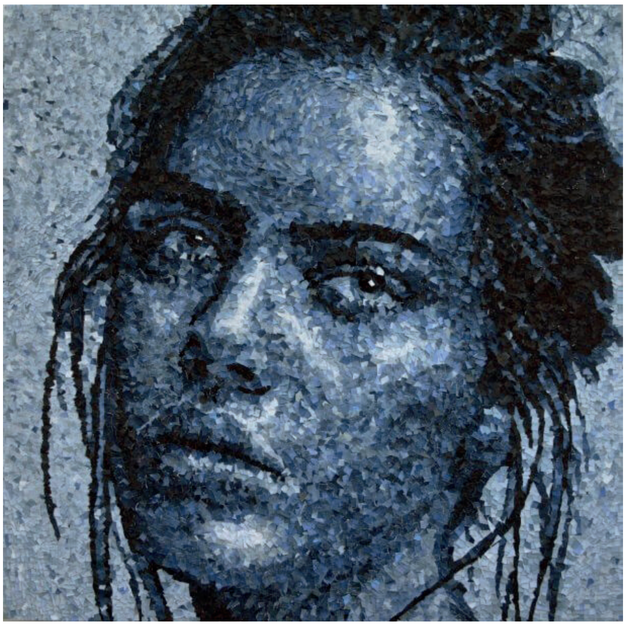
Amazing Art Mosaics from plastic pieces

Let me introduce Deniz Sagdic to you.

Deniz Sağdıç is a Turkish artist. She was born in 1982 in Mersin, Turkey, and began her art education in Mersin University's Faculty of Fine Arts in 1999 where she graduated at the top of her class in 2003. She moved to Istanbul where she established her own studio. Deniz created her own plastic style which became the artist's signature. She tried different mediums as painting, video art and photography. Deniz started a project called Ready ReMade where she uses daily use objects for creating art. Her latest material is used denim. She cuts it into small pieces and stripes and uses those as a painter uses different colours. Other materials she uses are used zips and buttons, but also used paper and cardboard. *"You can scratch the denim up like dressing a marble and can sculpt it into different shapes, without the fear of it being broken like a marble. You can transform it into 3D forms like a sculpture by stretching, bending, and folding…"* (Interview with Deniz Sagdic at fashionunited.uk)

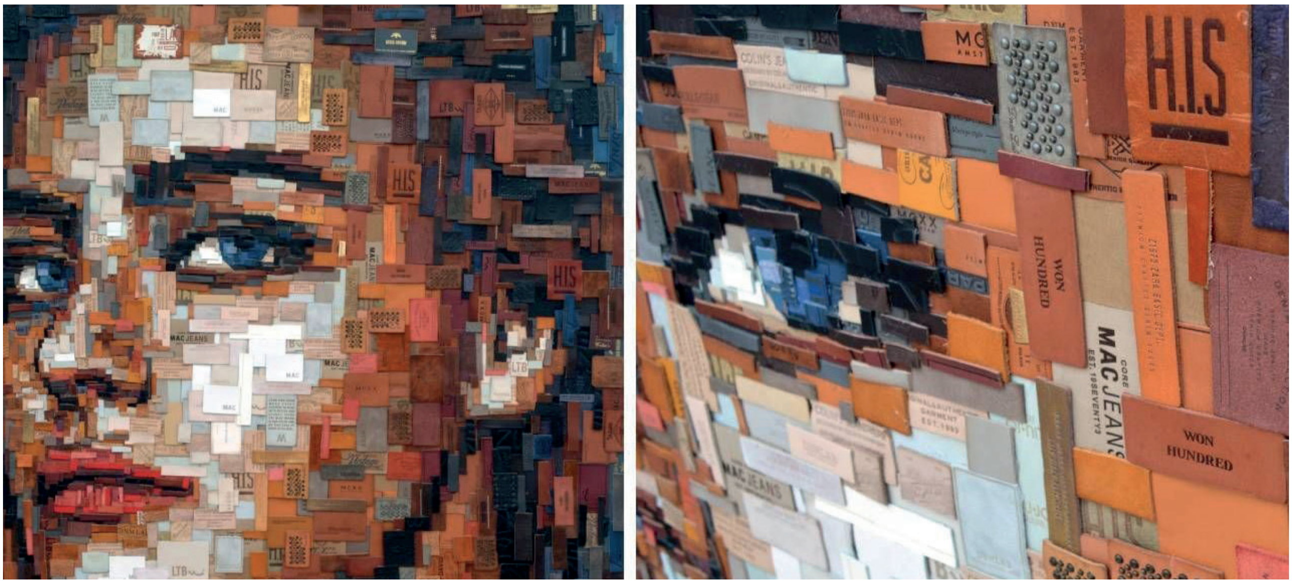
A friend made me aware of Deniz when some of her portraits were exhibited at the new Istanbul Airport. These large formatted art objects caught a lot of attention and I was immediately fascinated by her work.

Deniz gives used products a second chance showing the value of discarded clothes and brings them to new life.



This portrait spreads a sad atmosphere to me it seems she is looking for help, such a strong expression. On the left side you can see the pieces of old jeans in their different blues.

Her message is also that art can be created using all kinds of materials, there are no limits. She directs our intention



Deniz brings this face to life with a very specific expression by uing different pieces of paper and cardboard.

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to the problem of fast fashion that I presented already in Volume 18 of Detritus Journal. Although the portraits immediately cast a spell over us the aspect of the reuse of garments and fabrics needs to be highlighted.

Can art like one I present in this issue can bring people to think more of the value of their dresses and fabrics, will the reader buy less fashion? This depends on each single person watching this page, some may care more most may be not. But what in my view is important to make people aware of the thousands of tons of used dresses ending up on dumps in Africa and South America or that are burnt in MSW incinerators and may be this problem will enter their subconscious which may indirectly change peoples´ behaviour buying less and more long - lasting dresses.

I have selected some of her incredible portraits, showing for some of them the materials she used. The expression of the different faces is very concise independent of the materials used.

Deniz calls this art series “Being a Human”: “*I try to bring into view that art is human-specific but also its properties which explain human being are inseparable, one and sole. My art works arose in this context and also remove the obstacles that I mentioned, also this time try to contact with human being in its own contents. Every works purpose to deal with an argument one by one or entirely*”.

In the next edition I will show an installation of Goncalo Mabunda collects scrap metals and recycles weapons used in the 15 year long civil war in Mozambique, Africa.
From: Making Africa – A Continent of Contemporary Design, Vitra Design Museum.

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