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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

KEY FACTORS IN WASTE MANAGEMENT SYSTEMS AND THEIR VISIBILITY

Waste generation is inevitably connected to the socio-economic status of a region or country. Increasing population, rapid urbanization, increasing economic activity and an increase in society's living standards, mainly in major cities, has led to substantial growth in global waste generation. Specific socio-economic conditions prevail in developing countries, including rapid population growth, rural-urban migration, lack of funds and low-skilled cheap labour. Sustainable waste management (SWM) systems in emerging economies often lack facilities, and suffer insufficient service coverage, improper disposal and treatment methods that can lead to major environmental and social problems.

Direct adoption of developed cities' approaches without proper consideration of the local circumstances may lead to unsustainable future waste management in developing cities (Mukhtar et al., 2016). The significance of SWM factors in developing countries has been evaluated in many studies according to specific contexts including: socio-economic impacts on waste generation, recycling, waste to resource initiatives, the collection of municipal waste and disposal of waste. The roles of specific factors are not always well-defined nor their influence measured.

Multiple factors affect the development of SWM, including legislative, environment, social, technical, health, market demand and economic aspects. These key factors need to be identified and their role(s) understood to ascertain whether proposed waste management plans are duly tailored to local requirements and are viable in environmental, social and economic terms. The key factors relevant to waste management systems can vary due to the differences between individual cities' characteristics.

Some factors are usually measurable by specific indicators or scales, quantifiable, considered in decision-making and implementation processes, and publically accessible (e.g. demographic indicators): these factors may be termed "visible". In contrast, there are "invisible" factors that not usually measured or quantified but still likely to influence waste generation, behaviour and operational practices, and perceptions about waste. These factors potentially influence the need for development of a waste management system but are qualitative (e.g. behaviour, understanding and awareness) and may be important if local conditions are to be recognised and addressed in the design and implementation of waste management systems.

Various factors in SWM play different fundamental roles in waste management practices. These include: policy and strategy, age and aging communities, community behaviour and interactions, the socio-economic impacts on waste generation, recycling, waste to resource initiatives, the collection and disposal of municipal waste. The degree of influence of invisible factors is perhaps varied and sometimes unclear in terms of impact. Nevertheless, in principle they serve as a starting point to design more effective waste management strategies or policies based on tangible local trends or evidence, rather than adopting best practices from elsewhere which may not address local characteristics, customs, uniqueness or waste composition.

A comprehensive list of 43 fundamental SWM factors, derived from the literature and classified according to the PESTLE (political, environmental, social, technological, legal and economic) system. are summarised in Table 1. They were evaluated by global experts from developed and developing countries via a Delphi survey to determine whether they were considered as visible or invisible in practice.

The global experts agreed on the classification of 24 factors. Experts from developing countries classified 27 factors as visible whilst those from developed countries classified 20 factors as visible. There was agreement that 15 factors are visible: local government plan, government priorities, seasonal variations, local/national events, skilled workers and experts, application of suitable technology, facilities availability, international directives, local policies, producers responsibility, relevant SWM law, incentives from waste, availability of funds, interest and tax, and incentives. There was agreement that 8 factors are invisible: corruption, influence of politicians, environmental guidelines, environmental targets, religion, ethnicities, discrimination and philosophical change.

There was also agreement in that no clear consensus was observed for the rate of technology change. Thus, experts from developed and developing countries generally agreed on the visibility of environmental, technological, legal and economic factors, suggesting that these four categories have emerged as important. The classification of political and social factors showed less consistency between developing and developed countries; social and political factors are important but difficult to measure or assess because of their complexity.

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TABLE 1: Summary of fundamental factors that are important in the development of SWM according to developed, developing countries and their matched factors between both types of countries.

PESTLE class	Developed countries	Developing countries	Overall matched
Political	Local government plan Government priorities	Government stability Local government plan Government priorities Bureaucracy	Local government plan Government priorities
Environmental	Environmental guidelines Environmental targets Environmental awareness	Environmental guidelines Environmental targets Geographical landform Environmental awareness	Environmental guidelines Environmental targets Environmental awareness
Social	Seasonal variations Resource consumption patterns Socio-demographic indicator	Resource consumption patterns Rural-urban migration Socio-demographic indicator Seasonal variations Cultural Local national events Attitude behavior gap Resistance to change	Seasonal variations Resource consumption patterns Socio-demographic indicator
Technological	Skilled workers and experts Application to suitable technology Facilities availability Research and development	Skilled workers and experts Application to suitable technology Facilities availability Research and development Rate of technology change	Skilled workers and experts Application to suitable technology Facilities availability Research and development
Legal	International directives Local policy Producers responsibility Relevant SWM law	Local policy Producers responsibility Consumer accountability Relevant SWM law	Local policy Producers responsibility Relevant SWM law
Economic	Availability of funds Trade restrictions on waste Economic growth patterns Incentives Interest and tax	Availability of funds Trade restrictions on waste Third sector restrictions Economic growth patterns Interest and tax Incentives	Availability of funds Trade restrictions on waste Economic growth patterns Interest and tax Incentives

From the above it is clear the need to adopt new perspectives in the selection of factors considered in the development and optimisation of local waste management systems. By definition, invisible factors are not currently recognised as contributors to waste management systems, and even though they may be locally very important, they may have been neglected in decision-making processes. Recognising and making use of selected invisible factors within a local context may hasten the implementation and effectiveness of initiatives taken towards the development of SWM systems.

Factors in waste management may vary in influence and change dynamically alongside urbanization; this dynamic varies from one country to another and so factors need to be re-evaluated periodically. Different combinations of factors influence the development of SWM systems. Alongside the use of a reliable evidence-base, addressing the factors – visible and/or invisible - that strongly influence local conditions is crucial if municipalities in developed and developing countries are to move towards more effective, locally optimised sustainable waste management systems.

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REFERENCE

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EFFECTS OF SOLIDS CONCENTRATION AND SUBSTRATE TO INOCULUM RATIO ON METHANE PRODUCTION FROM FERMENTED ORGANIC MUNICIPAL SOLID WASTE

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ABSTRACT

The organic fraction of municipal solid waste (OFMSW) is usually stored under variable humidity conditions and extended periods before processing in anaerobic digestion plants. Digestate from previous batches is used as inoculum and mixed with previously stored (fermented) OFMSW for methane production under different substrate (OFMSW) to inoculum (digestate) ratios. Although both the moisture content during the storage of OFMSW and the inoculum concentration in the feed mixture to the anaerobic reactors are determining factors for the process, no studies were found on how these combined parameters affect methane production. This research is focused on determining how humidity affects OFMSW during storage (fermentation) and how the substrate to inoculum ratio affects the methanisation of the previously stored OFMSW. OFMSW with solids concentrations of 10, 20, and 28% was stored/fermented at 35°C for 15 days. The methanisation of the previously fermented OFMSW was allowed at S/I ratios of 0.5, 1.0, and 1.5. The fermentation results show that ethanol and acetic acid accounted for 90% of all products (metabolites). The lowest solids concentration allowed the highest fermentation degree with the highest VFA and alcohols production. Compared to fresh OFMSW, methane from fermented OFMSW increased 32%, and the time required to reach the maximum methane production decreased between 11 and 40%. For fermented OFMSW, the S/I ratio of 1.0 is the best condition to produce methane. ANOVA shows that, independently of solid concentration during storage, the S/I ratio is the main parameter to consider for methane production and reducing reaction times.

1. INTRODUCTION

In anaerobic digestion (AD) facilities, temporary storage of the organic fraction of municipal solid waste (OFMSW) is a necessary practice due to seasonal changes in the type of wastes, collection practices, the distance of the sources to the processing facility, limited capacity of collection and transport systems, and limited processing capacity (Worrell and Aarne, 2011). Storage takes place from the moment the OFMSW is produced and placed in containers at the generation sites (houses, shops, public spaces, among others), where organic material begins to decompose. Subsequently, depending on composition, humidity, and other environmental conditions during storage, different fermentation processes could occur (Nilsson et al., 2018). Within a short time of storage, readily biodegradable substances are rapidly transformed into volatile fatty acids (VFA) and alcohols, causing their accumulation. These processes need to be controlled appropriately to avoid inhibition during the subsequent methanogenic step (Fantozzi and Buratti 2011).

To reduce the risk of process failure by acidification during anaerobic digestion, controlled storage has been proposed as pretreatment. In this stage, the conversion of readily biodegradable substances is oriented to ethanol production rather than VFA (Wu et al., 2015). Ethanolic fermentation positively affects methane production because ethanol production consumes less alkalinity than VFA fermentation (Zhao et al., 2016). The ethanol fermentation path allows the methanogenic microorganisms to use chemical energy more efficiently (Pipyn and Verstraete, 1981). As a general concept, acid, and alcoholic fermentations before methanisation enable the control of specific process parameters and adjust operational conditions for increased subsequent methane production (Schievano et al., 2014).

In European countries, where the controlled storage (si-

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lage) of agricultural residues and energy crops to produce biogas is a common practice, studies show that controlled storage (silage), as a pretreatment before anaerobic digestion, allows the improvement of methane production from solid organic substrates (Pakarinen et al., 2011; Wang, 2010; Herrmann et al., 2011). Lü et al. (2016) consider mesophilic food waste storage as pretreatment to enhance methane production: They conclude that hydrolysis and subsequent methane production improve drastically with storage time (up to 12 days). Although OFMSW storage for weeks or even months is usual in biogas plants, no studies related to fermentation processes during storage have been found. OFMSW storage before processing in biogas plants in Europe, China, and other countries where industrial biogas production is standard is mandatory as the initial step of the whole process (Stürmer, 2017; Al Seadi et al., 2008; Qian et al., 2016.

Solids concentration is a critical parameter during silage. OFMSW arrives at the processing facilities under different humidity conditions, and the microbial dynamics and reaction kinetics depend on the bioavailability of water in the environment (Zhou et al., 2018; García-Bernet et al., 2011). Storage or silage is the previous step to methanisation. The concentration of the archaea responsible for this step is crucial as they are responsible for consuming VFA and avoiding alkalinity to drop below specific limits. Higher concentrations of methanogenic microorganisms are related to process stability; they can consume the previously produced VFA without allowing the media to become acid or decrease alkalinity beyond recovery (Hosseini et al., 2019). The main objective of this research deals with the effects of OFMSW storage (silage) under different solids concentrations and the consequences of silage on subsequent methane production.

2. MATERIALS AND METHODS

2.1 OFMSW sampling and characterisation

Three times a week, source-separated OFMSW is transported to the Coyoacán transfer station in Mexico City, where sampling was made according to ASTM D5231-92 (2016). Approximately 100 kg from 11 trucks were separated and thoroughly mixed using a skid-steer loader and shovels. Then, the quartering method was applied two times to reduce the bulk to approximately 200 kg. Undesired materials, such as plastic bags, stones, and wood, were manually separated. The remaining "clean" OFMSW was distributed in two-liter freezing bags and frozen at -20°C. A sample of fresh OFMSW was characterized for pH, total solids (TS), volatile solids (VS), COD, VFA, and ammonia nitrogen (NH4-N). VFA and NH4-N were determined in the liquid fraction after centrifugation. All determinations were made according to Standard Methods (APHA, 2017).

Table 1 shows the characteristics of fresh OFMSW. OFMSW contains 72% moisture and, from the total solids, 79% corresponds to volatile solids. Fresh OFMSW is naturally acid with a pH value of 5.2 and a VFA concentration of 10 gCOD/kgVS. Determined as COD, 40% of OFMSW is soluble.

TABLE 1: Characteristics of the organic fraction of municipal solid waste.

Parameter	Unit	Value
pH	-	5.2 ± 0.1
Humidity	%	72 ± 1.0
VS/TS	-	0.79
Total COD	g/kg_{TS}	1140 ± 13
Soluble COD	g/kg_{TS}	445 ± 9
Total VFA	$g_{COD/}kg_{TS}$	8.0 ± 0.5
NH_4-N	g/kg_{TS}	1.7 ± 0.1

2.2 OFMSW fermentation

Six batch reactors were operated at 35°C for 15 days, using 1,000 mL glass bottles with a reaction volume of 800 mL. Shredded OFMSW was placed in the reactors, and total solids concentration (TS) was adjusted to 10, 20, and 28%. The highest TS concentration corresponds to fresh OFMSW. To adjust the other solids concentrations, tap water was added. Three reactors with different solids concentrations were connected to an automated biogas monitoring system (AMPTS II, Bioprocess Control, Sweden), and the remaining three reactors were used for sampling. No mixing was performed in the reactors connected to the biogas monitoring system. The fermentation took place with the native microorganisms, and the pH was not adjusted at any time. In the sampling reactors, the content was mixed manually before obtaining samples. For every sample, determinations were made for total solids (TS), volatile solids (VS), soluble COD, ethanol, methanol, and VFA. VFA and alcohol concentrations were calculated based on the actual VS when samples were taken. The degree of acidification, or acidification rate (ηa), defined as the fraction of soluble COD converted to VFA, was calculated according to equation 1. To calculate the acidification degree, it is necessary to express the different determined VFA as COD equivalent. The sum of all VFA COD equivalents was divided by the total soluble COD of OFMSW.

According to Bolzonella et al. (2005) and Favaro et al. (2013), OFMSW contains enough naturally-occurring microorganisms to perform the anaerobic acid fermentation. Under this consideration, it was decided that supplementary inoculum for the fermentation was not necessary.

2.3 Adaptation of methanogenic inoculum

The inoculum used for the methanisation of the OFMSW fermentation products was digestate produced in the laboratory and adapted to OFMSW. For 58 days, a reactor was semi-continuously operated at 35°C. The reactor was fed with the same fresh OFMSW used in the fermentation stage and was run under the following criteria: a) Bicarbonate alkalinity to total alkalinity ratio (α index) higher than 0.6 (Sun et al., 2016); b) VFA concentration was kept under 1.5 g/kgOFMSW (Saveyn and Eder, 2014) and c) stable biogas production was considered when methane concentration was consistently over 60% in the biogas (Schievano et al., 2008). Before using the digestate for the methane production tests, it was kept under the same environmental conditions without feed for six days.

TABLE 2: Experimental arrangement for the methanisation tests.

	S/I ratio	Solids concentration during storage ($\%_{TS}$)		
		10	20	28
Fermented OFMSW	0.5	R1	R2	R3
	1.0	R4	R5	R6
	1.5	R7	R8	R9
Fresh OFMSW	0.5	R10	R11	R12
	1.0	R13	R14	R15
	1.5	R16	R17	R18

2.4 Methanisation of fermented OFMSW

For the methanisation, the substrate to inoculum (S/I) ratio was adjusted to 0.5, 1.0, and 1.5 as volatile solids to reach a reaction volume of 300 mL in 500 mL flasks. As a reference, another set of nine flasks was also placed in the temperature-controlled water bath and fed with fresh OFMSW instead of the fermented one (Table 2). Immediately after filling the flasks, the remaining air was washed out with nitrogen to guarantee anaerobic conditions. pH was not adjusted, and the experiment ran without buffer. Incubation took place at 35°C, without mixing, for 25 days, connected to an automated biogas counter (AMPTS II, Bioprocess Control, Sweden). A control containing only the inoculum was also monitored to determine endogenous methane production. Methane concentration in the biogas was determined daily by gas chromatography.

The S/I ratios for this research were selected according to articles reporting similar results. Di Maria et al. (2012) report that, for industrial OFMSW anaerobic digestion, lower S/I ratios are common (0.3 to 0.5, based on TS) in order to increase the methane production rates and to reduce malfunctioning risks. Karthikeyan and Visvanathan (2013) report values for successful batch dry anaerobic digestion of 10 to 60% inoculum (digestate from previous batches). Motte et al. (2013) conclude that S/I ratios between 2 and 6 can be managed in lab-operated reactors. Considering these reported results, the S/I ratios selected for this research include variations.

2.5 Analytical methods

pH, total COD, total (TS), volatile solids (VS), and ammonia nitrogen (NH4-N) were determined according to Standard Methods (APHA, 2017). Soluble COD was determined by photometry after 0.45 µm filtration. NH4-N was determined in the liquid fraction after centrifugation with a relative centrifugal force (RCF) of 4704 x g for 7 minutes (5,000 rpm, rotor diameter 16.7 mm). Ethanol and VFA (acetic, propionic, isobutyric, butyric, isovaleric, valeric, and hexanoic acids) were determined using a gas chromatograph (HP 5890 GC System) equipped with flame ionization detector (FID), Stabilwax column - DA, with hydrogen as the carrier. The sample was previously filtered using 0.22 µm cellulose filters. The biogas composition (CO_2 and CH_4) was determined using a gas chromatograph (SRI 8610c) equipped with a thermal conductivity detector, stainless steel column packed with silica gel (8600-PK1A), helium as carrier gas with a 27 mL/min flow rate. The detector temperature was 150°C.

2.6 Statistical analysis

The experimental results of methane production and reaction time were analyzed using multiple linear regression, considering a confidence level of 95%. Regression was used to make estimates or predictions for the dependent variables. The statistical analysis tool Solver for Excel was used.

3. RESULTS AND DISCUSSION

3.1 OFMSW fermentation

3.1.1 Effects of solid concentration on pH

Figure 1.a shows the pH behavior for the different solids concentrations. Since pH was not controlled, during the first day, in all cases, there was a rapid decrease to values near 4 for the two higher TS and slightly under 4 for the lowest TS. In reactors with higher TS (20 and 28%), pH did not vary significantly during the remaining 14 days. In the 10%TS reactor, pH dropped to values below 4.0 that can be attributed to lactic and formic acid formation, whose pKa values are 3.86 and 3.77, respectively (Xu et al., 2012). After day 6, pH increased to values slightly over 4, and it remained, with minor changes, until the end of the experiment. From those results, it can be concluded that pH decreases with decreasing solids concentration, at least during the first days. It has been reported that at low pH values, the fermentation of highly biodegradable substrates is limited. At such low pH values, VFA are in a dissociated form and can quickly diffuse through the cell membrane, lowering the internal pH and thus inhibiting cell activity (Warnecke and Gill, 2005). However, a positive aspect of low pH values is that, by inhibiting microbial activity, organic matter and energy losses are limited, allowing the preservation of the substrate during long periods and preserving the potential methane production (Wang, 2010; Jiang et al., 2013). Kalač (2012) evaluated the loss of organic matter during fermentation of energy crops, observing that, for total solids concentrations between 15 and 30%, the losses were minimal when the pH values varied between 4.1 and 4.45.

3.1.2 Hydrolysis

In all cases, on the first day of experimentation, soluble COD decreases (Figure 1.b). The heterogeneity of the substrate, together with the multiple and complex reactions during fermentation, makes that COD determinations cannot be as precise as desired. However, soluble COD's relative stability was observed from day 3 until the end of the experiment with average values of 301, 373, and 423 g/kgVS for solids concentrations of 10, 20, and 28%, respectively. The acid conditions, with pH values between 3.9 and 4.1, inhibited the hydrolytic and other microorganisms' activity. According to Nizami Bern. (2009), effective hydrolysis can occur when pH values are between 5.5 and 6.5. At pH values lower than 5.0, organic matter solubilization is limited (Zhou et al., 2018). In Figure 1.b, no tendency in the behavior of soluble COD can be observed. At the end of the fermentation, a reduction of volatile solids was observed: For the 10, 20, and 28% solids concentrations, the decrease was 12, 14, and 5 %, respectively.

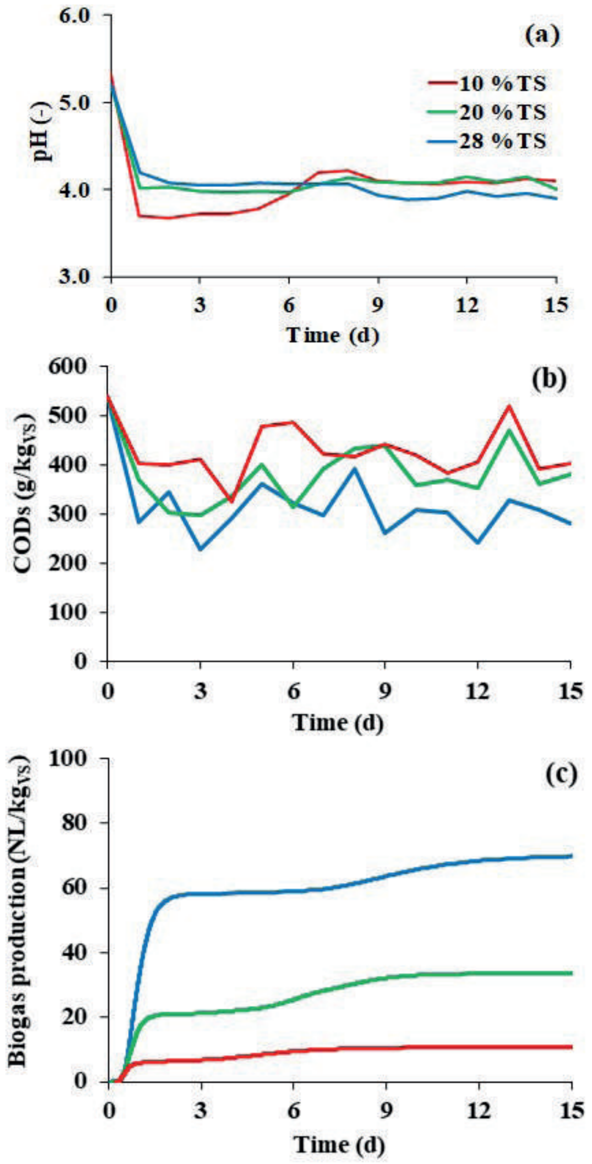

FIGURE 1: pH (a), soluble COD (b), and biogas production (c).

3.1.3 Production of fermentation gases

Figure 1.c also shows that the production of gases increased significantly during the first two days before the pH values decreased; this can also be compared with the sudden COD decrease during the same period. After the third day, pH and gases production stabilized because, as previously stated, lower pH values inhibited the biochemical activity. The higher gases production corresponds to the lower solids concentration.

3.1.4 VFA and alcohols production

Figure 2 shows that, for all three TS, total VFA production increased slowly and constantly until day 10, and then it continued without significant variations. Figure 2 also shows that VFA concentrations increased with decreasing solids concentrations. The highest VFA concentration was recorded on day 9 with 133 gCOD/kgVS in the reactor with 10%TS while, for the reactors with 20 and 28%TS, the highest VFA concentrations were 91 and 56 gCOD/kgVS, recorded at days 15 and 13, respectively. The highest total VFA concentrations correspond to maximum acidification rates of 52, 25, and 15% for TS of 10, 20, and 28%TS, respectively. The acidification rates indicated in Figure 2, calculated at the end of the experiment for the initial soluble COD, are only 18% for 10%TS, 14% for 20%TS, and 7% for 28 TS: Acidification rates are inversely proportional to TS. This means that only a small fraction of OFMSW was transformed to VFA. These yields are lower than those reported in other articles where low pH is considered the inhibitor of acidogenic microorganisms (Wang et al., 2014). Silva et al. (2013) determined a maximum OFMSW acidification rate of 32% under mesophilic conditions and pH-controlled at 6.0.

With a different purpose, Moretto et al. (2019) evaluated VFA production from OFMSW under mesophilic conditions and at different pH values, reporting acidification rates of 55, 79, and 81% for pH of 5.0, 7.0, and 9.0, respectively. These results confirm that the acidification rate is strongly dependent on pH and, to reach a maximum VFA yield, the pH needs to be controlled between 5.0 and 6.0 (Gottardo et al., 2015). At the same time as pH can act as an inhibitor, it is also an essential factor to determine the VFA composition during fermentation (Jiang et al., 2013), especially on the production of acetic, propionic and, butyric acids, whose formation is more sensitive to pH variations than other VFA (Liu et al., 2012). In this research, the primary acids produced were acetic, propionic, and butyric in all cases. The acetic acid in all three reactors represents more than 90% of the total VFA determined at the end of the experiment. This a positive feature considering that acetate is the preferred substrate by methanogens. In the reactor with 10%TS (Figure 2.a), greater diversity on VFA production was observed as valeric, and hexanoic acids were also present. Two phases of fast VFA production were identified: The first peak at the end of day 2 resulted in an increase of approximately four times the acetic acid concentration to the initial value. Ethanol production also increased about five times its concentration during the first two days. Zoetemeyer (1982) states that lower pH values favor the production of long-chain fatty acids. The second phase of acetic acid production can be associated with two processes: Firstly, acetogenesis from ethanol can be observed as acetic acid concentration increases and ethanol concentration decreases (reactors with 20 and 28%TS) or increased fast during the first two days, as in the case of the reactor with 10%TS (Figure 2.a). Secondly, VFA with more than three carbons were transformed through acetogenesis and made acetic acid concentrations increase. Yuan et al. (2006) report that, during anaerobic fermentation, VFA with more than three carbons are easily degraded to form acetic acid. For the two lower TS concentrations, acetic acid production increased steadily while, for the highest TS concentration, the increase was slower. In all three cases, after day 10, acetic acid production practically stopped and remained almost unchanged until the end of the experiment.

After acetic acid, propionic acid accounted for the most significant VFA fraction. As with acetic acid, propionic acid

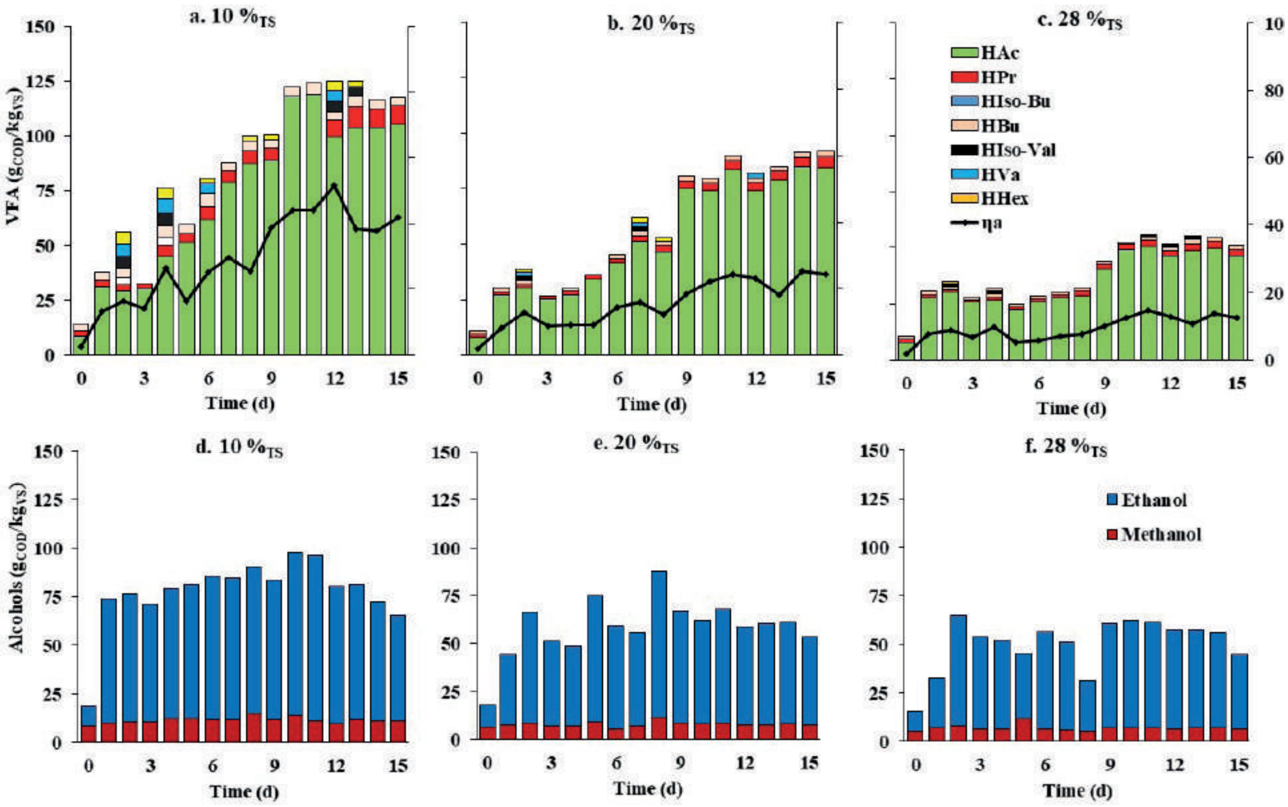

FIGURE 2: VFA and alcohol production, and acidification rate (ηa). HAc (acetic acid), HPr (propionic acid), HIsoBu (isobutyric acid), HBu (butyric acid), HIso-Va (isovaleric acid), HVa (valeric acid), HHex (hexanoic acid).

concentration was inversely proportional to TS concentration. This concurred with another study where propionic acid concentration increased as TS decreased, and pH was low (4.0 to 4.5) (Wang et al. 2014). Propionic acid accumulated at lower pH values. Thermodynamically, higher hydrogen partial pressure inhibits acetogenic microorganisms, and propionate cannot be processed: During anaerobic digestion, hydrogen and acetate are consumed to form mainly and CO_2 making that acetogenesis runs continuously (Henze et al., 2008). Butyric acid represents the third largest VFA fraction. Its concentration did not change significantly over time because, as with propionic acid, acetogenic microorganisms were inhibited and could not process butyric acid.

Ethanol and methanol production was also monitored (Figure 2). In all cases, during the first two days of fermentation, a rapid increase in ethanol production together with H2 and CO2 production could be observed. The fast gases production (Figure 1.c) was also observed during this same period. He et al. (2012) report similar results; they studied the hydrolysis and acidification of food waste at 35°C with 18%TS and finding that ethanol and acetic acid constituted 99% of the metabolites after seven days of fermentation. Dogan et al. (2009) report that during OFMSW fermentation with 25%TS, ethanol and acetic acid were the primary metabolites produced when the pH value ranged between 3.5 and 4.0. Figure 2 shows that the lack of soluble carbohydrates can explain the ethanol concentration's relative stability after day 3 as the particulate material's hydrolysis decreased. Also, lower pH values inhibit the metabolism of alcohol-producing bacteria or yeast.

On day 11, the maximum ethanol concentration in the reactor with 10%TS (Figure 2.d) was 84 gCOD/ kgVS while, for reactors with 20 (Figure 2.e) and 28%TS (Figure 2.f), the maximum ethanol concentrations were 58 and 59 gCOD/ kgVS, respectively. The highest ethanol concentration was recorded in the reactor with the lowest solids concentration. For the three reactors, ethanol represents 52% of the total concentration of metabolites in terms of abundance. The production showed no significant variations over the whole experiment (Figure 2). Methanol also was determined. Methanol is an essential final product from pectin metabolism (Schink and Zeikus 1980), which is part of the cell walls of most fruits and vegetables found in OFMSW. The methanol concentration did not change significantly over time, and this can be explained as this metabolism product cannot further develop under stress, such as low pH values. Also, any other organisms can use it as substrate under these conditions.

3.2 Methanisation of fermented OFMSW

3.2.1 Adaption of inoculum

To produce an adapted inoculum for the methanisation tests, a batch reactor was operated with a mixture of OFMSW and anaerobic granular sludge at a ratio of 0.5 gVS OFMSW per gram VS of granular sludge. As shown in Figure 3.a, in the mixture of OFMSW with the granular sludge, both pH and alkalinity ratio (α index) resulted in values considered adequate for methanogenesis (Sun et al., 2016). From the start, the VFA concentration in the reactor increased, presenting the highest concentration on day 6, but without negatively affecting the methanogenic activity; at this time, the methane concentration in the biogas was over 60% (Figure 3.b). The highest methane production rate was recorded during the first 6 days. After day 25, the methane production continued increasing at a slow regular rate until the end of the experiment.

The reactor was operated for 48 days until the parameters considered adequate for an adapted inoculum were met. The digestate used to seed the OFMSW for methane production had an α index of 0.8, and total VFA concentration of 5.0 gCOD/kgVS. The biogas had a methane concentration of 64%, complying with the criteria to be considered an adequate inoculum.

3.2.2 Methane production

To evaluate their methane production, every one of the OFMSW fermentation products from the experiments at 10, 20, and 28%TS were inoculated with the adapted methanogenic digestate to adjust substrate to inoculum ratios (S/I, as volatile solids) to 0.5, 1.0, and 1.5 (Figure 4). The characteristics of the adapted digestate used as inoculum were pH = 8.3, humidity = 89%; VS/TS = 0.73; total COD = 340 g/kgVS; NH4-N = 31 g/kgVS. Fresh OFMSW was used as control. Shortly after mixing the different substrates with inoculum, the pH stabilized to values near 7 (Table 3). Mixing the adapted inoculum (pH 8.3) with fresh and fermented OFMSW caused the overall acidic pH values to increase to values near neutrality. For all cases, the initial pH values decrease as the S/I ratio increases, and the reactors with fermented OFMSW showed lower pH values than the ones with fresh OFMSW. Although the initial pH values decreased according to solids concentration, they cannot be considered potential inhibitors. During the reaction, pH increased to values slightly alkaline, indicating healthy methanisation. Figure 4 shows the methane production curves for all 18 combinations indicated in Table 3.

The initial slopes for fresh OFMSW are slightly higher than those for fermented OFMSW and, after several days, the final methane production and the reaction times for fermented OFMSW are higher than for fresh OFMSW. At the reactions' end, the pH values were near 8.0 in all cases, meaning that the pH drop at the beginning could have influenced the process. However, methanogenesis consumed VFA rapidly, preventing pH from being an inhibitor.

The lower S/I ratios produce methane rapidly during the first 3 to 4 days, and then, the reaction reaches an end to continue as endogenous methane production (Figure 4). Fresh OFMSW finishes methane production (359 NL/kgVS) earlier than fermented OFMSW, and their final values are lower than those of fermented OFMSW (410 NL/kgVS) (Table 4). The curves corresponding to the S/I = 0.5 (Figure 4.a) produce methane slower than those with the lower substrate concentration: the methane production from fermented OFMSW runs until day 9, and, after day 10, the production is caused by endogenous substrates. For S/I = 1.0 (Figure 4.b) and fresh OFMSW, during the first six days, the curves run parallel to those of fermented OFMSW and finish earlier than those of fermented OFMSW. The methane production curves for S/I=1.5 (Figure 4.c) behave differently than the other two cases: Methane production is slower and takes more than 15 days to finish; fresh OFMSW with the highest S/I requires more than 23 days to complete. Independent of the TS concentration, Figure 4 shows that, for S/I=0.5 and 1.0, the initial slopes are practically the same for all curves and that the end of methane production is different for every one of them. It is important to note that all curves display the same performance for S/I=0.5 and 1.0, with only one stage during methane production. For S/I=1.5, the curves present similar behavior, but with different values; differently than the other two S/I values, for S/I=1.5, the highest TS concentrations report the highest methane yields where fermented OFMSW produce more methane than fresh OFMSW. The curves for S/I=1.5 present two stages, where the first and second stages end faster for the higher TS concentrations than for the lower ones.

Table 4 shows that fermented OFMSW with 10%TS and S/I=1.0 resulted in the most significant difference compared to fresh OFMSW with 99 NL/kgVS (32% more for fermented than for fresh OFMSW). The next most significant difference is in reactors with 20%TS and S/I=1.0 with 30% more methane production for fermented than for fresh OFMSW. These results conclude that the S/I ratio of 1.0, the same amount of fermented OFMSW and inoculum, is

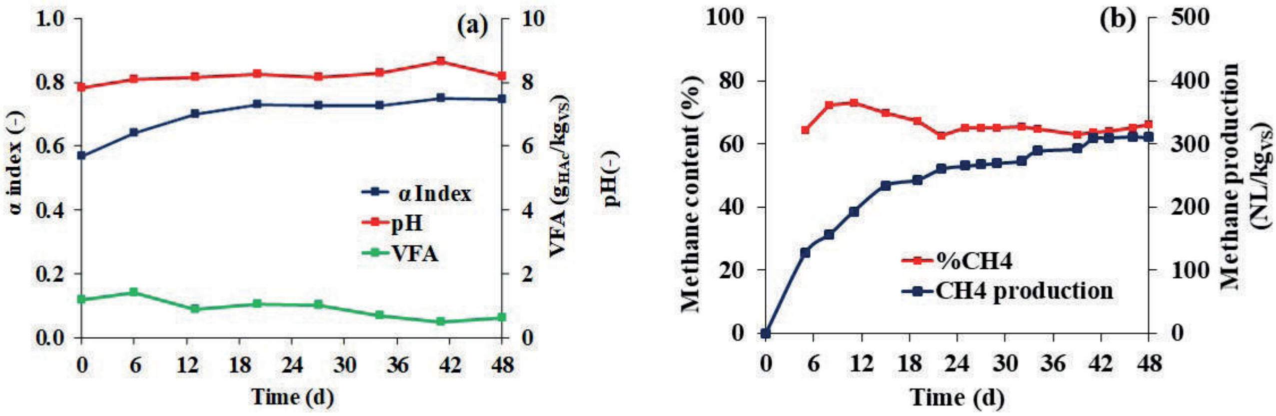

FIGURE 3: Behaviour of reactor operational parameters during inoculum adaptation.

TABLE 3: pH values in the combinations of fermented and fresh OFMSW with adapted methanogenic inoculum.

$\%_{TS}$	S/I ratio	Fermented OFMSW		Fresh OFMSW	
		Initial	Final	Initial	Final
10	0.5	7.0	8.2	7.6	8.2
	1.0	6.8	8.1	7.3	8.1
	1.5	6.3	7.3	7.2	8.1
20	0.5	6.8	8.2	7.6	8.3
	1.0	6.2	8.1	7.3	8.1
	1.5	5.8	8.1	7.1	8.1
28	0.5	6.9	8.2	7.6	8.2
	1.0	6.3	8.2	7.3	8.1
	1.5	6.0	8.0	7.1	8.2

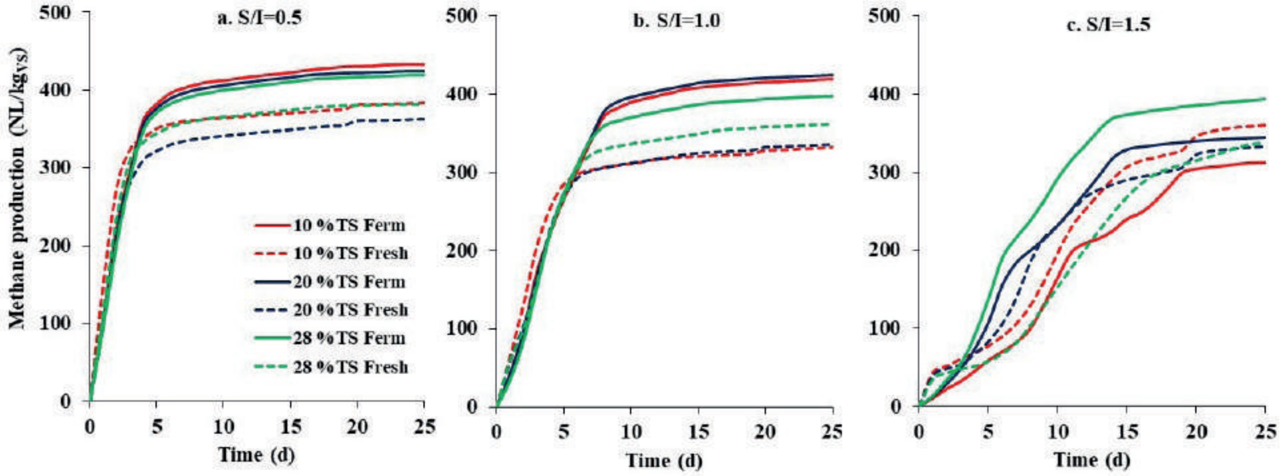

FIGURE 4: Specific methane production of fermented OFMSW (Ferm) and fresh OFMSW (Fresh) for 25 days according to different S/I ratios and TS.

the best combination of all the ones tested. Methanisation at low S/I ratios of fermented OFMSW resulted in reaction time reductions between 11 and 40% compared to fresh OFMSW. These results show that fermentation of OFMSW increases the subsequent methane production and, at the same time, decreases the reaction time. Fermented OFMSW with the highest S/I ratio of 1.5 reports the lowest methane production and the highest reaction time when compared to fresh OFMSW: the increase in reaction time is proportional to solids concentration with 9, 29, and 56% for 10, 20, and 28%TS, respectively.

The lowest solids concentration during silage is responsible for the highest methane production, indicating that decreasing VS during silage did not significantly affect the subsequent methane production (Table 4). Borreani et al. (2018) observed similar behaviour in a system with decreasing organic matter; they report VS losses of 20% and no decreases in methane production.

Table 4 shows the methane production from fermented and fresh OFMSW for the different TS concentrations and S/I ratios. According to Holliger et al. (2016), the reaction finishes when methane production values do not variate more than 1% as an average of three consecutive days. The reaction time corresponds to the time needed for the reaction to finish. According to Figure 3 and Table 4, methane production of fermented OFMSW improved between 2 and 32% compared to fresh OFMSW. In only one case, the reactor with 10%TS and S/I=1.5, methane production was lower for fermented OFMSW than for fresh OFMSW in 14%. Similar results were reported by Schievano et al. (2014); they determined an increase between 23 and 43% in energy recovery through methane production in the process of anaerobic co-digestion of corn silage and fruit and vegetable residues in a two-stage process. Liu et al. (2006) observed 21% increase in methane production from the digestion of domestic solid waste in batch reactors operated in two stages. On the other hand, Voelklein et al. (2016) recorded an increase of 23% when they used food as a substrate for digestion in a two-stage methanogenic reactor.

Figure 5 shows the methane production rates (slopes from curves in Figure 3) according to the substrate to inoculum ratio and substrate concentration. The slopes for 20 and 28%TS for fermented OFMSW are similar without significant differences between them, indicating that no differences in methane production rate can be differentiated from one another (compare with Figure 4). Coincidentally, for fermented OFMSW, all three lines cross at a S/I ratio of 1.0, proving that the previously proposed conclusion that the S/I ratio of 1.0 is the best condition to produce methane. The lowest solids concentration behaves differently: it

TABLE 4: Methane production and reaction time of fermented and fresh OFMSW.

$\%_{TS}$	S/I ratio	Methane production (NL/kg_{VS})		ΔCH_4 production (%)*	Reaction time (d)	
		Fermented OFMSW	Fresh OFMSW		Fermented OFMSW	Fresh OFMSW
10	0.5	410	359	+14	7	9
	1.0	408	309	+32	9	15
	1.5	306	357	-14	23	21
20	0.5	403	338	+19	8	9
	1.0	404	312	+30	10	12
	1.5	335	328	+2	22	17
28	0.5	397	363	+9	9	9
	1.0	377	336	+12	10	12
	1.5	376	337	+12	25	16

**Represents the variation in methane production of fermented OFMSW compared to fresh OFMSW*

has a higher slope, meaning that lower S/I ratios produce methane more rapidly than higher S/I ones when the S/I ratio is under 1.0. When S/I ratios and solids concentrations are high, methane can be produced less rapidly as solids concentrations increase.

Figure 5 indicates that, for fresh OFMSW, the methane production rates are different that fermented OFMSW. The lines cross at the highest S/I ratio, indicating deficient methane production at higher substrate concentrations. As expected, the rates increased with decreasing S/I ratios and, among the three tested fresh OFMSW concentrations, the lowest, corresponding to 10%TS, present higher methane production rates.

Higher methane production from fermented OFMSW can be explained as a function of the acidification rate. The higher the acidification rate, the higher is the methane production reached under lower substrate concentrations. The metabolites produced during fermentation are readily available for methanogenic archaea. Fresh OFMSW does not have metabolites from fermentation. Similar processes are reported by Buffière et al. (2018), who investigated the hydrolysis of cattle slurry and corn silage mixtures in two-stage systems: They found that, after 48 h of hydrolysis, the methane production increased significantly, and concluded that in a two-stage anaerobic digestion process, first-stage fermentation improved the substrate availability to methanogens. The use of inoculum adapted to the substrate has proven to be a determining factor in achieving higher methane yields (Campuzano, 2015; Brown and Murphy, 2013). In this study, the adapted inoculum was responsible for higher methane production rates.

3.3 Regression analysis

The results were analyzed using multiple linear regression to determine the influence of the operational parameters over methane production and reaction time. ANOVA significance test for methane production from fermented OFMSW shows that, for a p-value higher than 0.05, solids concentration, as %TS, does not significantly contribute to methane production and reaction times. The results show too that, for a p-value lower than 0.05, the substrate to inoculum ratio, S/I, is the most critical parameter. This means

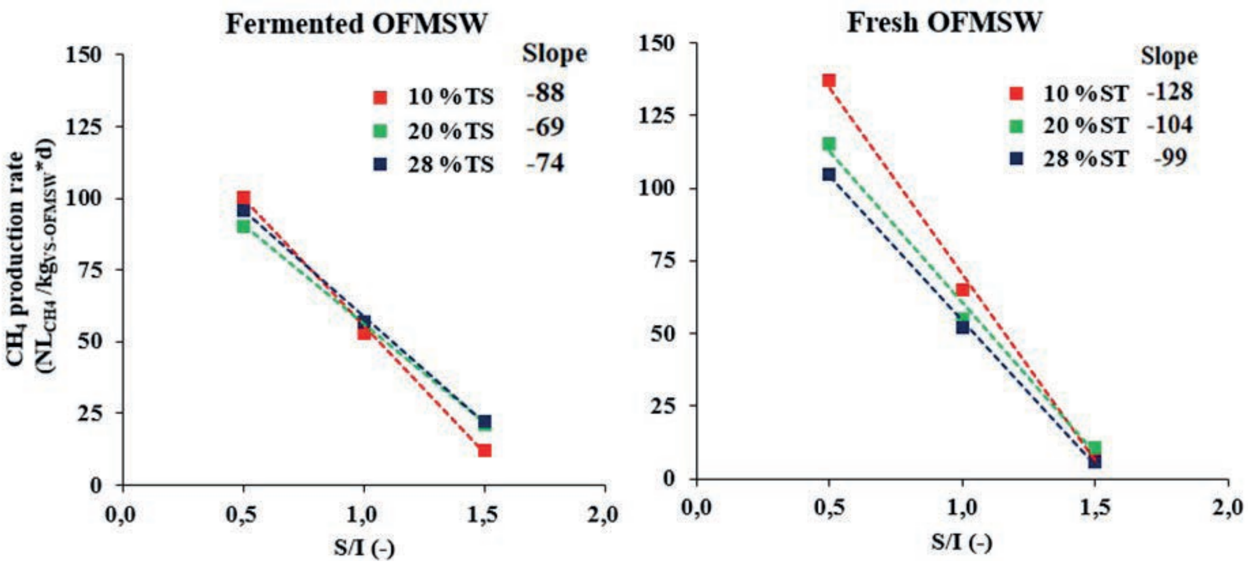

FIGURE 5: Methane production rates according to the substrate to inoculum ratio, S/I, and TS.

that, independently of solids concentration during OFMSW fermentation, when using high inoculum concentrations for methanisation of fermented OFMSW, methane production increases, and the reaction time decreases. The lowest p-value obtained for the reaction time was 0.002, and it shows that methanisation of fermented OFMSW at a low S/I ratio has more importance than reducing the reaction time.

For fresh OFMSW methanisation, p-value were 0.500 and 0.898 for S/I and %TS, respectively. When p-values are higher or equal to the significance value, then these parameters have no significant influence over methane production from fresh OFMSW. For lower reaction times, the analysis shows the p-value for S/I and %TS of 0.000 and 0.036, respectively. The meaning of these values is that lower S/I ratios allow a faster VFA uptake and other metabolites during the methanisation of fresh OFMSW without increasing methane production. Several authors report that solids concentration significantly affects methane production (Fernández et al., 2008; Dong et al., 2010; Abbassi-Guendouz et al., 2012; Campuzano, 2015). However, they do not consider the influence of inoculum concentration. These authors conclude that higher methane yields can be obtained with increasing solids concentration. Uncertainty arises from the results of Forster-Carneiro et al. (2008). They also experimented with fresh OFMSW varying the concentrations of both substrate and inoculum concentrations, concluding that the reactors with lower substrate concentrations produce more methane; unfortunately, they do not make any comments on the combined effects of the two variables.

4. CONCLUSIONS

During OFMSW fermentation, VFA and alcohol production increased with decreasing solids concentration. Together acetic acid and ethanol constitute more than 90% of all metabolites. OFMSW fermentation allowed 32% more methane production for the lowest solids concentrations than for fresh OFMSW, indicating that the VS loss during fermentation does not affect subsequent methane production. For fermented OFMSW, the S/I ratio of 1.0 is the best condition to produce methane. Methanisation of fermented OFMSW at low S/I ratios resulted in reaction times reductions between 11 and 40% than fresh OFMSW. Methane production from fermented and fresh OFMSW increased substantially with a decreasing substrate to inoculum, S/I, ratios. Independently of substrate solids concentrations, high inoculum concentrations increase methane production and reduce methanisation times.

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METHANE PRODUCTION AND BROMATOLOGICAL CHARACTERISTICS OF THE DIFFERENT FRACTIONS OF ORGANIC MUNICIPAL SOLID WASTE

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ABSTRACT

In some countries, garden trimmings are not considered part of urban solid wastes. Lignocellulosic substances contribute to heterogeneity, complicating the analysis of the organic fraction of municipal solid waste (OFMSW) and, subsequently, for methane production. Some of the substances contained in OFMSW are readily biodegradable, and others are not. This work analyses OFMSW from Mexico City and the methane production from its separate components. From OFMSW, nine fractions were visually identified and separated. Including bromatological and fibre analysis, the characterisation of OFMSW and its components was made to determine how the different substances influence methane production. Together, branches, dry leaves, fresh garden trimmings, unsorted wastes (mainly garden trimmings), kitchen paper, and waste vegetables represent 56% of OFMSW in weight. Fruit waste and unsorted organics contribute to 60% of the total methane production. Except for branches and dry leaves, methane production increases inversely with the content of lignocellulosic compounds. Animal waste, having the highest concentrations of proteins and lipids and the lowest in lignocellulosic substances, is characterised by the highest level of methane production. Fibre-rich fractions in OFMSW contributed with little or no methane production. Higher concentrations of lignocellulosic substances in the fractions resulted in lower methane production rates.

1. INTRODUCTION

By 2050, the world is expected to increase waste generation by 70 percent, from 2.01 billion tonnes of waste in 2016 to 3.40 billion tonnes of waste annually (Hoornweg et al., 2013). Individuals and governments make decisions about waste management that affect the daily health and cleanliness of communities. Increasing waste trends are particularly intense in less developed countries (Kaza et al., 2018). Waste production trends not only increase resource stress but also contribute to greenhouse gases. A transformation toward resource-circular systems and sustainable municipal solid waste management is necessary (Wainaina et al., 2019). Growing environmental pressure has caused regional/national targets to divert waste from landfills and increase the recycling and recovery rate. While developed countries struggle to reach a zero MSW production through recycling, developing countries struggle to avoid open dumpsites by implementing controlled landfill sites (Kumar et al., 2019; Manjunathaa et al., 2019).

A preferred treatment method for the organic fraction of municipal solid waste (OFMSW) is anaerobic digestion (AD) because it allows the production of methane as fuel, and the resulting solids (digestate) can be used for soil improvement (Möller and Müller, 2012). The whole AD microbial degradation process is divided into four consecutive biological processes: 1) the hydrolysis of complex organic molecules to soluble monomers takes place in the first step; 2) acidogenesis or fermentation is the process by which the soluble monomers from hydrolysis are converted to alcohols, volatile fatty acids (VFA), namely acetic, propionic and butyric acids, and CO_2 and hydrogen; 3) acetogenesis is the step where several of the previously

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produced VFA and alcohols are converted into acetate, which is an essential molecule used by methanogens as substrate and 4) methanogenesis is the final step where different archaea can use acetate, CO_2 and hydrogen to produce methane as a final product (Bajpai, 2017).

Methane from the organic fraction of the municipal solid wastes (OFMSW) is a potential energy source. Results from several studies indicate that biogas produced by microbial activity does not contain only CO_2 and CH_4, but also other compounds that need consideration when using this biogas as fuel to generate electricity (Rasi et al., 2006). Papurello (2019) demonstrates efficient biogas sampling procedures and precise analytical methods. Several undesired compounds need to be removed from biogas before using it as fuel. There are several standard processes to remove CO_2, hydrogen sulphide, and other sulphur compounds from biogas, but special attention needs to be paid to organic silicon compounds that cause abrasion in the engines (Ohannessian et al., 2008).

OFMSW heterogeneity (Naroznova et al., 2016) and complex composition (VALORGAS, 2010) are limiting factors of OFMSW for biogas production. Methane potential depends on substrate characteristics and biodegradability (Campuzano and González-Martínez, 2016). According to reports from VALORGAS (2010), Kobayashi et al. (2012), Naroznova et al. (2016), and Alibardi and Cossu (2016), the knowledge of OFMSW composition and characteristics allows the improvement of urban solids waste management and subsequent methane production. Eventually, it is possible to determine which fractions or components can be considered for methane production and which ones need to be avoided during source separation. According to the classifications in different countries, the contents of fiber-rich substances can significantly vary. For example, kitchen paper and garden trimmings are accepted as organic waste in countries like the United Kingdom, Finland, and Denmark. Still, they are not allowed in other countries like Norway and Sweden (Naroznova et al., 2016).

Geographic and socioeconomic aspects affect OFMSW composition (VALORGAS, 2010). Studies have been performed to determine how much methane or hydrogen can be produced according to the bromatological and chemical characteristics of the different OFMSW fractions (Kobayashi et al., 2012; Alibardi and Cossu, 2016; Naroznova et al., 2016; Edwiges et al., 2018). Every study classifies OFMSW according to their specific objectives, and there are essential differences between selected components. In Japan, Kobayashi et al. (2012) analyzed Kyoto OFMSW, and they found, in kitchen wastes, animal rests, vegetables, paper, cereals, food wrappings, tea and coffee bags, and some garden trimmings. In Italy, in an anaerobic digestion plant, Alibardi and Cossu (2016) found meat, fish, cheese, fruits, vegetables, pasta, bread, and some unidentifiable substances. In Denmark, Naroznova et al. (2016) characterized OFMSW in the city of Halsnæs. They classified the fractions as animal food waste, vegetable food waste, kitchen paper, vegetation waste, molded fibers, animal straw, dirty paper, and dirty cardboard.

Using bromatological and physicochemical analysis, several authors found that OFMSW fractions have different influences on methane production during anaerobic digestion. One concern about fiber-rich compounds in OFMSW are lignocellulosic compounds and their relative concentration of cellulose, hemicellulose, and lignin (Teghammar et al., 2010, Triolo et al., 2011). Edwiges et al. (2018) found that the biochemical methane potential (BMP) improved when the wastes contained higher amounts of lipids while lignin negatively affected methane production. Xu et al. (2014) also noted that methane production was negatively affected when the substrate was lignin-rich. Extractable substances, such as cellulose and other compounds, are desirable because they have a positive effect on methane production. Labatut et al. (2011) observed that the highest methane productions were from substrates rich in fat and carbohydrates and that the lowest rates were obtained with lignocellulosic-rich substances.

Lignocellulosic biomass is the most important and more abundant product from photosynthesis. In vegetal biomass, hydrogen bridges, forming microfibers with hemicellulose and covered by lignin compounds (Taherzadeh and Karimi, 2008), bind cellulose chains. Hemicellulose is the essential union between lignin and cellulose fibers, providing rigidity to the cellulose-hemicellulose-lignin compounds, resulting in highly recalcitrant compounds (Hendriks and Zeeman, 2009). High fiber concentrations in organic wastes indicate low biodegradability, and lignin presence is undesired during methane production in anaerobic digestion (Alibardi and Cossu, 2016; Fonoll et al., 2016). According to Campuzano and González-Martínez (2016), lignocellulosic compounds represent approximately 40% in weight in Mexico City's OFMSW, meaning that every kilogram of OFMSW contains 225 gVS/kg, and, from them, only 135 g are biodegradable. The remaining 90 g are slowly biodegradable or not susceptible to transformation to biogas; the authors did not mention which components contain higher amounts of lignocellulosic compounds.

Even though OFMSW is a potential source of bioenergy, it is crucial to determine how its composition affects the anaerobic digestion process. Independently of the specific local legislation, there will always be different substances in OFMSW. Paper and cardboard are generally present and also are cellulose-rich compounds (Kobayashi et al., 2012; Naroznova et al., 2016; González-Miranda et al., 2016). Fruits and vegetables are cellulose and fiber-rich substances and represent almost 74% of OFMSW (Nielfa et al., 2015; Naroznova et al., 2016; Edwiges et al., 2018). Animal rests are present in OFMSW in lower quantities and are not fiber-rich (Kobayashi et al., 2012; Naroznova et al., 2016).

Considering the discrepancies observed in other published research about OFMSW classification, this work aims to determine the influence of different compounds in the identifiable OFMSW fractions on methane production. For this purpose, it was necessary to identify and quantify OFMSW components according to their physicochemical and chemical characteristics.

2. MATERIALS AND METHODS

OFMSW from Mexico City was used for this purpose, considering that garden trimmings and market wastes are allowed as part of OFMSW.

2.1 OFMSW sampling and classification

OFMSW sampling and conservation was made on one day when source-sorted organic wastes are collected. Considering that between 380 and 450 trucks discharge solid wastes every day in the Coyoacán transfer station, eleven trucks were randomly selected and, from each one, approximately 100 kg were separated and thoroughly mixed; from the resulting amount, 200 kg were set apart according to the quartering method (ASTM D5231-92, 2016). Inorganic materials and plastics were hand-separated and discarded. The sample was distributed in 2-liter freezing bags, and they were immediately frozen at -20°C.

In the laboratory, approximately 20 kg of slowly defrosted OFMSW were placed in trays, and, carefully, all components were visually identified, manually separated, and grouped according to their apparent origin. To homogenize the separated and identified fractions before analysis, they were ground using a 0.35 W electric disc mill (DelRey, Mexico).

2.2 Biochemical methane potential tests

For biochemical methane production (BMP), an AMPTS II system from Bioprocess Control AB (Sweden) was used. The inoculum was anaerobic granular sludge from the wastewater treatment plant of a large beer factory in Mexico City. The granular sludge was washed three times using tap water and concentrating it using a centrifuge to separate exogenous dissolved substances from the solids. For BMP, VDI 4630 recommendations were followed: Inoculum to substrate ratio (ISR, as volatile solids) was 4:1; temperature, 35°C and per triplicate (VDI 4630, 2016). According to experiences from previous projects, the duration of the tests was set to 21 days (Campuzano and González-Martínez, 2015). Biogas sampling and analysis were performed daily. A blank only with inoculum was used as a reference (blank).

The theoretical biochemical methane production (TBMP) is widely used to estimate methane potential. Triolo et al. (2011) calculated TBMP after modifying the method proposed by Møller et al. (2004), where they included lignin with the following empirical formula, $C_{10}H_{13}O_3$, and with it, they calculated the TBMP of lignin through the Symons and Buswell (1933) equation resulting in a value of 727.1 $NLCH_4$/kglignin. Triolo et al. (2011) propose Equation 1 to calculate TBMP. Equation 1 includes lipids ($C_{57}H_{104}O_6$), protein ($C_5H_7O_2N$), carbohydrates ($C_6H_{10}O_5$), and lignin ($C_{10}H_{13}O_3$); values expressed as g/kgVS.

$$TBMP=(C_{57}H_{104}O_6\cdot 1014+C_5H_7O_2N\cdot 496+C_6H_{10}O_5\cdot 415+C_{10}H_{13}O_3\cdot 727\cdot 0.001 \quad (1)$$

TBMP is used to evaluate the biodegradability of a substrate using Equation 2 (Triolo et al., 2011).

$$Anaerobic\ biodegradability = BMP/TBMP \cdot 100\% \quad (2)$$

2.3 Analytical methods

The different fractions and original OFMSW were analysed for humidity, total solids (TS), volatile solids (VS), chemical oxygen demand (COD), Kjeldahl nitrogen (KN), ammonia nitrogen (NH4-N), total phosphorus (TP), and pH. These determinations were performed according to APHA (2005). Protein, grease and fats, total carbohydrates, lignin, cellulose, and hemicellulose were determined according to Van Soest (Van Soest, 1963, Goering and Van Soest, 1970) and Official Methods of Analysis of AOAC International (AOAC, 2012). Biogas composition was determined using an SRI 8610c gas chromatograph equipped with a thermal conductivity detector and stainless steel silica gel packed column 8600-PK1A using helium gas as carrier at a flow rate of 27 mL/min.

For carbohydrates fractioning, the method proposed by the Cornell Net Carbohydrate and Protein System (CNCPS) was used. This method separates the fractions according to their degradability. Structural carbohydrates were calculated as the difference between neutral detergent fibre and non-soluble protein. Non-fibrous carbohydrates or non-structural carbohydrates are the difference between total carbohydrates and structural carbohydrates (Sniffen et al., 1992, Lanzas et al., 2007).

3. RESULTS AND DISCUSSION

Approximately 20 kg OFMSW were overnight defrosted at 4°C. The visually identified fractions were: Food waste from animal origin (animal waste), flour products, fruits, vegetables, kitchen paper, dry leaves, branches (garden cuts), fresh trimmings, and a fraction was called unsorted as these wastes could not be visually identified or separated but mainly were related to garden trimmings and dry leaves. Table 1 shows a description of the substances found in every fraction; Figure 1 shows the fractions' images.

TABLE 1: Identified components in OFMSW.

Fraction	% OFMSW	Visually identified components
Fruits	36	Rests, peelings, and seeds from oranges, lemon, pineapple, watermelon, bananas, papaya, mamey (Pouteria sapota), mango, and avocado peeling and seeds, grapes, and tamarind shells and seeds
Vegetables	13	Jicama, carrots, different types of chilies, peanuts, potato peeling and rests, red beet, peas, fresh corn grains, garlic, onions, and seeds from sunflower
Animal waste	8	Red and white eggshells, beef leftovers and bones, chicken skin and bones
Flour products	3	Tortilla (typical Mexican flatbread from maize)
Fresh trimmings	6	Bugamvilia, fresh grass, pine needles, eucalyptus, palm, and ash tree leaves
Dry leaves	7	Leaves from different unidentified trees and maize leaves.
Branches	4	Different unidentified small and thin tree branches (from trimmings)
Kitchen paper	2	Paper napkins, kitchen paper, and some wax paper
Unsorted	21	Mostly from dry leaves and trimmings, in small pieces, unidentified

FIGURE 1: The identified OFMSW fractions (branches are not shown).

3.1 Characterisation of OFMSW fractions

3.1.1 OFMSW fractions

The fraction with the highest weight percentage is fruits (36%) followed by unsorted (21%) and then vegetables (13%) (Table 1). The fractions with the lowest contribution in OFMSW are kitchen paper (2%) and cereal waste (3%). Considering unsorted, dry leaves, garden trimmings, and branches together, garden wastes amount to 37% in weight; this last value is significant compared to reports from other countries (Naroznova et al., 2016; Kobayashi et al., 2016).

Vegetables, fruits, and fresh trimmings present the lowest solids concentrations with values from 22 to 28%. In comparison, the fractions with the highest solids concentrations are branches with 55%, followed by animal wastes and flour products with 45 and 44%, respectively (compare humidity in Table 2). When comparing these results with the ones Naroznova et al. (2016) found in Denmark, similarities can be found: they report 24% for vegetables and, in this research, 23 and 22% were determined for fruits and vegetables, respectively. For wastes from animal origin, Naroznova et al. (2016) report 41% solids concentration and 45% are observed in this research.

The fraction with the lowest VS corresponds to vegetables with 192 g/kg and the highest to branches with 475 g/kg. Naroznova et al. (2016) report values similar to the ones in this research: food animal wastes, 344 g/kg, vegetables, 223 g/kg, and vegetation wastes (garden trimmings), 240 g/kg; in the case of kitchen paper, they report 491 g/kg compared to 265 g/kg in this work. The VS/TS ratio is an indicator of the organic material concentration related to total solids: The highest VS/TS ratio belongs to fruits with 0.9, followed by cereals with 0.87, branches, 0.86, and kitchen paper, 0.86; the lowest value is for animal wastes

TABLE 2: Characterization of OFMSW fractions.

	[3]Humidity%	VS/TS	[3]VS g/kg	[3]Protein	[2]Lipids	[3]Total Carbohydrates	Hemicellulose*	Cellulose*	Lignin*
				g/kg$_{VS}$					
Animal	55±0.6	0.70	320±2.6	373±45.6	390-396	70±7.5	3	4	39
Kitchen paper	69±0.1	0.86	265±1.3	62±2.4	202-204	970±44.3	20	207	32
Flour	56±0.4	0.87	380±3.5	123±5.3	119-122	406±16.1	232	53	32
Vegetables	78±0.4	0.84	192±2.8	121±5.5	95-97	464±25.8	63	102	65
Fruits	77±0.2	0.90	206±4.3	94±3.0	78-83	505±12.2	7	121	32
Branches	45±0.1	0.86	475±4.9	44±2.3	93-100	451±14.5	155	37	401
Fresh trimmings	72±0.2	0.81	227±1.6	130±9.3	147-155	504±12.2	51	111	69
Dry leaves	64±0.2	0.84	306±2.4	89±5.9	156-176	394±13.6	49	98	68
Unsorted	[5]66±0.5	0.73	[5]252±4.0	131±4.3	170-189	294±18.6	62	33	165
OFMSW	[5]71±1.1	0.78	[5]228±3.0	168±4.7	133-136	791±41.3	9	102	53

*[3] Three replicas; [5] Five replicas; [2] In lipids only two replicas were made; both values are reported. *Average of three replicas; no standard deviations are available.*

with 0.7, containing eggshells and bones, and unsorted materials with 0.73. Campuzano and González-Martínez (2016) report a VS/TS ratio of 0.75 for other OFMSW samples in Mexico City; VS/TS of the OFMSW sample taken for this research is 0.78.

From Table 1 and Figure 2, the following observations can be drawn:

- Wastes of animal origin is the fraction with the lowest concentration of lignocellulosic substances (46 g/kgVS) and the highest in fat and protein.
- Fruits have the third-highest concentration of carbohydrates; fat and protein concentrations are relatively low. The concentration of lignocellulosic compounds is low but, with 160 g/kgVS, it is three times higher than the amounts in wastes of animal origin.
- Dry leaves contain high hemicellulose and lignin concentrations with 49 and 68 g/kgVS, respectively. Confusing can be that fat concentration is relatively high in dry leaves; it needs to be considered that the maize leaves found in OFMSW are disposed of after being used to wrap a traditional meal called tamales (fatty).
- Vegetables present higher concentrations in protein with 121 g/kgVS, and the concentration of degradable carbohydrates is higher than the average among the fractions with 299 g/kgVS. This fraction reports lower lignin concentrations than dry leaves but higher in cellulose and hemicellulose with 102 and 63 g/kgVS, respectively.
- Fresh trimmings, as expected, have higher cellulose and lignin concentrations with 111 and 69 g/kgVS, respectively. Nevertheless, it presents lower hemicellulose concentrations and is the third in protein concentration. The relatively high lipids concentration suggests that this fraction was previously in contact with other fat-rich materials.
- Kitchen paper has the highest cellulose concentration with 207 g/kgVS and the lowest lignin concentration with 32 g/kgVS; it also has the highest degradable carbohydrates concentration and the second place in lipids with 203 g/kgVS.
- Unsorted materials have third place in lipids and protein concentrations and one of the highest lignocellulose concentrations. Lignin is the second highest with 165 g/kgVS and, after animal rests, the lowest in degradable carbohydrates.
- Flour-rich substances present higher lignocellulosic concentrations, being hemicellulose the highest with 232 g/kgVS. Protein, lipids, and degradable carbohydrates are 123, 121, and 121 g/kgVS, respectively. As no amylase was used to determine the fiber, starch could not be avoided. Most probably, the starch interferes with the determination, and hemicellulose reports slightly higher values than expected.
- Of all fractions, branches have the highest lignocellulose concentrations with 401 g/kgVS. It has the lowest concentrations of protein and lipids and relatively high concentrations of degradable carbohydrates.
- OFMSW can be considered as a general average value of all determinations shown in Table 2.

The general characteristics of Mexico City's OFMSW are similar to the organic solid wastes from other countries. Table 3 shows a comparison of the OFMSW characteristics found in several papers with the ones reported in this research. Grinsted, Prague, Lisbon, and Padua report TS above 300 g/kg; under 300 g/kg are Mexico City, Verona, Copenhagen, and Southampton. Other cities like Cadiz, Kerala, Canton and Beirut, report values under 200 g/kg. Similar values can be found for VS in these same cities. The VS/TS value reported in this research is 0.78, which is

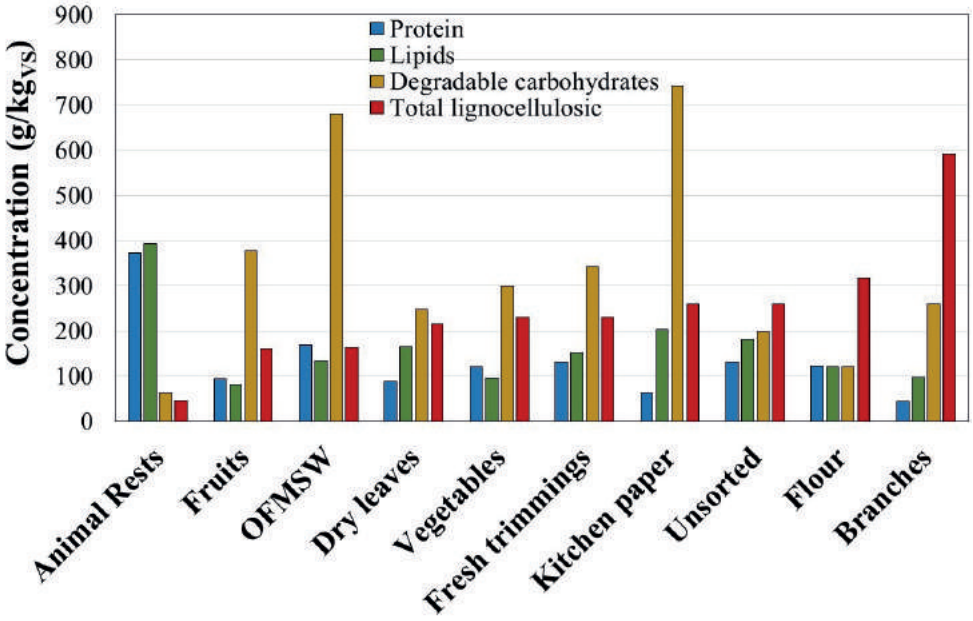

FIGURE 2: Comparison of bromatological characteristics in the separated fractions. The fractions are organized in ascending concentration of total lignocellulosic substances.

TABLE 3: Comparison of the OFMSW characteristics of this research with other countries. All values are wet based (raw OFMSW)

City	TS g/kg	VS g/kg	VS/TS -	KN g/kg	TP g/kg	COD g/kg	Reference
Kerala, India	187	169	0.91	1.04	-	-	Sajeena Beevi et al., 2015
Padua, Italy	305	281	0.92	7.7	1.16	575	Alibardi y Cossu, 2015
Verona, Italy	288	228	0.79	28	2.4	347	Bolzonella et al., 2005
Lisbon, Portugal	338	276	0.82	5.1	1.7	-	VALORGAS, 2010
Luton, UK	237	218	0.91	7.4	1.2	-	VALORGAS, 2010
Southampton, UK	277	244	0.88	8.9	1.9	-	Banks et al., 2011
Beirut, Lebanon	186	172	0.93	-	0.7	-	Ghanimeh et al., 2012
Prague, Chech Republic	325	231	0.71	4.5	0.7	-	Hanc et al., 2011
Cadiz, Spain	172	74	0.43	26.0	-	140	Forster-Carneiro et al., 2008b
Canton, China	184	113	0.62	4.2	0.4	-	Dong et al., 2010
Karlsruhe, Germany	255	225	0.88	7.8	-	350	Nayono et al., 2009
Copenhagen, Denmark	283	250	0.88	7.4	1.4	-	Davidsson et al., 2007
Grindsted, Denmark	356	307	0.86	6.3	-	431	Hartmann and Ahring, 2006
Mexico City, Mexico	297	223	0.75	5.4	1.8	304	Campuzano and González-Martínez, 2015
This research	290	228	0.78	6.1	3.9	294	

slightly lower than most of the values reported for other cities; the lowest VS/TS values of 0.43 and 0.42 correspond to Cadiz and Canton. Important differences are observed in macronutrients: KN presents values between 1.04 and 28 g/kg for Kerala and Verona, respectively. Grindsted, Copenhagen, Karlsruhe Luton, Lisbon, and Padua report similar KN to this research. TP values vary between 0.4 g/kg in Canton to 2.4 g/kg in Verona, and, surprisingly, this research reports 3.9 g/kg TP, the highest of all values. Table 3 shows that fruits, vegetables, and fresh trimmings contain elevated phosphorus concentrations. Except for Cadiz, other cities present higher COD values than the one from this research. No explanation can be offered about how COD can be related to TS because the reported values in Table 3 do not show any correlation.

3.1.2 Bromatological characteristics

According to the characteristics of the substances identified in the fractions, the following observations can be made.

Protein. As expected, animal wastes present the highest protein concentration with 373 g/kgVS and branches and paper, the lowest with 44 and 62 g/kgVS, respectively. The highest values agree with the ones reported by Kobayashi et al. (2012) for kitchen wastes and wasted animal rests. Alibardi and Cossu (2016) report higher concentrations for meat, fish, and cheese rest. Edwiges et al. (2018) report similar values as those in this work for protein in fruits and vegetables of 15.9%VS. Campuzano and González-Martínez (2016) report a total value of 152 g/kgVS for OFMSW, like the one in this research of 168 g/kgVS. From the total protein in OFMSW, animal wastes contribute with the highest value of 27%, and the lowest is for paper with less than 1% (Figure 2).

Lipids. Like protein concentration, animal wastes present the highest value with 393 g/kgVS; the lowest value is for fruits with 81 g/kgVS. Kobayashi et al. (2012) report similar results, and Alibardi and Cossu (2016) report similar results for fish, meat, and cheese wastes. For Brazil, Edwiges et al. (2018) report an average value of 4.5%VS for fruits and vegetables. From the total grease and oil in OFMSW, the highest concentration is for unsorted wastes with 25%VS and the lowest for paper with less than 2%VS (Figure 2).

Carbohydrates. Table 2 shows that the highest carbohydrate concentration is for paper with 970 g/kgVS. Coinciding with this work, Kobayashi et al. (2012) report 959 g/kgVS for wrapping paper. Alibardi and Cossu (2016), González-Miranda et al. (2016), and this work report that the lowest value for carbohydrates is for animal rests. Campuzano and González-Martínez (2016) report 529 g/kgVS for OFMSW, while this research determined a higher value with 791 g/kgVS. As a percentage of OFMSW, fruits correspond to 41% of the total carbohydrates, and animal wastes are the lowest with 2%.

Cellulose. Table 2 shows that kitchen paper has the highest cellulose concentration with 207 g/kgVS, followed by fruits with 121 g/kgVS. Kobayashi et al. (2012) and González-Miranda et al. (2016) show that paper also contains the highest cellulose concentration and Naroznova et al. (2016) indicate dirty carton with the highest cellulose followed by moulded fibres and fruits and vegetables with 120 g/kgVS. Kobayashi et al. (2012) report lower cellulose contents in wastes from animal origin with 1.6 g/kgVS, and Naroznova et al. (2016) also note lower cellulose content in waste animal origin with 2%VS. Edwiges et al. (2018) report average cellulose values in fruits and vegetables of 17.1%VS; in this research, the average cellulose concentrations in fruits and vegetables are 12.1 and 10.2%VS, respectively; from the total cellulose concentration in OFMSW, fruits contribute with 50% and wastes from animal origin with less than 1%.

Hemicellulose. Like cellulose, wastes from animal origin present the lowest concentration with 3 g/kgVS. In contrast, Kobayashi et al. (2012) found that "other kitchen wastes" have 2.7%VS and Naroznova et al. (2016) found 3%VS hemicellulose in dirty paper. González-Miranda et al. (2016) report similar values to those in this work for fruits with 1%VS. (10 g/kgVS). This work's highest values for hemicellulose are for flour (cereals) with 232 g/kgVS. Kobayashi et al. (2012) report the highest values for coffee and tea, while Naroznova et al. (2016) and Alibardi and Cossu (2016) note this for straw for pets and in vegetables. Edwiges et al. (2018) show average values in fruits and vegetables of 9.4%VS. From the total hemicellulose in OFMSW, unsorted wastes contribute 25% and animal wastes with less than 1%.

Lignin. The lowest lignin concentrations determined in this work were for paper, fruits, and flour with 32 g/kgVS, while the highest values are for branches with 401 g/kgVS, followed by unsorted wastes with 165 g/kgVS. Naroznova et al. (2016) found that, in their analysis, the lowest content was for animal rests with 2%VS and the highest for straw for domestic animals; they report the same lignin value for paper and dirty paper with 30 g/kgVS. Considering fruits, Naroznova et al. (2016) and this research agree on a lignin concentration of 4.5%VS and González-Miranda et al. (2016) and Edwiges et al. (2018) with a slightly higher value of 6.4%VS. Of the total lignin content in OFMSW, unsorted wastes contribute 37%, and paper with less than 1%.

Considering that lignocellulosic compounds are not readily biodegradable, Figure 2 compares the different OFMSW fractions according to the concentration of total lignocellulosic substances and degradable carbohydrates together with protein and lipids. Degradable carbohydrates are the difference between total carbohydrates and structural carbohydrates (cellulose and hemicellulose). The lowest values for total lignocellulosic substances are for animal wastes and the highest for branches. Degradable carbohydrates are high for kitchen paper with 743 g/kgVS, followed by OFMSW and fruits with 680 and 377 g/kgVS, respectively. Figure 2 also shows no direct relationship between carbohydrates and the concentration of lignocellulosic compounds.

3.1.3 Nutrients in OFMSW fractions

Table 4 shows COD and Kjeldahl and ammonia nitrogen, as well as total phosphorus concentrations. The highest COD corresponds to fresh trimmings with 1,395 g/kgTS, followed by fruits, animal wastes, unsorted, and flour with values between 1,287 and 1,222 g/kgTS. From the OFMSW fractions, the lowest COD is for branches with 742 g/kgTS. Considering total COD in OFMSW, fruits and unsorted together contribute 52% of the total.

Table 4 shows that values for Kjeldahl nitrogen are highest in animal rests (protein) with 42 g/kgTS. The lowest KN value was found in kitchen paper. The fractions animal rests, fruits, and unsorted contribute 72% of the total Kjeldahl nitrogen.

For ammonia nitrogen in OFMSW fractions (table 4), the highest value is for animal rests with 1.7 g/kgTS and the lowest for flour products (mainly tortilla) with 0.4 g/kgTS. All other fractions contain low NH4-N concentrations and, compared to KN, they can be considered negligible for the purpose of methane production. Like KN in OFMSW fractions, NH4-N contribution is mainly attributed to unsorted, fruits, animal, and vegetables, with a total of 77% of the total. Campuzano and González-Martínez (2016) report similar values for Kjeldahl nitrogen for OFMSW with 18.2 g/kgTS.

OFMSW is mixed at the origin, during transportation, separation, and selection of fractions; these procedures contribute to transferring fluids among the fractions and the liquids' partial homogenization. Total phosphorus concentrations in branches are lowest with 2.4 g/kgTS, which is less than 1% of OFMSW; vegetables have the highest with 18.9 g/kgTS (15% of OFMSW). Fruits follow with 15.9 g/kgTS (35% of the total in OFMSW). In contrast, González-Miranda et al. (2016) report that unsorted has the highest value with 67.6% of the total in OFMSW and paper the lowest with 0.4%.

3.2 Methane production

Except for branches, the curves in Figure 3 show diauxic behaviour. During the first three days, methane production increased rapidly, and then it slowed down and, after sever-

TABLE 4: COD, Kjeldahl nitrogen (KN), ammonia nitrogen (NH4-N), and total phosphorus (TP) in OFMSW fractions.

	COD*	KN**	NH_4-N**	TP*	COD	KN	NH_4-N	TP
	g/kg_{TS}				% OFMSW			
Fruits	1287±71.4	13.4±0.4	1.0±0.07	15.9±0.8	29	22	20	35
Vegetables	1311±90.9	16.3±0.7	1.3±0.08	18.9±0.2	10	9	11	15
Animal	1274±45.6	42.0±1.8	1.7±0.12	8.3±0.4	12	30	18	8
Flour	1222±58.4	17.1±0.7	0.4±0.03	9.0±0.1	4	4	1	3
Fresh trimmings	1395±81.0	16.5±1.2	1.1±0.09	14.0±0.4	6	5	5	6
Dry leaves	1224±72.4	12.0±0.8	1.2±0.10	10.5±0.3	8	6	8	7
Branches	742±23.4	12.8±0.6	1.3±0.01	2.4±0.3	5	3	8	1
Kitchen paper	1430±64.0	8.5±0.4	0.7±0.06	10.5±0.5	3	1	1	2
Unsorted	1265±77.2	15.3±0.5	1.4±0.12	12.9±0.6	23	20	28	23
OFMSW	1014±47.2	21.1±0.5	3.6±0.11	13.5±0.6	100	100	100	100

** Seven replicas, ** Three replicas*

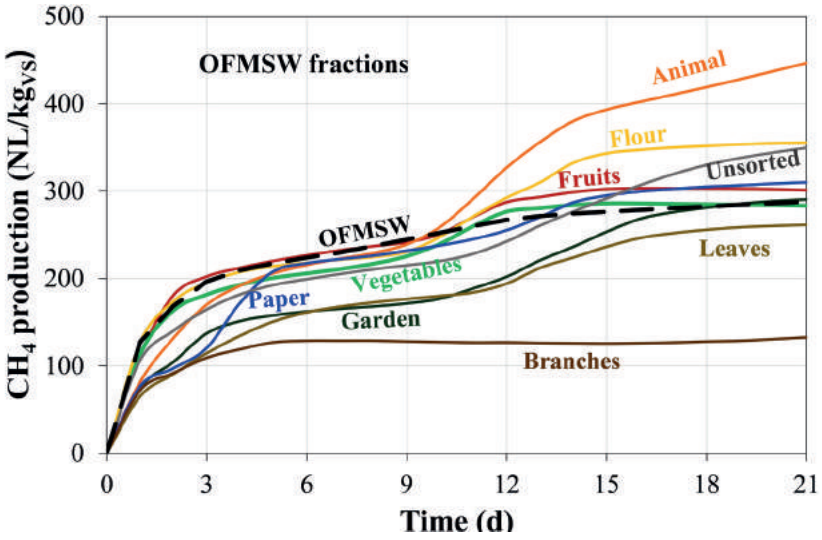

FIGURE 3: Methane production from OFMSW fractions.

al more days, it increased again to reach a point where the production became deficient until the end of the 21 days.

Animal waste and unsorted are the only fractions where low methane production continues after day 21. Branches produce methane only until day 5. On the third day, fruits and vegetables show increasing methane production, representing more than 60% of the methane produced in 21 days, indicating that these fractions contain readily biodegradable substances. From day 4 to 9, all fractions produced little methane, and, after day 10, the production increased in all fractions except in branches. After day 18, the methane production receded except for animal rests and unsorted organics. The diauxic behaviour can be related to several processes during anaerobic digestion: Readily degradable substances will be transformed first, and then other substances that require longer degradation times. Analysis of the methane production curves in Figure 3 allows determining the behaviour of the previously described sequence (Campuzano and González-Martínez, 2015).

Table 5 shows methane production and concentration in the biogas after 21 days for every fraction and OFMSW. The highest methane production belongs to animal wastes with 447 NL/kgVS, and the lowest belongs to branches with 133 NL/kgVS. This can be related to their composition: Animal waste has the highest protein and lipids concentrations and the lowest in lignocellulosic compounds; branches have the highest concentrations in lignocellulosic substances and the lowest in protein and lipids (see Figure 2). Labatut et al. (2011) conclude that they obtained the highest methane production from substrates rich in fat and carbohydrates and the lowest rates with lignocellulosic-rich substances. Kitchen paper absorbs fluids from other fractions, and because of this, it presents a relatively high methane production with 310 NL/gVS. It can be concluded that methane production decreases with increasing concentrations of lignocellulosic substances. Xu et al. (2014) also noted that methane production was "negatively affected" when the substrate was lignin-rich and that extractable substances, such as cellulose and other compounds, are desirable because they have a "positive" effect on methane production. When Xu et al. (2014) calculated the methane production based on volatile solids, they concluded that low methane

TABLE 5: Biogas and methane production after 21 days' anaerobic digestion. Because of the characteristics of this test, only one curve was determined for every substrate.

Fraction	CH$_4$ in biogas %	CH$_4$ production NL/kg$_{vs}$	TBMP NL/kg$_{vs}$	Anaerobic Biodegr. (%)	CH$_4$ in fraction %
Fruits	55	301	361	84	35.1
Vegetables	56	283	397	71	12.0
Animal waste	62	447	641	70	11.7
Flour	53	355	375	95	3.3
Fresh trimmings	61	291	477	61	5.5
Dry leaves	61	261	425	61	5.8
Branches	64	133	599	22	1.6
Kitchen paper	60	310	662	47	1.9
Unsorted	58	350	489	72	22.7
OFMSW	55	288	586	49	100

production is related to low VS and vice versa. Fruits represent the highest contribution to total methane production in OFMSW with 35.1%, followed by unsorted with 22.7%. Fruits, unsorted, vegetables, and animal rests together represent 81.5% of the total methane production from OFMSW. Table 5 shows that methane concentration in the biogas for OFMSW was 55% and that animal rests, fresh trimmings, dry leaves, branches, and kitchen paper had methane concentrations above 60%, indicating healthy anaerobic digestion.

Figure 4 compares experimentally determined methane production (BMP) with the theoretical one (TBMP), with and without considering the presence of lignocellulosic substances in the fractions. TMBP represents the amount of methane produced from all organic material in the sample, and it does not consider the complexity of the organic substances. Although lignocellulosic substances are considered recalcitrant to microbial degradation, several of their components, such as cellulose and hemicellulose, can be transformed under anaerobic conditions (Paul and Dutta, 2018). When these components are closely linked to lignin, they become unavailable to the microorganisms. As lignocellulosic compounds are not readily biodegradable or biodegradable, Figure 4 shows that, in all cases, TMBP has higher values than BMP.

Except for animal waste, all other fractions show that TBMP with lignocellulosic substances is higher than without them. This difference indicates that lignocellulosic substances contain biodegradable molecules, most probably hemicellulose and cellulose. Surprisingly, flour showed the highest biodegradability with 95%, followed by fruits with 84% and unsorted with 72%. As expected, branches has one of the highest TBMP and the lowest experimental BMP with 22% of the theoretical. Figure 4 also shows that TBMP, considering the presence of lignocellulosic substances, in all cases, has higher values than without them, but these differences are slight. It can be concluded that the presence of lignocellulosic substances in the different OFMSW fractions contributes little to methane production.

In fruits, vegetables, and unsorted, the TBMP without lignocellulosic substances and the experimental values are similar, meaning that BMP was produced from readily biodegradable substances. This can also mean that lignocellulosic substances have lower lignin concentrations or that the lignin clusters did not prevent biodegradable carbohydrates from biodegradation.

4. CONCLUSIONS

From Mexico City's OFMSW, nine fractions were visually identified, from which branches, dry leaves, fresh garden trimmings, unsorted wastes (primarily garden wastes), kitchen paper, and vegetable wastes together contain lignocellulosic compounds with 56% in weight. Together with fruit-waste and unsorted organics account for 60% of total methane production. Branches contain the highest concentration of lignocellulosic compounds, and it delivers the lowest methane production. Much differently, animal rests have higher protein and lipids concentrations and lower lignocellulosic substances leading to the highest level of methane production. Fibre-rich fractions in OFMSW contributed with little or no methane production. The methane production obtained in the laboratory from all fractions and OFMSW resulted in lower values than the theoretical ones.

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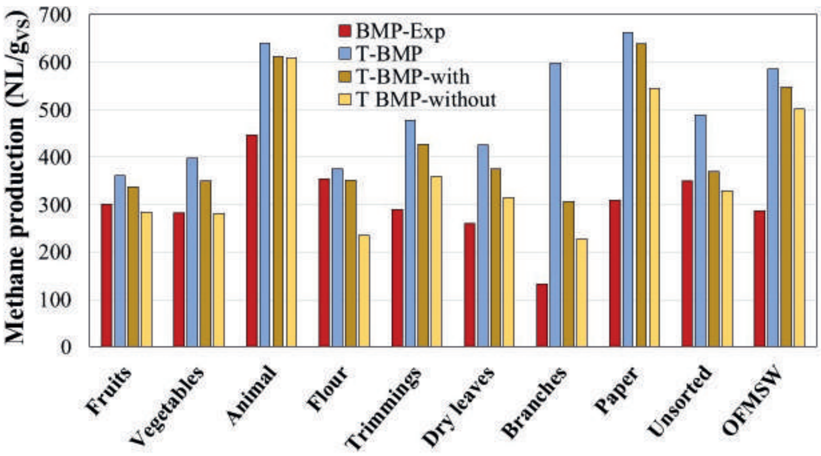

FIGURE 4: Methane production from OFMSW fractions after 21 days reaction. BMP-Exp (experimental); T-BMP (theoretical BMP); T-BMP-with (lignocellulosic substances); T-BMP-without (lignocellulosic substances).

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3-ROUTES PLATFORM FOR RECOVERY OF HIGH VALUE PRODUCTS, ENERGY AND BIO-FERTILIZER FROM URBAN BIOWASTE: THE REVENUE PROJECT

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ABSTRACT

This innovative Biorefinery platform is based on the integration of a mild thermal pre-treatment and a solid/liquid separation unit to parallel-integrated bioprocesses specifically selected on food waste distinctive chemical composition: a liquid fraction, rich in readily fermentable sugars, to be transformed into valuable biobased products, and a solid organic residue to enhance biomethane production generating a fully hygienized digestate to be recycled. The preliminary results in terms of VFAs yields and composition from the acidogenic stage, and the methane conversion rate from the anaerobic digestion of the solid residue, are here presented. The integration of a mild thermal pretreatment promoted the transformation and the conversion rates into volatile fatty acids through the optimization of operational pH and HRT of the primary fermentation step. Secondary fermentation reaction as chain elongation to MCFAs (as caproic acid) occurred in the same reactor without external donor addiction probably because of the in situ formation of lactate. The solid residue fed into semi-pilot anaerobic digesters assured process stability (no acidification phenomena occurred) despite the short HRT of 20d. The weakening of the lignocellulosic matrix by the thermal pre-treatment had a positive impact on the anaerobic conversion increasing methane production (+20%), in particular during weekend, when feeding was stopped.

1. INTRODUCTION

In circular bioeconomy, the recycling of biowaste is a crucial strategy for optimizing the use of the available biomass resource base in order to promote a comprehensive sustainable FW management system, stimulated by the increasing awareness of the exhaustion of non-renewable natural resources (Agler et al. 2011; Braguglia et al., 2018; Coma et al., 2017; Kleerebezem and van Loosdrecht 2007). Urban biowaste is traditionally seen as a costly problem in economic and environmental terms. This equation can be reversed by designing more effective recovery and processing systems to turn organic waste into a source of value and contribute to restoring natural capital. Biorefineries could be a central technology in this endeavor (Alibardi et al., 2020). Operating in a similar way to petrochemical refineries, they employ a range of techniques – such as feedstock pretreatments, biological processes, enzymatic conversions – to transform source separated biowaste into valuable chemicals, products and energy. In Italy, in the last decade, intensive source separation of biowaste (food waste, FW, and green park waste, GW), has been extended also to medium/large towns and metropolitan areas, attaining best-practice performances in terms of quantity and quality of the collected biowaste.

Anaerobic digestion (AD), regarded by EU waste legislation as ideal recycling operation in the framework of the circular economy, is currently considered as one of the best environmental-friendly alternatives, because of its limited environmental footprints, high potential for energy recovery, producing carrier material for biofertilizers. However, the economic value of biogas and digestate is limited, enabling the development of alternative processes that yield higher-value endproducts. Carboxylic acids as acetate, propionate, butyrate and valerate, have a wide variety of applications in the chemical industry, mainly as bulk

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industrial chemicals (Arslan et al., 2016), in the production of bioplastics (Zacharof and Lovitt 2013; Valentino et al., 2019), biofuels and chemicals (Lee et al., 2014), or in biological nutrient removal. The conversion of organic waste to bulk chemicals is 3.5 to 7.5 times more profitable than its conversion to biomethane (Pfaltzgraff et al., 2013). At the same time, the European energy target from renewable sources (Eurostat, 2020) is 20% (by 2020) and at least 32% by 2030 (Italy reached 17.8% in 2018).

Moreover, it is well known that anaerobic digestion of Food waste to produce biomethane is often limited by system instability induced by FW characteristics. The availiability of prompt biodegradable carbohydrate fraction of FW was shown to allow the establishment of stable and active fermentative populations (Tonanzi et al., 2018) with the reduction and progressive inhibition of methanogenic biomass, thereby resulting in VFA accumulation and a pH decrease until biomethane drop (Capson-Tojo et al., 2016; Tonanzi et al., 2020, Zhang et al., 2012).

The innovative 3-Routes Platform proposed here is based on the integration of a thermal pretreatment capable of significantly increasing the fraction of fermentable organic carbon from FW. The final goal is changing the status of the feedstock to become more suitable for the production of high-value products as short-medium chain fatty acids from the liquid stream, and biomethane (and hygienized digestate) from the solid residue.

Pretreatment has been a major argument of research during the past 30 years and the AD improvement in terms of increased solubilization and H_2/CH_4 yields are well established, in particular for sludge (Braguglia et al., 2018; Carrère et al., 2016; Cesaro and Belgiorno, 2020). Very few studies (Kleerbezem et al., 2015; Zhang et al., 2019), aimed for determining the impact of pretreatment on VFAs production potential from FW as proposed here. Mild thermal pretreatment can, in fact, significantly alter both physical and chemical properties of FW, resulting generally in increased solubilization (Pagliaccia et al., 2016, 2019; Montecchio et al., 2017), in reduced lignocellulosic matrix, and improved digestate hygienization (Ariunbataar et al., 2015). Cellulose, hemicellulose and lignin containing wastes as OFMSW (source separated organic fraction) and FW (Romero-Cedillo et al., 2017), need further pretreatment in order to make the substrate more accessible to microbial degradation (Loow et al., 2016). Pecorini et al. (2016) reported increase of soluble COD, carbohydrates and proteins concentration in the pretreated OFMSW sample, while lignocellulosic and starch fraction were found to be lower in the pretreated FW by Pagliaccia et al. (2019).

The strategies for enhancing VFAs production is to promoting the acidogenic process, by inhibiting the methanogenesis, operating a bioreactor with optimized operative conditions and feeding patterns. The selective production of organic acids from various feedstocks is an emerging field of research. Which fermentation products are produced during the fermentation step, under which conditions and by which microorganisms represent open questions receiving increasing attention in recent years, but to our knowledge, only few studies focused the attention on hydrolysis enhancement with feedstock pretreatment, by treating sludge not FW (Morgan-Sagastume et al., 2011; Zhang et al., 2019). Numerous environmental factors determine the product spectrum of mixed culture fermentations, and the microorganisms dominating the system. This project fills the knowledge gap by investigating the impact of operative conditions on fermentation process performances (VFA composition, yields, stability) together with quantitative, predictive relationships between the complex microbial community structure and the fermenter functional outputs (Crognale et al., 2021). Correlation between the microbial community structure and the fermentation process performances is still a field full of gaps, especially in the case of semi-continuous long term processing with real feedstock.

The innovative rationale of this approach, never investigated up to now, is to exploit the potential of thermal pretreatment of food waste in solubilizing carbohydrates and proteins, in order to investigate the impact on acidogenesis in terms of VFAs yield and product spectrum. The economic value of VFAs produced during anaerobic digestion is intrinsically high, but further processing and conversion of this mixed carboxylates can enhance economic benefits even more (Marshall et al., 2013). One biological post-processing step investigated in this project is the microbial chain elongation of the short chain carboxylates into longer chain products as caproic acid, via syntrophic interactions, when reduced compounds as lactate or ethanol (electron donor) are added (Steinbush et al., 2011).

In this project, the challenge is to minimize (or completely avoid) the addition of external donor by steering the hydrolysis and acidogenesis stage introducing the thermal pretreatment step of the food waste substrate to improve both organics hydrolysis and the successive fermentation and/or chain elongation process by starting from real biowaste. The microbiome responsible for VFA production and chain elongation will be investigated by means of several biomolecular tools including high-throughput 16S rRNA gene sequencing. For the first time, in this project, amphiphilic ty nanoparticles will be applied to investigate the fatty acids adsorption potential from fermentation broth.

The preliminary results in terms of VFA composition and yield of semicontinuous fermentation tests of the liquid extract, and methane yield by the anaerobic digestion of the residual solid phase, are here presented and discussed, for untreated and thermal pretreated food waste.

2. MATERIALS AND METHODS

2.1 Substrates

FW was collected from the cafeteria of the research area “Roma 1” of the National Research Council. The cafeteria produces approximately 400 kg of FW per week, which consisted of mixed cooked and uncooked food such as pasta, rice, soup, bread, fruit and vegetable peelings. After sampling, the FW underwent an accurate manual screening to match the quantities of the various fractions with the chosen composition: 70% fruit and vegetable waste, 15% pasta and bread and 15% dairy residues. Successively, sorted scraps were shredded by a food processor, prior to being stored at -20°C. The anaerobic inoculum used for the start-up of the acidogenic tests was extracted from a lab-scale fermenter fed with FW and then acclimated to liquid extract, while the inoculum for the biomethane potential tests (BMP) and the anaerobic digestion of FW residues was obtained from a full-scale digester treating sewage sludge.

2.2 Thermal pretreatment

The frozen FW sample was thawed and mixed with tap water (1:4 w/w) to maximize the effectiveness of pretreatment, namely a hydrothermal treatment carried out in a bench scale autoclave Laboklav 25b, with total capacity of 25 L (SHP Steriltechnik AG, Germany). Operative temperature and pressure were set at mild conditions, namely T = 134°C and p = 3.2 bar. Retention time was set at 20 minutes on 2.5 L of sample. The applied specific thermal energy resulted around 9250 kJ/kgTS.

2.3 Liquid solid separation unit

The liquid/solid separation phase on raw and pretreated FW was performed with a bench scale centrifuge Rotanta 460 (Hettich, Germany) operating at 4600 rpm for 10 min. The liquid phase was successively filtered through a 0.5 mm mesh sieve to remove the coarse residue particles (Figure 1).

2.4 Analytical methods

Total and volatile solids were determined according to standard methods (APHA, 1998). The pH was detected by a portable pH meter Eutech Instruments pH 700. Soluble (sCOD), total COD and total nitrogen, measured in duplicates, were determined by means of COD Cell Test (EPA method 410.4) and the Nitrogen Cell test (EN ISO 25663) by Spectroquant Merck. Proteins and carbohydrates were measured on filtered liquid samples (glass filters GF/C Whatman, 1.2 μm porosity). Protein content was determined by bicinchoninic acid protein assay (Sigma Aldrich) using standard solution of bovine serum albumin (Smith et al., 1985). Carbohydrates determination was based on a modified DuBois method (Braguglia et al., 2012; DuBois et al., 1956). Total proteins content was estimated by multiplying the total organic nitrogen content by a factor of 6.25.

The biogas composition was measured using a Perkin-Elmer Auto System Gas Chromatograph equipped with a thermal conductivity detector (TCD) as described in Gianico et al. (2013). Volatile fatty acids (VFA) were analysed by injecting 1 μL of filtered (0.22 μm porosity) liquid sample into a Perkin Elmer Auto System gas-chromatograph equipped with a FID detector (flame ionization detector) as reported in Gagliano et al. (2015)..

2.5 BioMethane potential tests (BMP)

BMP tests on raw and pretreated FW were performed in mesophilic conditions (37°C) by using the Automatic Methane Potential Test System (AMPTS-II, Bioprocess Control, Sweden), equipped with 500 mL reactors, mechanically stirred and heated, and CO2 removal units. Substrate and inoculum were mixed at a feed to inoculum ratio (F/I) of 0.5 and 1, on volatile solids (VS) basis. More experimental details in Gallipoli et al., 2020.

2.6 Acidogenic step with liquid extract

Two parallel 3 Liters semi-continuous fermenters were operated at 37°C, HRT of 4 d and pH = 6 (controlled with Na_2CO_3 2.7 M addition, when needed), and manually shaken several times a day. For the start-up of the fermentation tests, both reactors were filled with 3 L of acidogenic inoculum. Two acidogenic fermentation tests were carried out on untreated FW extract (organic loading rate OLR = 5 gCOD/$L_{reactor}$ d) and pretreated FW extract (OLR = 9 gCOD/$L_{reactor}$ d), respectively. Methane was measured online (μFlow, Bioprocess Control, Sweden), after the CO_2-fixing unit.

2.7 Anaerobic digestion of the residue

Two parallel 8 Liters semi-continuous anaerobic digesters (CSTR) were operated at 37°C, at hydraulic retention time (HRT) of 20 d and OLR = 1.6 gVS/$L_{reactor}$ d by feeding untreated and pretreated FW solid residues. For the start-up, each reactor was filled with methanogenic inoculum and then acclimated to FW for a period of 1 HRT. The produced biogas was collected and sent through CO_2 traps (NaOH 3M solutions) and to the methane detection units (μFlow, Bioprocess Control, Sweden) for gas volume measurement at T = 0°C and p = 1 atm.

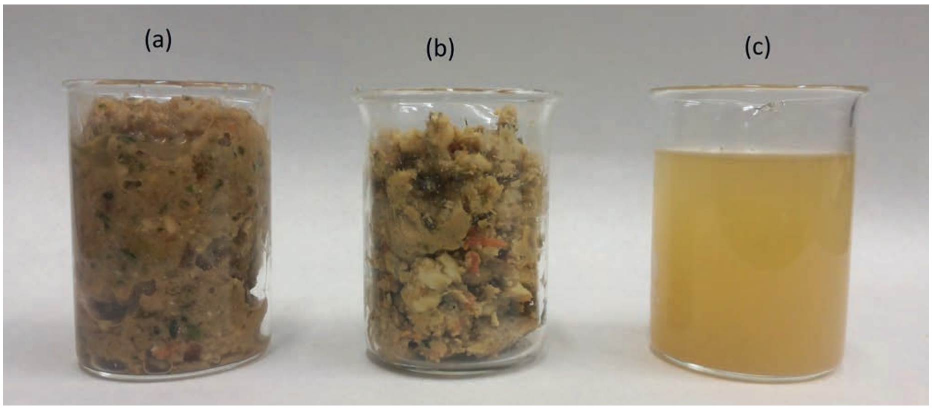

FIGURE 1: The Food waste (a), the solid residue (b) and the liquid extract (c) obtained after pretreatment and centrifugation.

3. RESULTS AND DISCUSSION

3.1 Characterization of household food waste

The FW characterization showed an average acidic pH of 4.5 ± 0.2, high content of water, due to the high percentage of vegetables, and high organic content (VS = 95±2%TS), highlighting high transformation potential of the substrate in bioprocesses as AD and fermentation. The organic matter was mapped in terms of proteins, carbohydrates, lignocellulosic components, lipids and Volatile Fatty Acids (VFAs). VFAs content in the fresh matrix was almost insignificant, accounting for not more than 2% of the total organic matter expressed as total COD, suggesting no pre-fermentation phenomena due probably to the rapid freezing storage after collection. Total carbohydrates accounted for 60%, while proteins and lipids fraction was 16% and 6% of the total COD, respectively. Soluble carbohydrates content (21% of COD) was high due probably to the high water content combined with the grinding phase acting as physical pretreatment. It is important to note that this type of FW presents also a lignocellulosic matrix (cellulose + hemicellulose + lignin) accounting for 19 ± 2% of the total solids, due to the high fraction of vegetable and fruit wastes (Pagliaccia et al., 2019).

The liquid (43%, w/w) and solid (57%, w/w) fraction deriving from raw food waste separation unit were characterized (Table 1). Most of the total solids, rich in organics (VS/TS=97%), remained in the residual fraction, with a total COD mainly in particulate form (~15% was soluble). As regards the liquid extract, all the COD was composed by soluble substances, in particular carbohydrates (around 70% expressed as COD).

3.2 Thermal pretreatment effect

The thermal pretreatment carried out with autoclave with an optimized duration of 20 min did not affect solids and organic content, but the inherent distribution between particle and soluble forms (Gianico et al., 2013). Pretreatment was effective in the solubilisation of the FW macromolecular fractions, resulting in a +57% increase in the soluble organics, both proteins and sugars. The pretreatment, despite the mild temperature and pressure conditions, was able to reduce the recalcitrant lignocellulosic matrix by breaking the interlinkages making more bioavailable the trapped compounds (Montecchio et al., 2017).

The main characteristics of liquid extract (83% w/w) and solid residue (17% w/w) deriving from the pretreated, diluted, FW are reported in Table 1. It is important to note that the thermal pretreatment resulted highly efficient in transferring the colloidal fraction of COD into the extract. The total COD of the raw extract was, in fact, totally constituted by soluble COD, while the pretreated one presented a noticeable fraction (around 30%) of sovra-colloidal COD, probably due to starch solubilization. Moreover, after the pretreatment, the residue was depleted by a significant fraction of solids, transferred into the liquid extract.

3.3 Effect of thermal pretreatment on acidification stage

In order to assess the potential of both raw and thermal pretreated FW to be fermented and/or to produce CH_4, different batch tests were performed at different F/I ratios (feed to inoculum ratio in terms of organics content) using an anaerobic inoculum from full-scale digester treating sludge containing also methanogens. Thermal pretreatment reduced the inert fraction of the FW from 22 to 17%, enhancing biodegradability. All tests reported a fast carbohydrate consumption: independently from the initial concentration, most of the soluble carbohydrates in the reactors was easily transformed into VFAs and hydrogen. Nevertheless, just after one day, operating the reactors at higher load (i.e. F/I = 1), dramatic VFA accumulation (mostly butyrate) took place, due to the fermentation of 70% of the bioavailable soluble carbohydrates.

The liquid extracts obtained from the solid-liquid separation unit (see Table 1) have been characterized in terms of COD before the acidogenic step, and appropriately diluted (only in the case of the untreated extract) in order to achieve the desired OLR.

A preliminary study on fermentation performances of the liquid "extract" deriving from raw and pretreated FW was performed in semi-continuous mode. Acidogenic fermentation was evaluated in terms of the amounts of VFAs produced, VFA composition, and fate of the carbohydrates. Promising fermentation yields of VFAs (from acetic to caproic acid) of 0.37 ± 0.3 g/gVS_{fed} and 0.49 ± 0.4 g/gVS_{fed} were obtained after 3 HRTs of operation, with raw and pretreated feed respectively, with comparable acidification degree (COD_{VFA}/sCOD) of 85 ± 2%. It was interesting to note

TABLE 1: Characteristics of liquid extract and solid residue of raw and thermal pretreated FW.

	Raw FW		Pretreated FW	
	Liquid extract	**Solid residue**	**Liquid extract ***	**Solid residue**
pH	5.1 ± 0.2	4.4 ± 0.2	4.7 ± 0.1	4.7 ± 0.3
TS (g/L)	68.8 ± 2.5	285 ± 14	28.0 ± 1.3	180 ± 12
VS (g/L)	58.5 ± 2.0	276 ± 10	25.2 ± 1.2	175 ± 10
COD (g/L)	75.7 ± 6.4	288.9 ± 29	34.1 ± 4.6	194.1 ± 27
sCOD (g/L)	72.6 ± 3.6	42.3 ± 6.8	25.6 ± 2.6	46.1 ± 9.2
Soluble Proteins (g/L)	18.4 ± 2.6	11.2 ± 1.7	3.8 ± 0.5	11.3 ± 1.5
Soluble Carbohydrates (g/L)	45.6 ± 4.4	23.2 ± 3.2	10.8 ± 2.7	24.5 ± 4.9

** Before thermal pretreatment, the FW sample was mixed with tap water (1:4 w/w)* that caproic acid was produced during both tests without external donor addition, and the pretreatment seemed to favor the chain elongation pathway. The yield observed at quasi-steady state for the pretreated FW extract was 0.08 gCaproate/gVS$_{fed}$ with respect to 0.06 gCaproate/gVS$_{fed}$ of the untreated one. VFA composition did not vary significantly between the two tests, predominant was always acetate followed by butyrate and propionate. However, in the case of the untreated extract, valerate was 17.6% against 7.8% of the pretreated (average quasi-steady state values), while the caproate fraction was comparable around 14% (Figure 2).

Undissociated acid concentration increase caused toxicity phenomena to reactor microbiome that controlled itself by not producing more caproic (see HRT 7, Figure 2) and kept the inhibition down despite the substrates availability. A continuous product extraction would address this limitation (Crognale et al., 2021; Kannengiesser et al., 2018).

3.4 Effect of thermal pretreatment on methane production

The effect of thermal pretreatment on biogas production rate of this food waste was previously assessed by single-stage anaerobic batch tests (Gallipoli et al., 2020). After an initial hydrogen production rate of 10 and 27 mL H_2/g VS$_{fed}$, the methane yield was statistically comparable, namely 0.38±0.014 and 0.37±0.010 mL CH_4/gVS$_{fed}$, for raw and thermal pretreated FW, respectively.

The performance of the parallel digesters fed with untreated and pretreated solid residue was monitored under the same OLR of 1.6 g VS/L.d and HRT of 20d. In particular, Figure 3 shows the cumulative methane production along the entire operation period of 70 days, including the production of Saturdays and Sundays, when the feeding was stopped. It is important to note that after 2 weeks of start-up, the slope of the curve remained almost constant (r^2 of 0.99) with 2.6 L/d and 2.8 L/d for untreated and pretreated residue, respectively, because of the remarkable methane production during the weekend despite the absence of feeding. It is therefore very likely that, because of the stop feeding, microorganisms had time to degrade and transform the bio-available cellulose and hemicellulose fraction of the residue into methane. Moreover, the thermal pretreatment likely undermined the lignocellulosic matrix, having a positive impact on the anaerobic conversion that was 0.31±0.01 against 0.26±0.01 L CH_4/gVS$_{fed}$ of the untreated one.

The typical instability problem due to acidification and methane drop observed just after the first HRT by feeding food waste (Tonanzi et al., 2018; Tonanzi et al., 2020), in this case was overwhelmed probably because of the lack of soluble bio-available COD (Figure 3).

4. CONCLUSIONS

Food waste is currently a significant environmental problem that needs to be properly tackled. In this Project, its conversion to added-value products and energy within a biorefinery perspective fits well in the Circular Economy approach as it promotes a more efficient utilization of renewable raw materials, contributing to the goals of the European Green Deal (i.e to strive for greener industry and to produce clean energy).The integration of a mild thermal pretreatment assured an efficient organics solubilization (in particular sugars) into the liquid phase favoring the transformation and the conversion rates into volatile fatty acids through the optimization of operational pH and HRT of the primary fermentation step.

Secondary fermentation reaction as chain elongation to MCFAs (as caproic acid) occurred in the same reactor without external donor addiction probably because of the in situ formation of lactate. The innovative technological components of the proposed biorefinery will also include the use of magnetic nanoparticles to recover the most val-

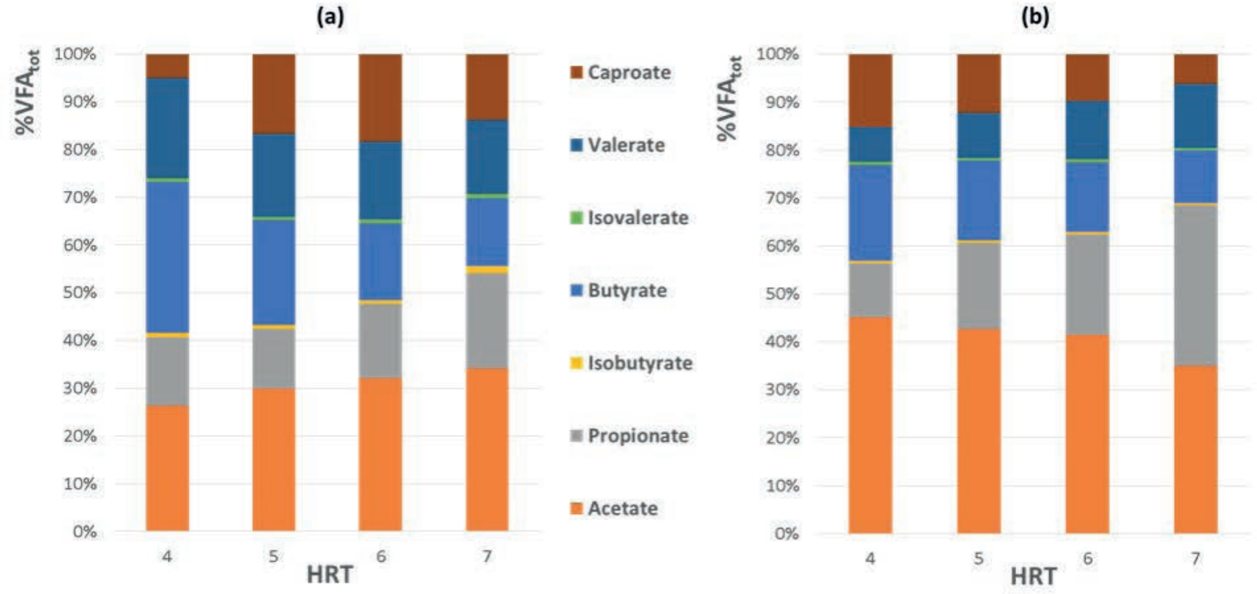

FIGURE 2: VFAs distribution of untreated (a) and pretreated (b) extract, at quasi-steady state conditions.

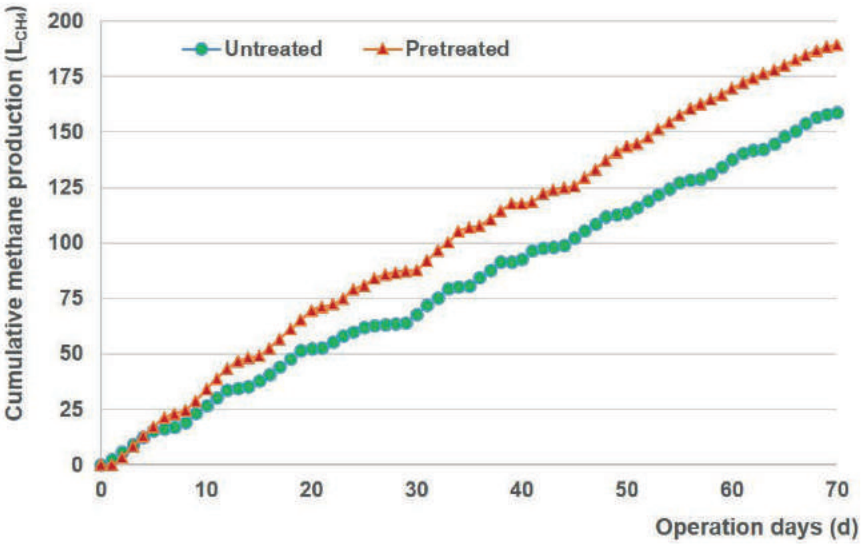

FIGURE 3: CH_4 production of untreated and pretreated solid residues during the long-term AD process (HRT 20d).

uable fatty acids for successive platform chemicals production.

The solid residue fed into the anaerobic digesters assured process stability (no acidification phenomena occurred) despite the short HRT of 20d. The weakening of the lignocellulosic matrix by the thermal pre-treatment had a positive impact on the anaerobic conversion increasing significantly methane production rate in particular during weekend, when feeding was stopped. During the Project, assessment of the hygienic properties of digestate (in terms of Salmonella and E.coli concentration) will be performed to assure a "safe" digestate to be recycled in agriculture, drastically reducing environmental waste impact, one of the prior societal challenges in many part of the world.

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A BRIEF OVERVIEW ON VALORIZATION OF INDUSTRIAL TOMATO BY-PRODUCTS USING THE BIOREFINERY CASCADE APPROACH

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ABSTRACT

The industrial processing of tomato leads to substantial amounts of residues, typically known as tomato pomace or by-products, which can represent as much as 10% by weight of fresh tomatoes. At present, these residues are either used as feedstock for animals or, in the worst case, disposed of in landfills. This represents a significant waste because tomato pomace contains high-value compounds like lycopene, a powerful antioxidant, cutin, which can be used as a starting material for biopolymers, and pectin, a gelling agent. This article presents an overview of technologies that valorize tomato by-products by recovering added-value compounds as well as generating fuel for energy production. These technologies include operations for extraction, separation, and exploitation of lycopene, cutin and pectin, as well as the processes for conversion of the solid residues to fuels. Data collected from the review has been used to develop a biorefinery scheme with the related mass flow balance, for a scenario involving the tomato supply chain of Regione Campania in Italy, using tomato by-products as feedstock.

1. INTRODUCTION

Tomatoes (Lycopersicon esculentum Mill.) are one of the major vegetables in terms of world production, being second to potatoes only. In 2017, the global production of tomatoes amounted to 182 million tons, with China accounting for 32% of the total, followed by the European Union, India, United States and Turkey as the major producers; Italy is one of the largest producers among European nations, and the industries of Regione Campania are leaders in the sector (FAO, 2019).

While most harvested tomatoes are sold fresh, about a third is processed for traditional products like canned tomatoes, juice, paste or puree, sauces, and ketchup. The processing of tomatoes generates substantial amounts of residues, known as tomato pomace or by-products, representing 2-10% of the whole fresh tomato (Gustavsson et al., 2011; Ventura et al., 2009). Tomato pomace is a mixture of tomato cuticle (or peel), seeds and small amounts of pulp that remain after processing (Ventura et al., 2009). It is often used in animal feed (Mizael et al., 2020) as a source of dietary fiber, as well as B vitamins, lycopene and to a lesser extent vitamin A, for example, in California (which accounts for 95% of US production) most of the pomace is added to cattle feed for dairy farms. The alternative to use in animal feed is landfilling; this, for instance, represents the prevailing end-of-life route in the US Midwest. The use of tomato pomace as soil amendment after composting denotes an additional, but less common alternative (Kakabouki et al., 2021). In Italy, tomato pomace is either given for free to farmers or is sent to landfill, which represents an added cost of 200 euros per ton for the producing companies. Overall, the use of tomato pomace is limited by the risk of ruminal acidosis caused by pomace acidity (Mizael et al., 2020) or the high moisture content (60-80% wt.) leading to fast spoiling and pollution hazard.

Tomato pomace is rich in nutrients and could be used as a potential source of fiber (60-70% on dry basis), protein (10-20% on dry basis), antioxidants or fat (5-10% on dry basis) (Del Valle et al., 2006, Brachi et al., 2016a). Table 1 reports the average composition of tomato pomace including the organic macro-components.

A recent study (Benítez et al., 2018) demonstrated that cutin is the main component of the peel fraction of tomato pomace; it is a non-toxic, biodegradable, water-

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proof, UV-blocking, amorphous, insoluble and infusible bio polyester made of esterified C16 and C18 hydroxy acids (Benítez et al., 2018). Del Valle et al. (2006) reported that tomato pomace is also a source of natural pectin (8% wt. on dry basis), a thickening agent used in the food industry. Knoblich et al. (2005) investigated the tomato peel composition and proved its potential use as a source of carotenoids, natural pigments, due to its health-beneficial properties. Their results, reported in Table 2, showed that lycopene is the most abundant carotenoid in tomato processing by-products.

Tomato seeds are a specific by-product of de-seeded canned tomatoes production. They are separated from the residual tomato pomace as well as from peels by floatation. The vegetable oil contained in seeds (i.e., 20-25% wt. on dry basis) can be recovered, for example by crushing in expellers. Tomato oil is brown in color with a strong smell and is considered suitable for dressing salads (Giuffrè and Capocasale, 2016). It contains saturated fatty acids up to 14-18% wt., and unsaturated fatty acids up to 76-80% wt. (Giuffrè and Capocasale, 2016). Its average composition is reported in Table 3.

The considerable number of high-value compounds in tomato pomace, advocates for the development of a strategy for their exploitation. Given that tomato processing by-products are classified as lignocellulosic biomass, they could be exploited through thermochemical, biological, and chemical conversion to obtain biogenic fuels, and then electricity and heat. For example, it has been reported that peels and seeds are suitable for combustion processes indeed their volatile matter and HHV (higher heating value) are significantly high after drying, which make them an interesting source for thermal energy production (Khiari et al., 2019) . Moreover, the high content of fatty acids makes tomato seed oil a good candidate to produce biodiesel (Giuffrè et al., 2016).

This article presents a brief review on the state of the art of the available techniques for the valorization of tomato by-products, in terms of extraction of high value compounds and production of energy and fuel. The review will be used as starting point for the development of a biorefinery model based on tomato pomace as feedstock and the evaluation of the related mass flow balances.

2. EXTRACTION AND APPLICATION OF HIGH VALUE COMPOUNDS

2.1 Lycopene properties and extraction

Lycopene (from the neo-Latin Lycopersicum, the tomato species) is a bright red carotenoid found in tomatoes and other red fruits, vegetables (red carrots, watermelons, gac, and papayas) and phytosynthetic algae. Due to the strong color and its solubility in organic matters, lycopene is useful as food coloring (registered as E160d) and is approved for use in the USA by the US Food and Drug Administration since 2005.

Lycopene has also been studied for its potential health effects. Promising data from epidemiological as well as cell culture and animal, studies suggest that the consumption of lycopene-containing foods may improve human health. To this end, several advanced drug delivery systems have been developed, to enhance the in vivo delivery of lycopene (Caseiro et al., 2020; Kaur et al., 2017).

In recent years, tomato peels have been proposed as a low-cost source of lycopene, compared to fresh tomatoes or Blakeslea trispora, a fungus of the division of Zygomycota, industrially used due to its ability to produce carotenoids (Górecka et al., 2020; Martínez-Cámara et al., 2018). As for the extraction of carotenoids from other plant materials, solvent extraction and supercritical CO_2 extraction are the most studied and optimized techniques for lycopene extraction from tomato residues (Fritsch et al., 2017). There are numerous variables that can influence the yield of lycopene extraction, but the solvent type is widely considered to be the most important (Kaur et al., 2008). Organic solvents and their mixtures are the most investigated due to their affinity with lycopene (Briones-Labarca et al., 2019; Pandya, 2017; Zuorro, 2020).

The EU allows a small number of solvents when lycopene is to be used within the food industry (including propanol, hexane, acetone, ethanol, methanol and ethyl acetate) and a maximum solvent residue of 50 mg per kg of lycopene (Commission Directive 2011/3/EU, 2011). The most promising environment-friendly alternative to traditional solvents is limonene, the major component in the essential oil of citrus fruit peels (Shakir Salih, 2015) and typically used in food industry as additive (Kim et al., 2013;

TABLE 1: Typical composition of tomato pomace on dry basis.

Pomace fraction	Content (% w/w)	Components	Water Content (%)	References
Pulp	0-15	Cellulose, hemicellulose, lignin	≈95	(Al-Wandawi et al., 1985; Benítez et al., 2018; Liadakis et al., 1995; Silva et al., 2019)
Peels	30-60	Cutin, pectin, hemicellulose, cellulose, extractives	≈80	
Seeds	35-55	Hemicellulose, cellulose, lignin, lipids	≈60	

TABLE 2: Carotenoid content of tomato peels on dry basis according to Knoblich et al. (2005)

Carotenoid	Lycopene	Lutein	Zeaxanthin	β-Carotene	cis- β-Carotene
Content (μg/g)	734	14.5	3.7	29.3	11.7

TABLE 3: Tomato seeds oil composition on dry basis according to Giuffrè and Capocasale (2016).

Fatty acids	Stearic acid	Oleic acid	Linoleic acid	Linolenic acid	Others
Content (% wt.)	20	25	50	2-3	2-3

Ravichandran et al., 2018). Limonene, obtained from orange peels, has been also tested for lycopene extraction with results comparable to conventional organic solvents (Chemat-Djenni et al., 2010).

Solvent extraction has usually low yields due to the complex structure of the cell walls where the lycopene is trapped; moreover, degradation of the carotenoids can occur due to high temperature and long extraction time, reducing the lycopene extract quality. To overcome this problem, several techniques have been proposed in the literature, including: sonication-assisted extraction (Yilmaz et al., 2017, Rahimi and Mikani, 2019), microwave-assisted extraction (Ho et al., 2015), enzyme-assisted extraction (Ranveer, 2013; Catalkaya and Kahveci, 2019) and Naviglio extraction (Naviglio et al., 2008).

The commercial alternative to solvent extraction is supercritical fluid extraction (SFE) with CO_2 being the most common working fluid. Studies on the extraction of lycopene by the means of SFE show that increasing the density of CO_2 with a co-solvent (hexane or ethanol), leads to higher solubilization of the carotenoids in the fluid and, thus to an improved extraction (Baysal et al., 2000; Cadoni et al., 1999). However, recent works demonstrated that the supercritical extraction of lycopene without the use of co-solvent can reach high yields when operational parameters are optimized (Pellicanò et al., 2020). The main advantages of SFE are the high target specificity, the short extraction times, the use of a non-toxic solvent and a reduced environmental impact (Wang and Weller, 2006). On the other hand, disadvantages could be represented by SFE higher operating costs, since high pressures must be applied to maintain the fluid in supercritical state, compared to the less energy demanding operational conditions of solvent extraction. The results together with the experimental conditions of the most recent works on the extraction of lycopene from tomato peels are reported in Table 4.

The extraction processes, mentioned above, produce a fat-soluble oleoresin that contains a high concentration of lycopene and all the hydrophobic compounds that were included in the pomace; oleoresin accounts for around 5% of tomato peels on dry weight (Brachi et al., 2016a). It is already available on the market and can be sold as produced or incorporated in products as tablets, capsules, soft gels, powders, and drinks (Nagarajan et al., 2017).

2.2 Cutin extraction and application

Cutin is the polymeric building block of the plant cuticle. It represents 40–80% wt. of the dry peels and consists of esterified fatty acids (Domínguez et al., 2015; Heredia, 2003). Cutin is mainly composed of mixture of C16 and C18fatty acids (Domínguez et al., 2011). These long-chain fatty acids (called cutin acids) represent innovative building-block chemicals for the synthesis of novel bio-resins and lacquers suitable as internal protective coating for metal food packaging. However, these natural compounds are not currently available commercially (Cifarelli et al., 2019). Tomato pomace and tomato peels have been proposed as a renewable source of this biopolymer, due to their high content in cutin and their availability. Cifarelli et al. (2019) reported three efficient, easy and environmentally safe procedures that could be commercialized for the extraction of cutin acids from tomato peels without the use of organic solvents, these include: i) alkaline hydrolysis of the tomato peel, ii) acid-free selective precipitation of cutin and iii) hydrogen peroxide-assisted hydrolysis. Notably, those authors noticed that the products were different depending on the method used in terms of appearance, solubility, degree of observed crosslinking and molecular weight. They

TABLE 4: Main experimental results for lycopene extraction from tomato pomace.

Technology	Experimental conditions	Extraction solvents	Yield of extraction [on 100 g of peels]	Reference
Solvent extraction	40°C for 5h and 1:30 feed to solvent ratio	50% acetone 50% ethyl acetate	7.91 mg	(Pandya, 2017)
	40°C for 30 min and 1:30 feed to solvent ratio	30.6% hexane, 32.8% ethanol 36.6% acetone	0.34 mg	(Zuorro,2020)
	RT and 1:1 feed to solvent ratio	d-Limonene	2.44 mg	(Chemat-Djenni et al.,2010)
	High pressure pre-treatment and RT for 24 hour and 1:2 feed to solvent ratio	60% hexane 40% ethanol	2.08 mg	(Briones-Labarca et al., 2019)
Ultrasound assisted extraction	90 W for 30 min and 1:35 feed to solvent ratio	50% hexane, 25% ethanol 25% acetone	9.4 mg	(Yilmaz et al., 2017)
	70 W for 10 min and 1:20 feed to solvent ratio	Sunflower oil	9.1 mg	(Rahimi and Mikani, 2019)
Microwave assisted extraction	400 W for 1 min and 1:20 feed to solvent ratio	Ethyl acetate	13.6 mg	(Ho et al., 2015)
Enzyme aided extraction	Enzymatic treatment with 2% pectinase at 4 h of incubation period and subsequent solvent extraction	50% hexane, 25% ethanol 25% acetone	20 mg	(Ranveer, 2013)
	Enzymatic treatment with a combination of cellulolytic and pectinolytic enzymes at 5 h of incubation period and subsequent solvent extraction	Ethyl acetate	N.A.	(Catalkaya and Kahveci, 2019)
Supercritical fluid extraction	80 min with a pressure of 550 bar at 60°C and 2ml/min of solvent flow	CO_2	37 mg	(Pellicanò et al., 2020)

also noted that cutin obtained through alkaline hydrolysis resulted the best raw material for bio-resin preparation (Cifarelli et al., 2019). Manrich et al. (2017) proposed a hydrophobic edible film consisting of tomato cutin and pectin, obtained using extraction of cutin using the procedure proposed by Cigognini et al. (2015). The procedure consisted in immersing dried peels in a solution of NaOH and then autoclaving at 121°C. The liquid phase was collected by filtration and then acidified to precipitate cutin, which was recovered by centrifugation, washed, and freeze-dried with a yield of 25% of dried tomato peels. Pure cutin did not lead to free-standing film and, therefore, pectin was added as a ligand to produce water-resistant tomato cutin-based films (Manrich et al., 2017). Benitez et al. (2018) proposed the production of a cutin-based polyester by melt-polycondensation without catalyst, after cutin extraction and depolymerization. Even if cutin-based resins still must be optimized, they are promising alternatives to commercial polymers used as coating for food packaging and represent a viable way to re-use industrial tomato by-products.

2.3 Pectin extraction and application

Pectin is a well-known, naturally occurring biopolymer that is finding increasing applications in the pharmaceutical and biotechnology industry. It has been successfully used for many years in the food and beverage industry as a thickening medium, a gelling agent, and a colloidal stabilizer (Commission Regulation (EU) No 231/2012, 2012). Moreover, pectin has several unique properties that have enabled it to be used as a matrix for the entrapment and delivery of a variety of drugs, proteins, and cells (Sriamornsak, 2003).

Although pectin is found in most plant tissues, the number of sources that may be used for the commercial manufacture of pectins are very limited. At present, commercial pectin is almost exclusively derived from citrus peels or apple pomace as by-products from juice manufacturing. The process involves extracting pectin via treatment of the pomace with hot dilute mineral acid; pectin is then recovered by precipitation with alcohol (May, 1990; Yapo et al., 2007). The main drawback of this process is represented by mineral acids, which are toxic and generate environmentally problematic effluents (Yapo, 2009). A potential alternative to mineral acids is represented by citric acid, which has been tested for pectin extraction with comparable results (Pereira et al., 2016, Cho et al., 2019). Adiletta et al. (2019) extracted and studied pectin contained in sugar beet processing residues. They proposed for the first time the valorization of sugar beet pulp to value-added chemicals and fuels by coupling the eco-friendly isolation of pectins via citric acid extraction with the upgrading of the residual pectin-free solid as a biofuel through torrefaction.

Del Valle et al. (2006) reported that tomato peels contain pectin at 8% wt. on dry basis. Grassino et al. (2016) developed a method to produce pectin from tomato peels; in their experiments, pectin was extracted from dried tomato peels using ammonium oxalate and oxalic acid as extracting solvents, in two steps. According to their results, it can be concluded that tomato peels are a suitable source for pectin that can be used to produce corrosion inhibitors and a valuable additive in food industry. Alancay et al. (2017) optimized the pectin extraction from tomato processing waste by using a mineral acid, i.e. HCl, thus obtaining a maximum yield of 280 g/kg of tomato pomace. The main results from pectin extraction experiments starting from different biomasses are reported in Table 5.

TABLE 5: Main experimental results for pectin extraction from different sources.

Source of pectin	Extraction acid	Yield of extraction	Reference
Sugar beet pulp (dried)	Sulfuric acid	16%	(Yapo et al., 2007)
Yellow passion fruit by-product (dried)	Lemon juice concentrate	10%	(Yapo, 2009)
Pomegranate peels (dried)	Citric acid	8%	(Pereira et al.; 2016)
Sugar beet pulp (dried)	Citric acid	25%	(Adiletta et al., 2019)
Apple peels (dried)	Tartaric acid	7%	(Cho et al., 2019)
Tomato peels(dried)	Ammonium oxalate and oxalic acid	32%	(Grassino et al. 2016)
Tomato peels (dried)	Hydrochloric acid	28%	(Alancay et al., 2017)

3. TOMATO RESIDUES TO ENERGY

3.1 Characterization, torrefaction and pelletization

Mangut et al. (2006) provided ultimate analysis, proximate analysis, and higher heating value (HHV) of tomato pomace, after drying, with a residual moisture content around 10% wt. Their results show that the amount of sulfur is lower compared with that found in convectional fossil fuels. This outcome is interesting from environmental and technical standpoints because sulfur is well-known to generate important atmospheric pollutants and to negatively affect process plant components upon combustion. Mangut and coworkers also stated that with an HHV value of around 22 MJ/kg, tomato by-products (peels, seeds and pulp) represent an interesting energy source with a high potential for heat and electricity production (Mangut et al., 2006).

Rossini et al. (2013) focused on the characterization of the tomato manufacturing residues finalized to the energy recovery. In their study they reported the physical-chemical properties of tomato by-products, and their component parts, and highlighted that nitrogen and chlorine content are considerable, especially in the seeds, this is undesirable in combustion system due to corrosion of plant components as well as serious environmental problems. To this end, they suggested to use tomato peels for combustion and seeds for the production of vegetable oil.

Brachi et al. (2016b) carried out a comprehensive study on torrefaction of dried tomato peels in a fluidized bed of inert particles, which included identification of key performance parameters and development of an experimental procedure to determine their values. Their results indicate that tomato peels are a good candidate for the torrefaction treatment. The authors also observed that higher temperatures and longer holding times (with a more marked effect of the torrefaction temperature) led to an increase in the calorific value of the torrefied tomato peels, with respect to untreated peels. For istance, when pomace is thermally treated for 30 min at 285°C the calorific value is increased by a factor of 1.2 for the torrefied biomass, with a 40% reduction in the O/C elemental ratio and an improved hydrophobicity. These positive effects of the torrefaction treatment occurred while maintaining the mass yield (approximately between ~75% and ~94%) and energy yield at satisfactory levels. The authors also demonstrated that the fixed bed torrefaction does not ensure a consistent quality of the torrefied solid product and, consequently, a reliable determination of the key process performance parameters.

Ruiz Celma et al. (2012) investigated the feasibility of pelletizing tomato by-products for use as solid fuels. They produced fuel pellets by forcing the feed product through 6 mm diameter nozzles in a matrix pattern, after previous milling and air-drying process, carried out at 45°C drying air temperature and 1.3 m/s drying air velocity. Their pellets had a HHV of 20 MJ/kg, which is comparable to that estimated by Mangut et al. (2006) and to that of commercial wood pellets (Telmo and Lousada, 2011).

3.2 Chemical conversion of tomato seed oil

The use of vegetable oil for the production of biodiesel, a renewable source of energy, has multiple advantages: first, a reduction in the dependency on fossil fuels for energy production as well as a reduction of vegetal wastes; second, an increase of the economic value of crops and of vegetable oils; and third, a reduction of carbon emissions.

Giuffrè et al. (2016) firstly suggested that tomato seeds could be used for biodiesel production. The possibility of extracting oil from tomato seeds was already considered in the early 20th century. Seeds are obtained from pomace by sedimentation and pressed or extracted with solvent to produce oil, which can be refined using alkalis and then clarified with fuller's earth. The resulting oil is pale yellow and considered suitable for dressing salads. Giuffrè et al. (2016) reported that the physicochemical properties of tomato seed oil are comparable with those of rapeseed oil, that is currently used for biodiesel production in Europe. Giuffrè et al. (2017) reported a method to synthetize biodiesel from tomato seed oil. The transformation of a vegetable oil into biodiesel occurs via transesterification in a chemical reactor, in which the oil is mixed with an alcohol in the presence of a catalyst and heated. The most common alcohol used for transesterification is methanol, with potassium hydroxide being the typical catalyst. Glycerin is produced as by-products. This study showed that the resulting biodiesel can meet European regulations. In particular, the biodiesel with the best yield and composition was obtained at trans-esterification conditions with a temperature of 55°C, a reaction time of 1 h, an oil/methanol ratio 1/6 and a catalyst concentration of 1% wt. in oil.

4. BIOREFINERY MODEL AND MASS BALANCES

The previous sections demonstrate the significant value of tomato by-products and the availability of techniques for their valorization. In light of this, a new biorefinery model was developed for the sustainable co-production of fuel and chemicals from tomato pomace, with minimal generation of wastes. The model follows the biorefinery cascade approach (Keegan et al., 2013) and is sketched as a block diagram in Figure 1. Notably, Figure 1 only includes the main unit operations that are associated with each product.

The model assumes that tomato pomace is only composed of peels and seeds. These components are separated via floatation-cum-sedimentation and sent to different exploitation pathways. On the one hand, seeds are cold pressed with the resulting oil collected and purified. The oil is then sent to a transesterification section to produce biodiesel that meets European regulations via catalytic reaction in methanol media.

On the other hand, peels undergo several process steps in series for their complete exploitation. First, lycopene oleoresin is extracted using a mixture of organic solvents; the oleoresin is collected, purified, and used for preparing tablets that are sold as dietary supplement. Second, pectin is separated by oxalate acid extraction, collected by ethanol precipitation and washed to meet the appropriate purity for use in the food industry. Third, cutin is extracted from the solid residue, via alkaline hydrolysis; a cutin-based polyester is then produced via melt-polycondensation. The final solid residue, composed by seed residues and exhausted peels is dried, sent to a pelletizer and then torrefied to produce pellets with an HHV of around 20 MJ/kg.

A general mass balance of the biorefinery model was carried out using data from literature, as reported in previous sections, including feedstock characteristics and processes yields. The mass balance shows that from 100 ton of tomato pomace on wet basis, it is possible to extract 0.6 ton of lycopene-based oleoresin, and 3 ton of cutin and 1 ton of pectin from tomato peels, whilst producing 3.9 ton of biodiesel and 0.4 ton of glycerin from tomato seed oil and methanol. In addition, it is possible to send 91.4 ton of biomass to thermal treatment (drying, torrefaction and pelletizing), producing 18.2 ton of pellets and generating 73.2 ton of emission composed mainly by removed water and gas produced during torrefaction. The results obtained from the mass balance are shown in the Sankey diagram in Figure 2.

Finally, Table 6 shows an application of the biorefinery model to the Campania region in Southern Italy, which processes nearly half of all Italian tomatoes; this corresponds to about 2.2 Mtons of fresh fruits transformed every year (Tomato News, 2019) and leads to a yearly availability of tomato by-products of around 110 kton. The table illustrates the potential production capacity of a biorefinery plant based in the Campania region. Such a plant could produce

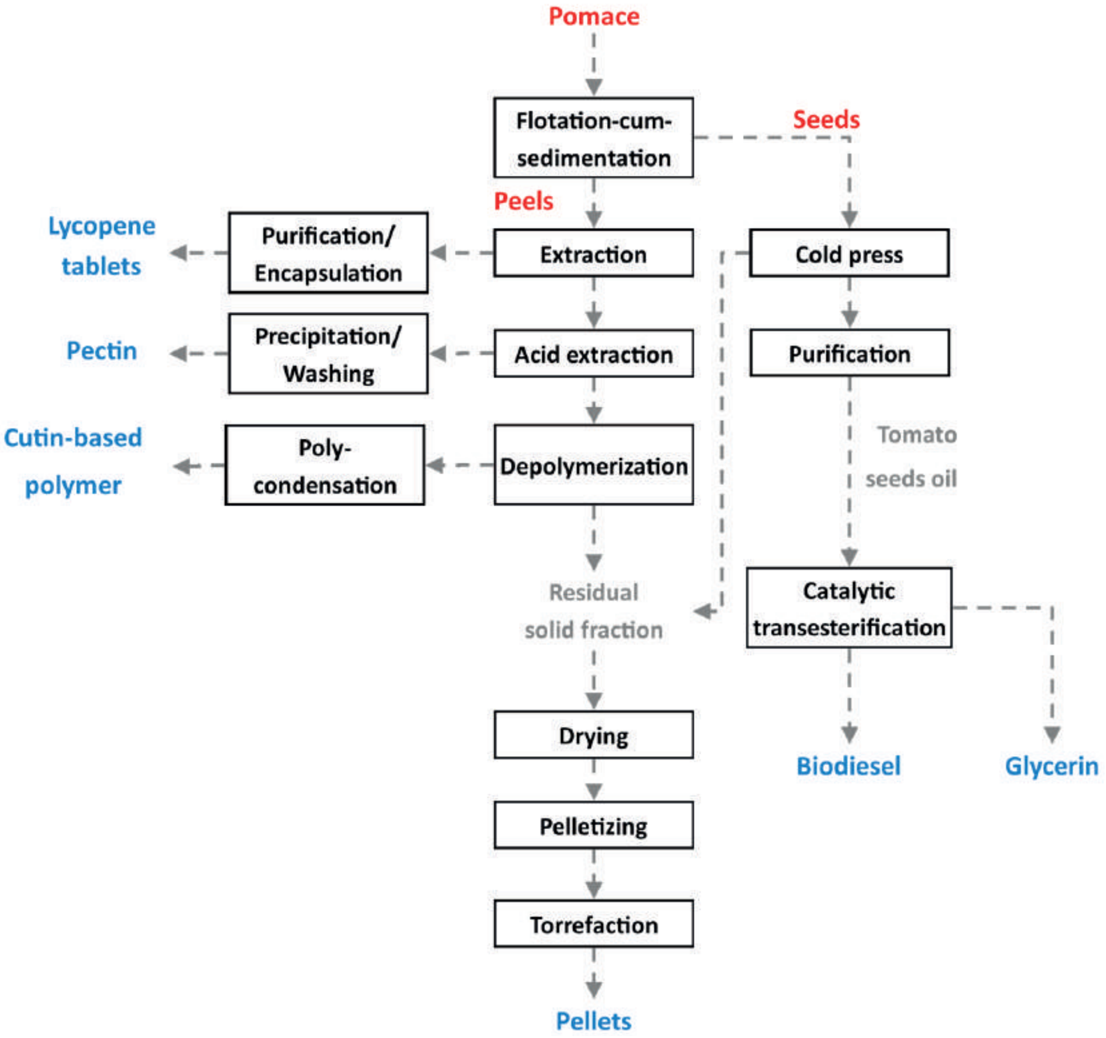

FIGURE 1: Biorefiney model for a full exploitation of tomato processing by-products (feedstocks are reported in red font, process units in black, intermediate materials in grey and biorefinery products in blue).

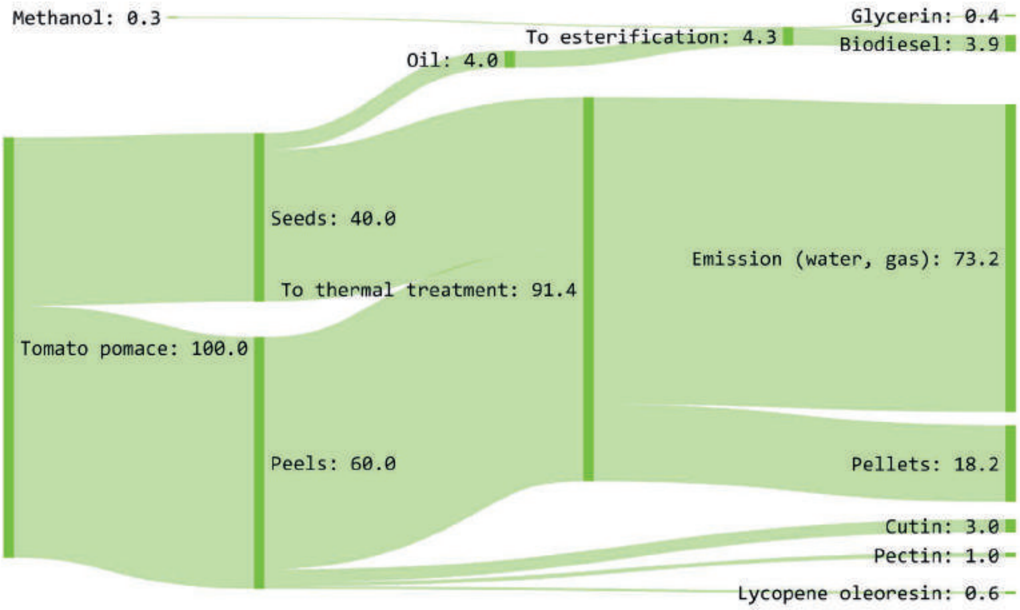

FIGURE 2: Mass balance for tomato by-products in tonnes.

up to 1.02 kton of pectin, which covers about 3% of the European demand (Pectin Market | Apple Pectin, Fruit Pectin, Citrus Pectin, 2019) and 0.41 kton of glycerin, that is 3% of the amount of glycerin, natural and synthetic, produced in Campania in 2019 (ISTAT, 2020). With respect to the fuel and energy sector, a Campania-based biorefinery plant could contribute with 20.35 kton of pellets, i.e.16% of all wood pellets consumption in Campania in the last 5 years (ISTAT), and with 4800 m^3 of biodiesel, i.e. 3 times the Italian demand of recent years (Statista - The Statistics Portal, 2020). Moreover, such a plant could supply all Italian population with lycopene tablets during cold months, ensuring the daily requirement of 10 mg of lycopene when fresh tomatoes are not available (Story et al., 2010).

5. CONCLUSIONS

This article aimed at contributing to the development and implementation of technologies for the exploitation of industrial tomato by-products that are technically feasible, economically convenient, and environmentally friendly. The article presented a thorough review of the technologies that have been proposed in the literature to valorize industrial tomato by-products, which are under-exploited and sometimes even landfilled. The review revealed that tomato pomace has high concentration of exploitable compounds: lycopene, for dietary supplement production, cutin for food packaging application, pectin as gelling agent in food industry, and tomato seed oil for biodiesel and glycerin production. Techniques to extract and purify these compounds have been demonstrated and optimized at various scales as such, they can be feasibly deployed in new process plants using tomato pomace as a feedstock. Moreover, due to their thermochemical properties, the solid residues after extraction steps, can be used for the production of pellets, after torrefaction.

Using information gathered from the literature review, it has been possible to develop a biorefinery scheme based on the valorization of tomato by-products for the sustainable co-production of fuel and chemicals, with minimal generation of waste. Although, the biorefinery scheme has only been analyzed in term of mass balances, it represents a valid background for the modeling and optimization of a multi-product biorefinery for the production of lycopene-based tablet, cutin-based polymer, pectin, glycerin and fuels, thus creating adding value and recovering from wasting the large amount of tomato pomace that is produced every year in the Campania region. Therefore, tomato by-products can be turned from a puzzling issue into a useful resource for the Campania in the frame of a circular economy approach.

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TABLE 6: Production flow of biorefinery products with tomato pomace as feedstock (base case: 2.2 Mton/y of processed tomatoes in Campania).

Product	Amount (unit/year)	Application	Commercial alternatives
Tablet containing 10 mg of lycopene	6.6 10^9 pills	Dietary supplement for lycopene intake, provides antioxidant properties that help cells fight damaging free radicals in the body	Pills containing lycopene extracted from fruits
Pectin	1.02 kton	In the food and beverage industry as a thickening agent, a gelling agent and a colloidal stabilizer	Pectin extracted from citrus peels
Cutin based polymer	3.7 kton	In the food packaging field	Polyester
Biodiesel	4.8 10^3 m^3	In pure form or blended with petroleum diesel at any concentration in most injection pump diesel engines	Biodiesel from vegetable oil
Glycerin	0.41 kton	It is also widely used as a sweetener in the food industry and as a humectant in pharmaceutical formulations	Generally obtained from plant and animal sources
Pellets	20.3 kton	Energy source with a high potential for heat and electricity production	Wood pellets

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MINERALOGICAL AND CHEMICAL CHARACTERIZATION OF CDW AS FUNCTION OF PARTICLE SIZE AND THERMAL TREATMENTS FOR POTENTIAL RECYCLING

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ABSTRACT

Construction and Demolition Waste (CDW) originating from the rubble produced by the 2016 seismic events in the Marche Region (Central Italy) has been studied, focusing on its mineralogical and chemical characteristics, to investigate its recycling potentials as a component for eco-sustainable building material or in the glass industry. The aim was to obtain a full characterization of the behaviour of this material at high T in order to determine the most advantageous conditions for vitrification, considered as an effective process for volume reduction as well as for immobilization of potentially hazardous elements. Vitrification experiments, carried out with thermal treatments as function of temperature/duration/particle size and aimed at amorphization, were carried out under atmospheric conditions, at different temperatures (1000-1250°C) and durations (2-8 hours). The study demonstrated that mineralogical composition remains homogeneous for grainsize <4 mm, thus suggesting that no sieving is necessary for recycling of the fine fractions, which are the most difficult to treat. Vitrification, although not achieved for the CDW sample up to 1250°C, due to high-Ca and low-Si contents, demonstrated that this CDW can produce an interesting refractory material and a porous/insulating material. However, experiments showed that full vitrification can be easily achieved by mixing urban waste glass and CDW, suggesting applications in the glass industry. Based on the chemical and mineralogical features of the products, other significant upgrading alternatives of recycling the CDW in different fields of applications are highlighted.

1. INTRODUCTION

The EU Waste Framework Directive 2008/98/EC (European Commission, 2008) states that all EU members should take all necessary measures in order to achieve at least 70% of re-use, recycling or other recovery of non-hazardous Construction and Demolition Waste (CDW) by 2020 (Whittaker et al., 2019 and references therein). Construction and Demolition Waste (CDW), in fact, has been identified as a priority waste stream covering about the 30% of total waste generation in EU (approx. 800 million metric tons; European Commission, 2016a, 2016b).

The composition of CDW varies strongly between the different member states and geographical areas, making an uniform approach and recovery strategy difficult. CDW is, in fact, highly heterogeneous, depending on the geological availability of lithotypes, construction locations and building styles. Concrete, bricks, rocks, sanitary ware, ceramic tiles, roof tiles, plaster, wood, glass, metals, plastic as well as hazardous waste like asbestos, are commonly present in CDW. Often, the main option for recycling this waste type is still downgrading applications, as aggregates for road construction or backfilling (DG ENV, 2011) and, with limitations due to diminished mechanical performances, as recycled aggregates in concrete (Pacheco-Torgal et al., 2013 and references therein). Due to the low economic costs of landfilling, lack of incentives and sometimes dated regulations, still virgin aggregates are preferred over the recycled aggregates (Pacheco-Torgal and Jalali, 2011; Marie and Quiasrawi, 2012). However, a few studies already deal with the possible incorporation of CDW in new high-grade construction materials, as for example in terrazzo tiles (Favaretto et al.,2017; Ansaloni et al., 2017; Whittaker et al., 2019; Stabile et al., 2021), interesting for the environmental advantages in terms of saving of geo-resources and energy over ceramic tiles production.

It is noteworthy though that up-grading applications of CDW in the construction sector are still difficult to achieve. In 2019, the European Commission produced a

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comprehensive report entitled 'On the Implementation of the Circular Economy Action Plan' (European Commission, 2019; Whittaker et al., 2019). The report proposes the development of reliable strategies and new technologies to: a) increase the percentage of CDW-derived materials in new residential construction, b) increase the technical and economic value of CDW-derived materials, c) minimize the future production of CDW from the next generation of buildings, d) increase building energy efficiency. All these points should encourage studies in this field. In view of the overall latest environmental norms, a strong effort has been devoted to the elaboration of treatment processes in order to transform CDW into recycled eco-friendly products. Recent studies deal with different techniques able to meet these requirements, including solidification within concrete, leaching with acids and other solvents, sintering, and vitrification (Haugsten and Gustavson, 2000; Jung et al., 2005 and references therein). Vitrification as the preferred treatment is supported by studies that indicate it represents the most promising waste management technique that: a) produces a stable vitreous material, b) can provide chemical stability, c) immobilizes toxic elements (e.g. cadmium, nickel, etc.) in glass structure and d) decomposes dioxins (Kavouras et al., 2003; Jung et al., 2005; Stabile et al., 2019).

Here a full chemical and mineralogical characterization of CDW, before and after thermal treatments is presented, carried out to investigate the effects of composition, grainsize, experimental temperature and duration on the vitrification process. The aim of the work is also to explore possible alternatives to landfilling, investigating the recycling potential, based on compositional/mineralogical characterization of CDW and its derived products.

2. MATERIAL AND METHODS

The main methodological steps carried out in this study for the full characterization and thermal treatments of the CDW sample and products are summarized in Figure 1 and presented in details in the results section.

2.1 Sample collection and sorting

The CDW used in this study was obtained from the centralized processing plant (COSMARI, in Macerata, Italy) which, after the 2016 seismic crisis that hit four Regions in Central Italy, is in charge of collecting, separating and distributing all the earthquake rubble of the entire Marche Region. The CDW is preliminarily removed of health- or environmentally-concerned materials like cement-asbestos, lead and electronic waste, as well as of all other recyclable metals. Wood, plastic, glass, gypsum are separated, while large rock blocks, if reusable as building materials, are left in storage for reuse.

The CDW, acquired by local processing plants, is then made available to buyers in the desired grainsize range. The CDW material used here has been first sieved in order to assess the grain size distribution below. This range is mostly unwanted by buyers interested to backfilling because too fine, and therefore not recyclable, hence usually destined to landfilling. The grainsize separation carried out has been used also to verify possible variations of CDW composition as a function of granulometry.

2.2 Mineralogical composition

The mineralogical composition was determined on the starting material and the different granulometries, as well as on the products obtained after the thermal treatments. Specifically, X-ray diffraction (XRD) data were collected with a Panalytical X'Pert powder diffractometer (Geology Division, University of Camerino), using Cu Kα radiation (λ=1.5418Å) and an applied voltage and current of 40 kV and 25 mA, respectively. Acquisition range is from 5° to 70°, step size of 0.02° and an acquisition time of 1 sec/step was used. After the thermal treatments, all products were checked for the presence of glass and/or crystals by both optical polarizing microscope and Electron Microprobe (EMP) and then were analysed by XRD, which allowed identifying the crystalline phases and evaluation of the quantity of amorphous materials vs. the degree of crystallinity of the products. The Reference Intensity Ratio (RIR) method (Hubbard and Snyder, 1998) based upon scaling the sample diffraction data to the diffraction of standard reference materials (software Match! https://www.crystal-impact.com), was used to quantify the phases present and comparing the results of all the experiments.

2.3 Thermal treatments

The thermal treatments of CDW, towards vitrification, were carried out at ambient pressure in air using a chamber furnace (Carbolite RHF 14/3, ~3 litre internal volume). For the procedure used, 5g of CDW powder were loaded in a platinum crucible (15ml capacity) and placed in the constant temperature zone of the furnace. Samples were kept in a drying oven at 110°C overnight before the experiments. Temperature was recorded by a platinum-rhodium thermocouple, located in close proximity of the sample. The starting temperature of the experiments was 300°C, heating rate was set at 20°C/min up to the final temperature (either 1000°C, or 1250°C) and samples were kept at the desired temperature for 2 to 8 hours. After the treatment, the crucible was taken out of the furnace and rapidly quenched by immersion in a water bath.

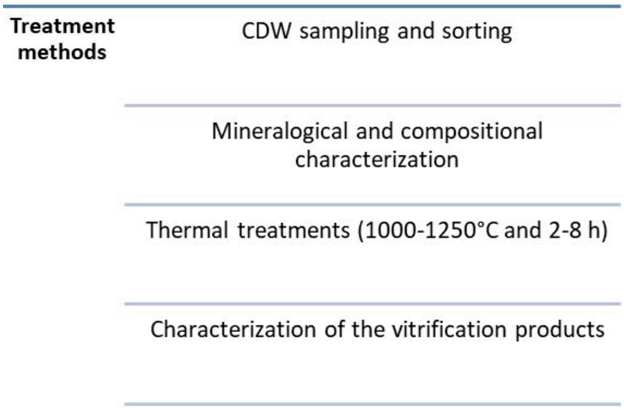

FIGURE 1: Methodological chart showing the sequence of treatments on CDW.

2.4 Chemical characterization

To determine the chemical compositions of the starting CDW sample, prior to thermal treatments, acid dissolution followed by ICP-MS analysis were employed in order to provide analyses of major, minor and trace elements. A lithium-borate fusion prior to acid dissolution was used for the major and more refractory elements, mainly major oxides and rare earth elements (REEs). The relative measurement uncertainties, validated on international standards, are 5% for trace elements and 10% for major elements.

Chemical analyses of the major elements in the samples treated at 1250°C and for the longest duration were obtained by electron micro-probe (EMP) using a CAMECA SX50 at the Consiglio Nazionale delle Ricerche (CNR)- Institute of Geosciences and Earth Resources (IGG) in Padova (Italy). Sample fragments obtained from the vitrification processes were embedded in 25.4 mm diameter epoxy plugs, polished to 1 µm quality, and then coated with an approximately 20 nm thick carbon layer for charge dissipation. The measurements were performed with an acceleration voltage of 20 keV, a beam current of 2nA for potassium, sodium, aluminium and silicon, all measured at first, and a beam current of 20 nA for all the other elements. All the analysis used a slightly defocused (ca. 10 µm) beam to minimize alkali volatility. The reference materials used for the analyses consisted of a range of silicate and oxide minerals of composition similar to the studied compositions, such as wollastonite, albite, orthoclase, TiO_2, Fe_2O_3, etc., following standard procedures. All measurements were made using wavelength-dispersive spectrometers. Sample compositions are given in oxides wt. % along with standard deviations in Table 1.

TABLE 1: Calcite and quartz proportions (wt.%) in CDW for different granulometries.

CDW sample grainsize (mm)	Calcite (wt. %)	Quartz (wt. %)
A6 <0.125	77.6	22.4
A5 0.125-0.257	76.3	23.7
A4 0.257-0.5	85.4	14.6
A3 0.5-1.0	86.3	13.7
A2 1-4	78.4	21.6
A1 >4	89.6	10.4
Average	82.3±5.5	17.9±5.7

Values obtained by Reference Intensity Ratio (RIR) method.

3. RESULTS AND DISCUSSION

3.1 Grain size distribution

The CDW sample before sieving is shown along with the cumulative grain size distribution in Figure 2 (a-b). CDW fractions divide almost equally between the grainsize range 1-4 mm (range of sand-gravel sizes), while the remaining fractions range from coarse to very fine sand (<1mm). This distribution reflects that the finest fractions (<2mm) can account for the majority of the CDW finer grainsize, compromising the use of CDW as backfilling materials towards final landfilling. This information therefore prompted us to investigate the possibility to use the finest grainsize as secondary raw material (SRM) in the ceramic or glass industry, by investigating their vitrification capacity in an attempt to find an upcycling application for CDW, which could be highly desirable, especially for the fine fractions for which there is a lack of effective recycling options.

3.2 CDW mineralogical and chemical characteristics

Six samples, with grainsize between <4 and <0.125mm (A1 to A6 in Figure 3) were analysed by XRD to identify the

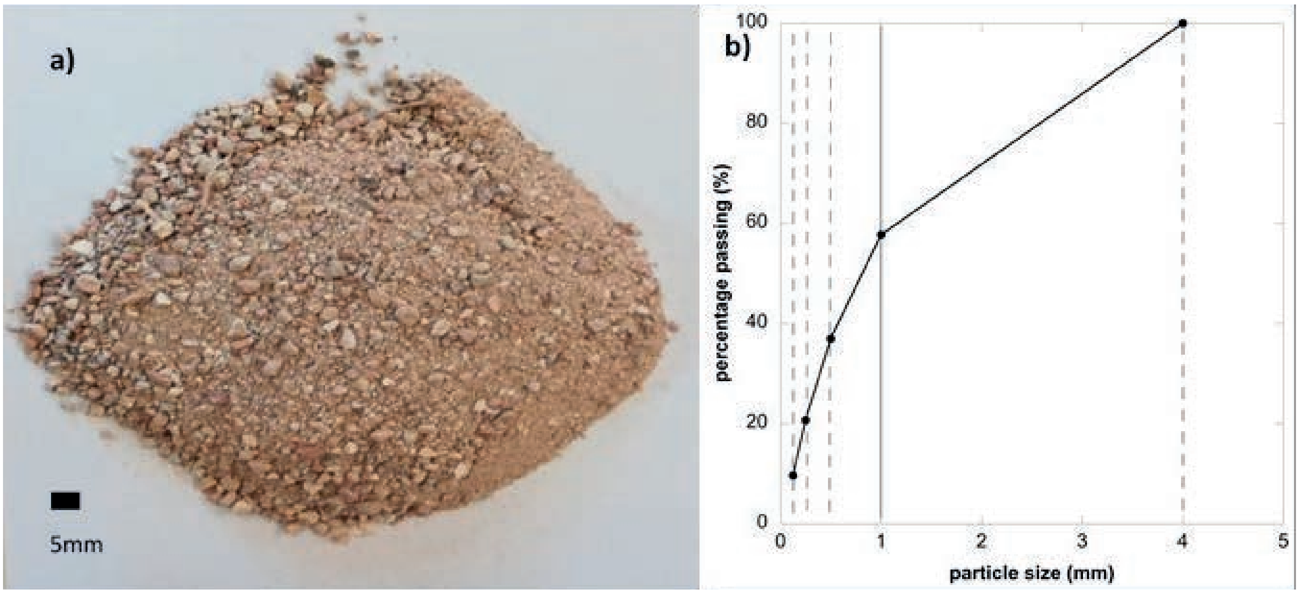

FIGURE 2: (a) CDW sample before sieving; (b) Rosin Rammler particle size distribution.

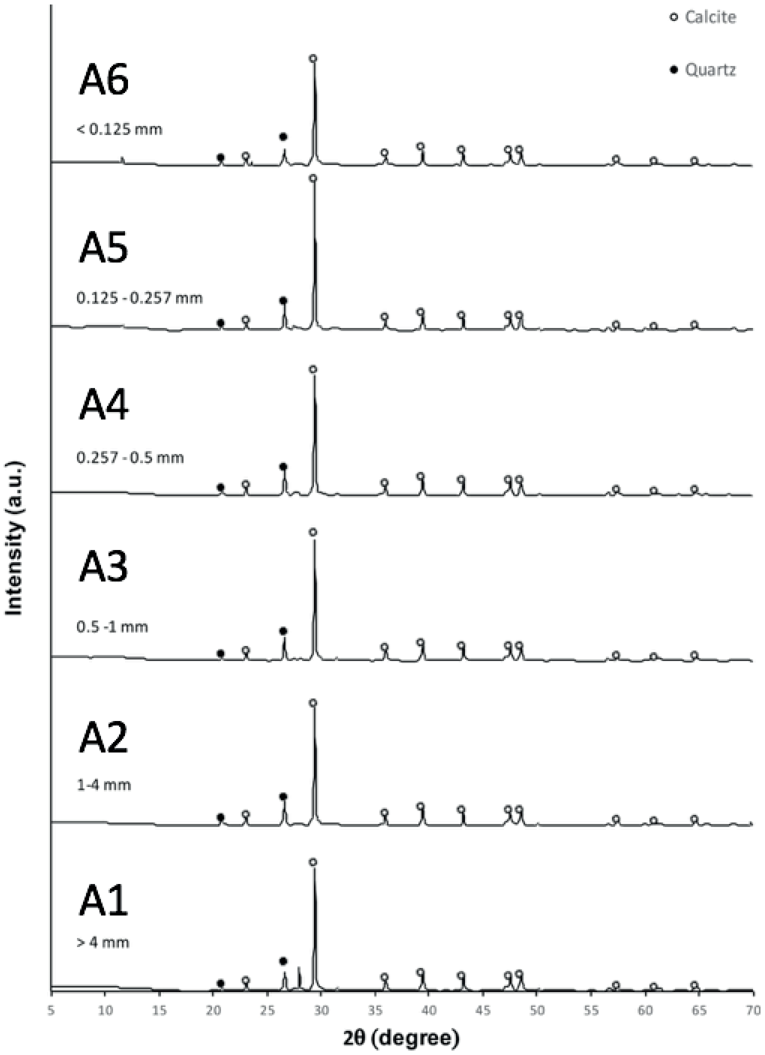

FIGURE 3: XRD mineralogical composition of CDW different granulometries. Notes: full circles are Quartz; empty circles are Calcite; grain-size intervals in mm are reported.

mineralogical content of the samples and verify if the particle size could show different mineralogical compositions, affecting therefore the products obtained upon the thermal treatment. The XRD data of the CDW samples indicate that the principal mineral species is Calcite, followed by Quartz and by trace amounts of phyllosilicates, visible only in the finest grainsize samples. Calcite is always strongly predominant in all the different granulometries analyzed (Figure 3, Table 1), averaging 82.3±5.5 wt.% for Calcite vs. 17.9±5.7 wt.% for Quartz, as determined via the RIR method. The samples are rather homogeneous in mineralogical composition and all very enriched in $CaCO_3$, as originating from a CDW composed predominantly by building rocks like limestones and travertines, or sandstones rich in the calcareous component, all common in the area, as well as occasional ornamental rocks like marbles and granites. XRD data show that there is no strong variation in composition between the different grainsize categories, apart the limited variability of Calcite and Quartz contents, all reflecting the building materials and architectural styles of the rubble provenance area. The building materials composing the CDW are in fact directly connected to the geological characteristics and availability of lithologies in the area. These are mostly used locally and have been extracted for centuries, as evidenced by the many historical buildings, some also affected by the earthquake. Some stone quarries are still in operation now. On the other hand, in the area there is also a long-term tradition of exploitation of clays deposits for the ceramic industry, which produced an established use of bricks/roof tiles as major building materials in constructions, since the XIV century. All these materials are therefore expected to contribute to the bulk composition of CDW from the area, as well as others like concrete or metakaolin from sanitary waste and limited plaster although it should be separated from CDW before crushing.

The contribution of all materials to CDW composition is evidenced by the bulk chemical data. The XRD data agree well with the chemical composition (Table 2) showing that this CDW is very low in silica, with Si coming mainly from

TABLE 2: Major oxide composition (wt%) of starting CDW and CDW treated at the highest melting temperature for 8 hrs (CDW-1250) reported along with literature CDW data and other comparable compositions.

	This study		Other CDW compositions			Other compositions	
Wt%	CDW-bulk	CDW-1250	Volpintesta (2019)	Favaretto et al. (2017)	Bianchini et al. (2005)	CR1 Concrete	FC1 Clay bricks
SiO_2	32.89	27.33 (1.51)	33.26	85.50	51.90	38.03	63.00
Al_2O_3	6.42	24.08 (2.11)	9.65	3.69	12.48	5.01	15.14
CaO	54.43	39.14 (0.42)	47.07	7.12	22.64	39.88	8.52
FeO*	2.79	5.64 (1.66)	4.16	1.41	5.17	1.80	5.03
MgO	1.60	2.85 (0.67)	2.87	1.34	4.45	12.21	3.22
Na_2O	0.58	0.64 (0.23)	1.05	n.d.	1.79	0.74	1.00
K_2O	1.29	0.32 (0.16)	1.52	0.92	2.17	1.06	2.57
Total	100	100	100	100	100	99	98.48

Notes: Microprobe analyses for CDW-1250 are based on 10 measurements, using analysis conditions as described in text. Measurement's uncertainty for CDW-bulk is 10 %, while standard deviations for CDW-1250 are reported into parenthesis. Totals have been normalized to 100%; FeO refers to total Fe expressed as FeO. CDW composition from Bianchini et al. (2015) corresponds to an average value; CR1 Concrete and FC1 clay bricks compositions are taken from Panizza et al. (2018).*

Quartz grains from sand and cement, or from clays constituting the ceramic components like mixed bricks, tiles and roof tiles. It contains abundant Ca, reflecting the presence of both cement/concrete and limestones or sandstones. In particular, major oxides are represented by CaO (54.3 wt%), SiO_2 (32.8 wt%), and Al_2O_3 (6.4 wt%), followed by other oxides in minor amounts (Table 2). Table 3 reports also the minor elements, including REEs, determined in the CDW sample, in total accounting for about 0.1 wt.%. In particular, elements such as Pb or other heavy metals, for instance, are considered to be harmful species that are not permissible for direct disposal or reutilization, although in this CDW sample they are well below allowed environmental protection limits. The chemical characterization, however, is particularly important for the impact CDW can have on the environment when recycled, to quantify the release of possible harmful elements with time, carried out using leaching tests (Kida et al., 1996; Kosson et al., 2002). The leaching properties of trace elements are determinant to learn about their chemical speciation and to gather information about the chemical properties of the waste before its eventual disposal into the environment or recycling into new waste-based products. Though, leaching tests are commonly widely used for other different waste materials, more hazardous than CDW, as for instance municipal solid waste bottom ash, that are notably richer than CDW in harmful elements (e.g. As, Cu, Cd, Cr, Hg, Ni, Pb and Zn; Li et al., 2004), which represent a significant risk for contamination of groundwater resources and surrounding soils via weathering.

TABLE 3: Minor elements (ppm by weight), including REEs, in the starting CDW of this study.

Minor elements (ppm)	CDW-bulk	Minor elements (ppm)	CDW-bulk
Sr	812	U	1.47
W	82	Er	1.24
V	49	Yb	1.19
Pb	40.7	Eu	0.52
Ce	25.9	Ta	0.40
La	15.4	Ho	0.40
Y	13.50	Tb	0.33
Nd	13.2	Lu	0.17
Th	4.66	Tm	0.12
Sn	4.00		
Pr	3.40		
Gd	2.34		
Sm	2.32		
Dy	2.10		

Notes: ICP-MS and acid dissolution have been employed with a relative measurement's uncertainty, validated on international standards, of 5%. Elements are reported in descending order of abundance.

If compared to other CDW analyses from the literature (Table 2), CaO is strongly predominant in this CDW, whereas SiO_2 and Al_2O_3, are considerably depleted, showing that the CDW compositional range is highly variable both in oxide abundances and proportions among them, depending on the different nature of the waste and provenance. Also, although CDW represents a problem in waste management and recycling on a global scale, it is worthwhile noticing that literature studies are still lacking of CDW chemical composition data, in spite of the importance of this information which can negatively affect industrial applications. For a better evaluation of the available sources, CDW data were compared using the $CaO–Al_2O_3–SiO_2$ ternary diagram (Figure 4), where the composition of mineral phases typically recognised in cements and ceramic materials were also reported. Raw material compositions required for ce-

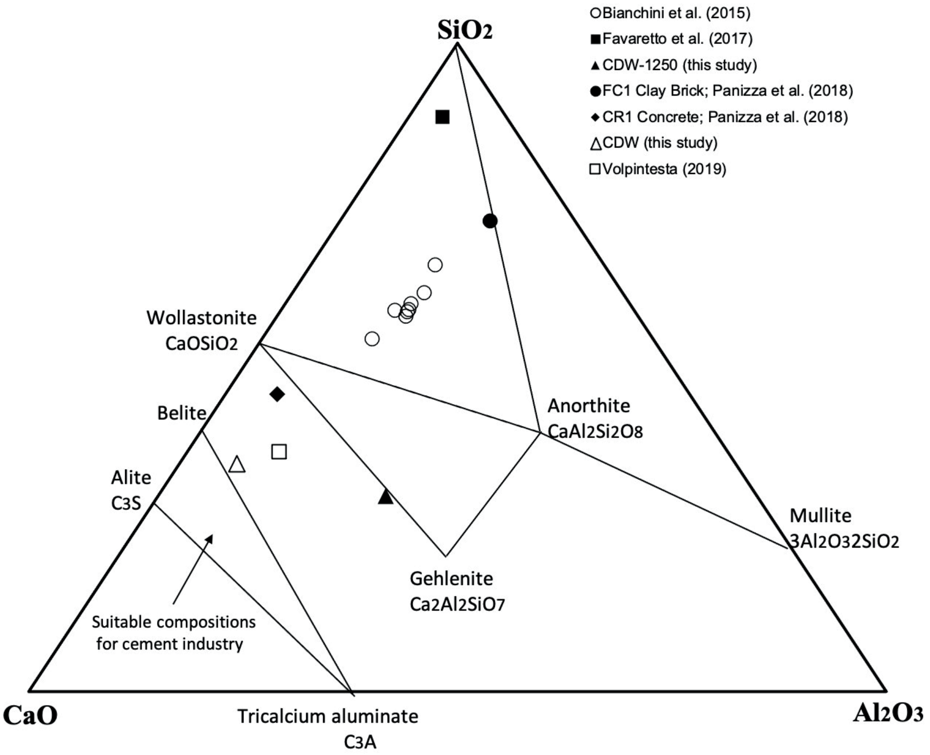

FIGURE 4: Ternary diagram describing the system CaO–Al2O3–SiO2 (wt%). Triangles are our CDW study composition (empty starting material, full after thermal treatment at 1250°C, circles are data from Bianchini et al. (2015) and full square from Favaretto et al. (2017). A clay brick and a concrete composition (Panizza et al. 2018) are reported as comparison.

ment preparation are usually included in the sub-triangle tricalcium silicate (alite; C_3S) - dicalcium silicate (belite; C_2S) - tricalcium aluminate (C_3A) (Manning, 1995). Components that could be suitable as secondary raw materials for the ceramic industry can be considered in the same diagram (Bianchini et al., 2005). In Figure 4, the CDW composition used in this study (empty triangle) shows a silica content considerably lower than the literature data of both CDW compositions from Favaretto et al. (2017) and Bianchini et al. (2005). Moreover, CDW (this study) composition is also far from CDW-1250 (full triangle in the Triplot), because the latter corresponds to a residual melt composition (Tab. 2) of the treated CDW_1250 sample with ca. 50% xls. Data from this study and from Volpintesta (2019) are more similar to each other, falling close to the sub-triangle containing compounds suitable for cement industry. Moreover, if we consider the Basicity Ratio (BR) of the different CDW materials, as a parameter expressed as the ratio between the CaO/SiO_2 oxides wt% or C/S (data from Table 2), the CDW of this study has a BR of ca. 0.6 compared to the literature C/S values that reach values strongly higher than the unity, up to an average value of 2.2 (as for Bianchini et al. 2015), or even 12 as for Favaretto et al. (2017) samples. This reflects a composition with a high proportion of concrete, limestone and ceramic components able to produce a CaO-rich and SiO_2-poor CDW. This information is useful for a better comparison between materials and suggests a possible recycling application of this CDW in the cement industry.

3.3 Thermally-treated CDW

The samples used as starting materials for the thermal experiments were selected from two different gransizes: sample A6 (<0.125 mm), the finest and most interesting fraction for recycling this CDW in a non-conventional way, and A1 (>4 mm), the largest size, which contains the least amount of Quartz of all the sample series.

The experiments were carried out on sample A6 in the temperature range 1000-1250°C for 2 hours at each T step. The XRD data were collected after each step in the same analytical conditions. Figure 5(a) shows that Quartz decomposes rapidly going from the experimental run at 1000°C to 1100°C and it is completely absent at 1200°C. At 1000°C Gehlenite and Larnite, both Ca-bearing silicate species, are already forming thanks to the decomposition of Calcite and to the availability of silicon and aluminium.

Calcite, in fact, is known to start decomposing slowly at 700°C, more rapidly at 750°C, to complete decomposition by 800°C (Karunadasa et al., 2019), releasing CO_2 (Trinidade et al., 2009) and forming lime, CaO. Portlandite, $Ca(OH)_2$, present in the 1200°C XRD pattern, formed as a secondary phase after CaO. The hydration and carbohydration processes are expected to occur after thermal treatment, although some Calcite is still evident at 1000°C, due to the fast recarbonatation of CaO in the sample holder during the XRD analysis, without indication, however, of significant volume changes (Setién et al., 2009). In contrast, the carbohydration processes of periclase (MgO), which could provoke a significant volume increase jeopardizing

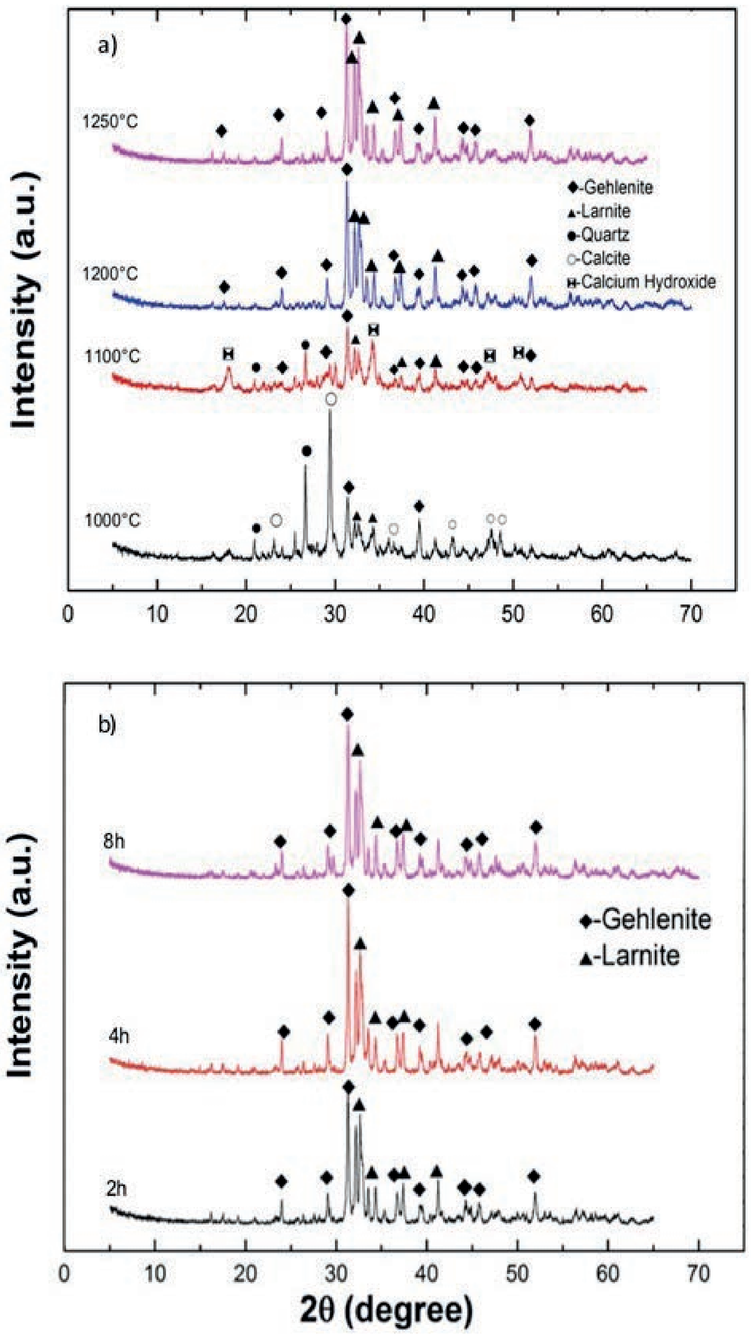

FIGURE 5: a) XRD analysis of CDW A6 sample treated at different temperature and same duration of 2 hours each; b) A6 at the highest temperature (1250°C) and different durations (2 to 8 h).

the use of this treated material in construction and building (Juckes, 2003), is however negligible because of the very low amount of Mg present in the CDW starting composition, which prevented the formation of MgO.

The newly formed species, Gehlenite and Larnite, already present in the experimental runs at 1000°C, at 1200°C are the only crystalline species present in the samples. They persist at increasing temperatures and are still present in the samples heated at 1250°C, independently from the duration of the thermal treatment, as shown in the runs lasting from 2 to 8 hours (Figure 5(b); Table 1), which indicate there is no significant variation in the XRD patterns, testifying that they are the only crystalline compounds present in the sample.

Figure 5(b) shows that there is no further variation of the mineralogical composition after experiments at 1250°C, even with increasing the experiment duration from 2 to 8 hours. Longer runs have been carried out (up to 16h) also with no difference in results. This means that the two phases are already stable even using the shortest experimental time of 2 hours, which could be interesting for the optimization of an industrial process.

Moreover, the comparison of XRD data between sample A6 (the finest) and A1 (the coarsest) collected from

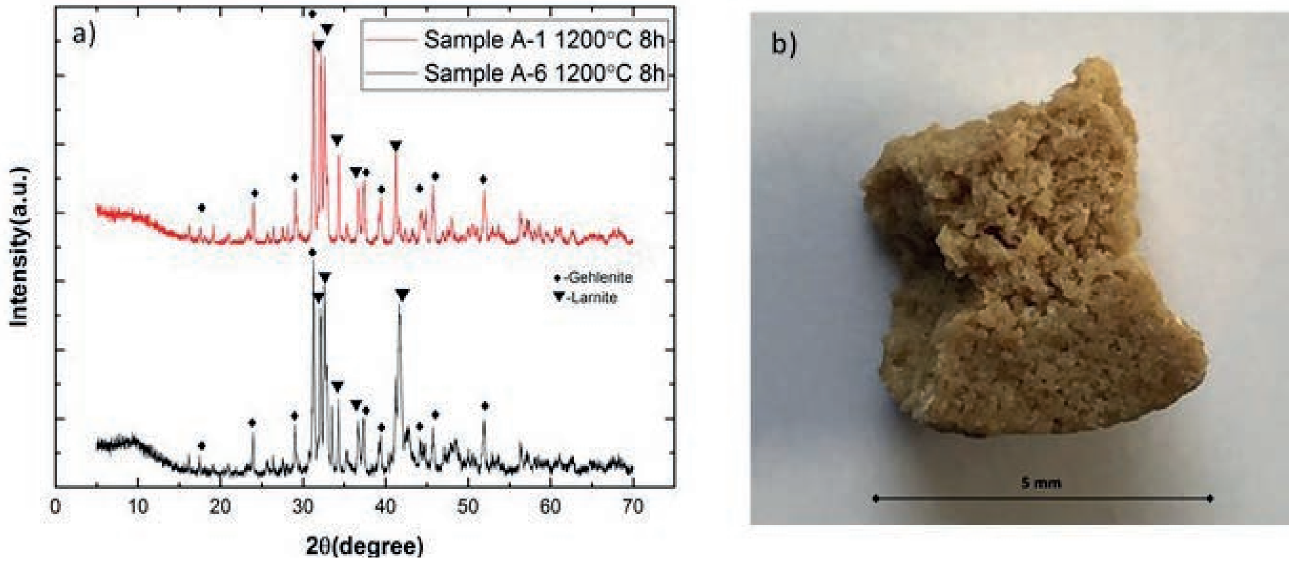

FIGURE 6: (a) Comparison of XRD data between the samples A1 and A6 after thermal treatment at the same temperature (1200°C) and duration (8h). (b) The porous material produced at high temperature from the CDW thermal treatment.

experiments at same temperature and duration (1200°-8h runs) (Figure 6a) show that the samples reach the same mineralogical composition, without significant variation as a function of granulometry. This suggests that sieving this CDW below 4mm size does not affect the thermal treatment results, producing the same type of final product (Figure 6b).

Gehlenite and Larnite form, above 750°C and 570-670°C, (Gonzalez-Garcia et al., 1990) respectively, from the reaction between CaO, SiO_2 (and Al_2O_3 for Gehlenite), thanks to the decomposition of the original CDW components. The composition of the two species is reported, as an indication, in the ternary diagram CaO-Al_2O_3-SiO_2 (Figure 4). In particular, Gehlenite ($Ca_2Al(AlSiO_7)$) is the Ca and Al-rich end member of the Melilite binary system (Gehlenite-Åkermanite) where Åkermanite is the Mg and Si-rich end-member, $Ca_2Mg(Si_2O_7)$. Gehlenite is also a high temperature phase forming in ceramics and is common in heterogeneous waste materials after treatment at high temperature, like municipal solid waste bottom ash (e.g. Schollbach et al., 2016; Stabile et al. 2019). Pure Gehlenite melts at 1595°C at ambient pressure, well over the temperature of these experiments, accounting for the lack of melting of this CDW sample. The structure of Gehlenite can incorporate the Al present in CDW and can also include a substitution of Al by Fe^{3+}, even up to 18 wt.% at 1237°C (Huckenholz and Ott, 1978), suggesting that Gehlenite structure could allocate also the ferric iron present in the original CDW (Table 2). Also, other transition elements could possibly substitute in Gehlenite, suggesting it might serve as a possible confinement phase for other metals.

Larnite Ca_2SiO_4 is the natural mineral species whose structure corresponds to the synthetic β-Ca_2SiO_4, forming as belite in Portland cement. The name larnite is connected to a series of Ca_2SiO_4 polymorphs (γ, β, a in order of increasing temperature of stability) where β and α phases are characterized by reversible polymorphic transitions and α presents also a higher and a lower temperature phases (Yamnova et al. 2009), difficult to detect because of peak overlapping in the XRD patterns of quenched samples. The low temperature synthetic γ phase has its natural analogue in the Ca-olivine (γ- Ca_2SiO_4) the calcium end-member of the olivine group of minerals.

In the experimental runs (Figure 5), Larnite is present at 1000°C and with increasing temperature there is a progressive increase of larnite presence and also an increase of the crystallinity degree of the two species, Gehlenite and Larnite, from the early crystal nucleation up to the highest experimental temperature (1250°C). This is testified by the increased sharpness and intensity of the diffraction peaks and by the decrease of the amorphous components, as evidenced by the flattening of the XRD pattern background. The early formation of larger amounts of glass may reflect metastable melting, followed by delayed nucleation and growth of larnite and gehlenite. The minor presence or even the absence of amorphous material at high temperature reflects the highly refractory nature of this Si-poor, Ca-rich CDW compositions investigated.

Calcium silicates are interesting materials for CO_2 sequestration (Santos et al., 2009): this is possible because, for example, Larnite presents the highest carbonation reaction rate among the calcium silicate, with 64–67 wt.% of CaO, about 50% more than that of wollastonite (42-44wt.% of CaO), interesting therefore for acting as a possible CO_2 structural confinement (via Ca-carbonate formation). While here the process of decarbonation takes place by a high temperature treatment of CDW, it is however interesting that the calcium silicates, on the other hand, could possibly be recycled as CO_2 storage materials. The carbonation process is not well documented yet in Larnite, although its β-Ca_2SiO_4 phase Belite is a major constituent of Portland cement where carbonation is an important process capable to reduce its environmental impact, helping also decrease curing time and increasing materials strength (Santos et al., 2009). A different and peculiar aspect relates also larnite and its possible role in an acid-neutralizing effect produced after the Cretaceous/Tertiary meteoritic impact, where a mechanism producing larnite from the interaction between acid rain and the vapour plume produced from the Ca-rich target rocks at the impact site, able to neutralize acidity in a short time preventing the acidification of freshwater and preserving the living organism within (Maruoka & Koeberl, 2004).

3.4 Further considerations

3.4.1 Vitrification

The presence of crystalline phases and their persistence in the thermally treated samples implies that complete vitrification is not accomplished for the studied CDW sample, using this thermal process (maximum T=1250°C, 8 hrs). In the treated samples, newly formed phases like Gehlenite and Larnite can be observed and are stable up to at least 1250°C. Thus, it is clear that the amorphous proportion in the samples, when present at these temperatures, is nonetheless limited by the refractory, Si-poor and Ca-rich bulk composition.

In fact, the CDW studied, although chemically belonging to the SiO_2-Al_2O_3-CaO system, does not contain amounts of network-forming elements in quantities that could facilitate the vitrification process by lowering liquidus temperatures. A silicate glassy material consists of a network of SiO_4 tetrahedra in which Si^{4+} can be partially replaced by other cations with similar ionic radius as network-formers; among these, Al_2O_3, TiO_2 or P_2O_5 usually act as glass forming oxides (Öveçoğlu, 1998; Colombo et al., 2003). On the other hand, other cations, such as alkali metals or alkaline earths, like CaO and MgO, are network modifiers and decrease the connectivity of the glass network (Schaffer et al., 2010). Therefore, if the starting chemical composition results lacking of network-formers, the formation of a glass phase can be expected only if more glass-forming oxides are added, preventing crystallization of high temperature-stable phases and lowering melting temperatures. Therefore, to reach vitrification at reasonable temperatures (<1300°C), a change of chemical composition of the starting CDW is necessary.

Although most wastes can vitrify without additions or fluxes (Neuville et al., 2017), the addition of a modest amount of glass cullet or other compounds (like quartz sand) can promote the vitrification, if such agents are not present in sufficient quantity. In fact, considering a glass-forming system, a wide range of elements can play a crucial role in stable and metastable phase separation processes, inhibiting nucleation and crystallization in response to a physical or chemical change (e.g. heating) (Galoisy 2006; Neuville et al., 2017). To overcome this problem and obtain higher amorphous contents, preliminary tests were carried out by combining CDW with silicate glass waste materials. In particular, special glasses, like borosilicate or lead-containing glasses, multilayer glasses (laminated glasses or windshields) or solar panel glasses, cannot be inserted in the normal waste streams, requiring special recycling techniques or missing recycling due to lack of applications or for the presence of contaminants which could be released in the environment. Thus, they are often destined to landfill if a separate recycling line is not available and can represent an important environmental issue (Barbieri et al., 2000; Colombo et al., 2003; Kavouras et al., 2003; Li et al., 2004; Stabile et al., 2019). In these tests, by mixing CDW with 30 wt.% of windshield glass, the amorphous proportion obtained in the thermally treated sample, determined by RIR method, reaches 58 wt.%, compared to 42 wt.% crystalline phases present. This value increases up to 75 wt.% if 50 wt.% of glass is added, making this process valuable for the production of a fully glassy materials by a mixing of wastes.

3.4.2 Applications

Apart from CDW recycling as a backfilling material, still the most used worldwide, other applications can be highlighted for the recycling of this CDW, which render useful this material, especially the finest unwanted grainsizes.

The vitrification data give us promising information about the propensity of this CDW materials for vitrification and suggests a procedure that could be adopted to improve recycling of low-quality waste materials as secondary materials in the glass industry, for example in the production of soda-lime glass (Na-silicate + CDW). Moreover, one possible application of vitrified CDW is contributing to the production of glass fibres (Scarinci et al., 2000), in alternative or in combination with other waste, such as bottom ashes from incinerators and milled glass fibres from the recycling of fiberglass insulation material (Lopez et al., 2012). On the other hand, this CDW proved to produce a refractory material when thermally treated, with a porous texture, possibly interesting as thermal insulator, and meriting further work concerning porosity evolution as a function of heat treatment conditions.

Other possible uses, some of which already mentioned before, can be proposed in view of the chemistry of this CDW, such as:

- as secondary raw material in the production of cement and concrete, depending not only on the composition, but also on the granulometry (Oliveira et al., 2020);
- for the production of geopolymers, incorporating CDW as either inert aggregates or partially reactive materials (Panizza et al., 2018);
- in acid correction in soil and freshwater in polluted areas, which can be of interest whenever, such as in this work, CDW is duly separated and characterized, while it encounters difficulties otherwise (Staunton et al., 2014), and providing that there is no presence of heavy metals;
- in mining areas of sulphide ore deposits, for the correction of Acid Mine Drainage (AMD), commonly carried out using limestones, which however could be partially substituted by CDW of similar composition due to its high Calcite content (Yilmaz et al., 2018).

Also, whenever CDW contains sufficient amounts of clay materials, the extraction from waste of nanoclays would represent an enhanced upgrading option, since nanoclay does have extensive use e.g., for the improvement of mechanical and wear properties of thermosetting polymer resins including those used in the construction sectors (Shettar et al., 2020).

4. CONCLUSIONS

The large differences among CDW samples of different provenance (and/or waste source) highlight the strong heterogeneity of CDWs coming from different regions or localities where they have been produced and collected. This observation suggests the need of a better characterization of CDW since the bulk chemical and mineralogical composition must be controls possibilities for finding novel upgrading industrial applications for this material.

From the mineralogical analysis, the CDW composition appears to be quite homogeneous and characterized by a strong predominant presence of Calcite over Quartz, independently of its granulometry. The results of this study show that a complete glassy state is not reached by the CDW samples, at the highest T used (1250°C) and despite even long experimental duration, producing a porous refractory material. This is due to the chemical composition of CDW, characterized by a lower amount of glass network-forming cations like silicon and aluminium in the sample composition, combined with a higher amount of calcium. However, mixing this CDW with a waste-glass component improves drastically the vitrification process, strongly promoting an increased amount of glass formation, thus opening a larger application field for this CDW.

Moreover, its chemical composition makes the studied CDW most likely suitable either for direct use as raw component or as a tool for compositional correction of other materials for the cement preparation and for the making of other cement-based building products. Additionally, CDW produced by vitrification shows a particular porous macro-appearance that may imply its possible application as filler in cement-based insulation materials.

The results obtained are in the view of reducing as much as possible the downgrading of CDW materials to proceed with their economically and structurally viable recycle. It needs to be suggested that, although EC 98/2008 directive indicated 70% of CDW to be recycled by 2020, this ambitious objective has proven difficult to be reached, due to the inconsistencies about the secondary raw materials supply and the consequent possibilities to recycle it in new materials for construction. Setting general rules, though necessary, led so often to dismiss the local situation about generated rubbles, their nature and inherent quality, together with their chemical composition and possible vitrification. This study is aimed at offering the evaluation of these characteristics to serve towards the fulfilment of circular economy goals by tailoring the treatment of local CDW to its effective recycling potential.

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TECHNICAL ANALYSIS OF FULL-SCALE CONSTRUCTION AND DEMOLITION WASTE TREATMENT PLANTS: CASE STUDIES OF THE LOMBARDY REGION, ITALY

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ABSTRACT

Construction and demolition activities in Italy and the Lombardy Region produce a considerable amount of wastes that can be valorised as secondary raw materials. The recovery of construction and demolition wastes is severely limited by the lack of consolidated and sustainable treatment chains and by the strong variability of their environmental characteristics that may generate potentially dangerous effects for the environment. The purpose of this study was to evaluate and technically define the current treatment chains in the Province of Brescia (Lombardy) by analyzing three full-scale treatment plants in order to highlight barriers to demolition waste recovery and propose possible solutions and improvement strategies. To best represent the entire management and treatment system, the analysis was developed on all the acceptance, treatment, and final quality control phases. Moreover, chemical composition and leaching data on demolition wastes and recycled aggregates were collected from the three treatment plants and statistically analyzed to assess their potential recovery according to the Italian legislation. Mixed non-hazardous waste is the fraction mostly managed by recycling plants and mixed recycled aggregates are the main products obtained from the treatment. These are mostly used in roads and in geotechnical applications. Chemical composition results showed that the pH is generally alkaline and tin and benzene are the most critical elements for both demolition wastes and recycled aggregates with respect to the regulatory limit values. Total chromium was identified as a critical compound in leachates. The results of the statistical analysis confirmed that chromium was mainly released by cement materials.

1. INTRODUCTION

Construction and demolition activities produce large quantities of waste materials, called Construction and Demolition Wastes (CDWs), classified as special waste according to the current Italian legislation (Legislative Decree n. 152, 2006) and codified by Chapter 17 of the European Waste Codes (EWC). In 2016, the total wastes generated by the construction sector in Europe (EU) accounted for around 36% of the total waste production, with 924 million tons (Eurostat, 2019). Italy, with a production of about 54.5 million tons and a per-capita production of 0.9 tons, is the fourth European country for CDW production after France, Germany, and the Netherlands.

The level of EU recycling varies significantly between the Member States: from 10% (e.g. Greece) to 90% (e.g. Germany, the Netherlands) (Giorgi et al., 2018). In Italy conflicting data on recovery rates are reported: from 10% (Legambiente, 2017) to 75% (ISPRA, 2019).

In the Lombardy Region (northern Italy) about 12.2 million tons of CDWs were produced in 2016, which represent 41% of all special wastes produced in the region (29.4 million tons) (ISPRA, 2019). Regarding the recovery, 16.5 million tons of CDWs were treated in the region: 73% were recovered through R5 (recovery of inorganic substances) operations, 14% were recovered through R4 (metal recovery) operations, 5% were recovered throughother operations, while 8% were disposed of in landfills (Lombardy Region, 2018).

The interest in CDW recovery is supported by both European (Directive EU 2018/851) and Italian (L.D. 152/2006) legislations, which aim to improve the efficiency of resource recovery (e.g. any operation the principal result of which is waste serving a useful purpose by replacing other materials which would otherwise have been used to fulfil a particular function, or waste being prepared to fulfil that function, in the plant or in the wider economy) as well as

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to facilitate the transition to a more sustainable material management and to a circular economy model. In particular, the Directive EU 2018/851 requires that at least 70% of the non-hazardous CDWs produced must be recycled/recovered by 2020. As a consequence, the sustainable CDW management and recovery has become a topic of debates at the national and regional level (Goh, 2016).

Generally, CDWs represent a large source of secondary raw materials, consisting roughly of concrete, wood, masonry, drywall, glass, plastics, metals and more (Bianchini et al., 2020; European Commission, 2017). The composition is therefore extremely variable and heterogeneous. This is mainly due to: the non-selective demolition techniques generally applied by breakers, to the lack of advanced treatment processes for recycled aggregate (RA) production, as well as to the high variability of materials used for building's construction (Ruggeri et al., 2019; Siddique & Cachim, 2018). Based on these considerations, over 90% of CDWs produced in Italy derive from non-selective microdemolition operations of the residential and non-residential building stock (i.e. maintenance and/or renovation works) and this significantly increases the overall CDW heterogeneity. Conversely, only 8% comes from entire building demolition (Altamura, 2015). As a result, RAs are mainly used in downcycling practices, i.e. recycling of waste in cases where the recycled product is of a lower quality than the original ones.

In the Lombardy Region, CDWs are mainly treated in dedicated mobile plants that carry out rough treatments, such as single crushing and optional screening. As stated by Borghi et al. (2018), 86.1% of CDWs is treated in mobile plants, while only 13.9% of CDWs is sent to fixed plants. The resulting RAs have, therefore, extremely variable physical and chemical characteristics that often do not comply with the national environmental standards (Ministerial Decree n. 186, 2006) required for their use as unbound aggregates in the construction sector. In particular, in terms of contaminant release, chromium and sulphates are the most critical compounds in CDW and RA leachates (Butera et al., 2014; Galvín et al., 2014). Specifically, total chromium is mainly released by ceramic materials and partly carbonated samples, while high sulphate levels are released by gypsum-based materials and other CDW compounds (e.g. mortar particles) (Barbudo et al., 2012; López-Uceda et al., 2019; C. S. Vieira & Pereira, 2018).

In this context, only a few studies have been developed on the in-depth analysis of the Lombard CDW treatment chains (Borghi et al., 2018; Pantini et al., 2019; Pantini & Rigamonti, 2016). The authors of these studies have assessed the environmental impacts of the Lombard CDW management system, by applying the Life Cycle Assessment methodology, to improve the environmental performance of the current system. Conversely, literature studies on the CDW treatment chains of the Province of Brescia (the biggest Lombardy province) have not been developed so far.

Hence, the aim of this paper was to operationally define the current CDW recovery and treatment chains of the Province by analyzing 3 selected recycling plants (geographically representing the whole province). In particular, based on the observed plant behaviour, the main purpose of this technical analysis was to highlight barriers and hurdles (logistical, economic and regulatory) that still limit the recovery of CDWs and the RA end-use and to propose possible solutions and improvement strategies.

The importance of this study stems from the fact that the CDW recovery is a common concern in many Member States (e.g. Spain, Greece etc.). At the European level, this issue is addressed through the development of guidelines that aim to support national authorities in achieving the EU target for CDW recycling (European Commission, 2016). In this context, the knowledge of real case studies regarding the experiences implemented by the treatment plants as well as the obstacles still present can certainly provide an important contribution to the identification of new strategies and policy instruments by national authorities applicable at a national and international level.

Finally, since the CDW heterogeneity and the treatment techniques adopted have a significant influence on the characteristics of the final recycled materials, the present work aims also to evaluate the technical (EC marking) and environmental properties of these materials. In particular, to identify possible correlations between RA characteristics and CDWs, data on chemical and leaching properties of both CDWs and RAs were collected from the databases of the three selected CDW treatment plants and then elaborated using a statistical approach.

2. MATERIALS AND METHODS

2.1 CDW treatment plants

Three authorized treatment plants, located in the Province of Brescia (the Lombardy Region - northern Italy), were analysed to study the CDW treatment system currently adopted. These plants, called treatment plant 1 (TP1), 2 (TP2) and 3 (TP3), are in natural aggregate excavation quarries and, over time, have adapted their technologies to the treatment and recovery of CDWs. In particular, in order to evaluate a situation as representative as possible of the regional reality, TP1 and TP3 are representative of mobile treatment plants, whilst TP2 is representative of a fixed treatment plant. As stated by Borghi et al. (2018), these are the two main recycling plant categories present in the regional territory.

Differently from TP2 and TP3, respectively located in the municipalities of Manerbio and Breno, TP1 manages four plants located in the neighboring municipalities of Brescia, hereinafter called:

- TP1$_{,1}$: treatment plant located in the municipality of Berlingo;
- TP1$_{,2}$: treatment plant located in the municipality of Orzivecchi;
- TP1$_{,3}$: treatment plant located in the municipality of Leno;
- TP1$_{,4}$: storage plant located in the municipality of Poncarale (active since 2017). This plant is authorized for only CDW storage. The accepted CDWs must be subsequently sent to the above-mentioned treatment plants (TP1$_{1,2,3}$).

TABLE 1: EWC codes authorized and processed by the treatment plants.

CDW treatment plant		Type	Common authorized EWC codes	Additional EWC codes	Authorized operations	
					R13 (m^3)	R5 (t/y)
TP1	TP1$_{,1}$	Mobile	170101 - concrete 170102 - bricks 170103 - tiles and ceramics 170107 - mixtures of concrete, bricks, tiles and ceramics 170504 - soil and stones 170802 - gypsum-based construction materials 170904 - mixed construction and demolition wastes	-	3,300	107,000
	TP1$_{,2}$				1,750	55,000
	TP1$_{,3}$				2,211	90,000
	TP1$_{,4}$				2,700	-
TP2		Fixed		170508 - track ballast	1,584	55,000
TP3		Mobile		170302 - bituminous mixtures	2,650	101,600

R5: recycling/reclamation of other inorganic materials
R13: storage of wastes pending any of the operations numbered R1 to R12, excluding temporary storage, pending collection, on the site where it is produced

All the treatment plants are authorized to process and manage only non-hazardous CDWs with EWC codes of chapter 17, as shown in Table 1.

From CDW treatment, RAs of different quality and grain size are produced.

2.2 Data collection

In order to best represent the CDW treatment chains currently operating in the Province of Brescia, the following historic data were acquired and collected from the three treatment plants analyzed.

2.2.1 CDW and RA flows

In order to evaluate the amount of CDWs managed and RAs produced by the three treatment plants, the following data were extracted and collected from the databases of the TP companies, on a yearly basis: total quantity of incoming CDWs, quantity of incoming CDWs according to EWC codes of Chapter 17 and, finally, quantity or RAs produced. In particular, the annual amount of CDWs entering the treatment plants was assessed by analysing the annual mandatory declaration (i.e Environmental Declaration Model – calld MUD in Italy) submitted by treatment operators to the territorially competent Chamber of Commerce, in accordance with Article 189 of L.D. 152/2006.

A six-year reference period was used for data collection (from 2013 to 2018). In this way, it was possible to set up the mass balance of the entire CDW management system of the 3 recycling plants.

At the same time, based on the acquired flow data, the current recovery efficiency of the treatment plants and the performances of their treatment systems was also assessed (i.e. ratio between the quantities of CDWs treated and RAs produced).

2.2.2 Quality control procedures carried out on incoming CDWs and outgoing RAs and treatment processes and technologies adopted

In this section, in order to technically outline the current CDW management and treatment systems, primary information and operational-management data were collected during technical visits performed at 3 recycling plants. The main goal was to identify the recovery solutions already applied in the region and the workflows implemented by recycling plants for CDW treatment and RA production.

Based on the requirements imposed by all 3 plant authorizations, data about CDWs acceptance and management criteria were firstly acquired. Subsequently, field visits were performed to gather information about the treatment processes and technologies adopted, as well as the type and actual end-uses of RAs produced. Moreover, the quality control systems implemented in the production of RAs were identified, based on both technical standards and environmental compatibility requirements.

2.2.3 Technical and environmental properties of both CDWs and RAs

To statistically evaluate the technical and environmental behaviour of CDWs and RAs, several certificates of EC marking, chemical characterization and leaching tests were provided by the 3 treatment plants and by the Italian Lombardy Builders' Association (ANCE Lombardy) and the National Association of Recycled Aggregate Producers (ANPAR).

All the certificates refer to a collection time period of 10 years. Over 1,400 certificates were collected in pdf format and subsequently reworked in digital format. Of these, 46 were related to EC marking, 865 certificates were related to chemical analyses and 535 related to leaching tests, divided as reported in Table 2. In particular, as regulated by the Italian Environmental Legislation (L.D. 152/2006 and M.D. 186/2006), chemical analyses refer to the CDW non-hazardousness assessment performed by the CDW owner (breaker) before sending the CDWs to the treatment plants, whilst the leaching tests refer to the RAs eco-compatibility assessment.

The leaching tests were performed according to the compliance test imposed by UNI EN 12457-2:2004. The procedure consists of a one stage batch leaching test at a liquid to solid ratio of 10 L/kg applied to materials with a particle size below 4 mm.

All the collected data were then processed and elabo-

TABLE 2: Number of certificates of EC marking, chemical analysis and leaching test in relation to TPs and material analyzed.

Certificates	Sample	TP1	TP2	TP3	Others (1)	Total
Chemical analysis	CDWs	140	342	283	14	779
	RAs	0	33	52	1	86
Leaching tests	CDWs	15	165	35	35	250
	RAs	134	31	98	22	285
EC marking	RAs	3	3	40	-	46

(1) *ANCE Lombardy and ANPAR databases.*

rated using the statistical software "IBM SPSS 19.0" in order to identify the variability ranges of chemical and leaching characteristics of both CDWs and RAs, as well as the potential critical parameters for recovery under the Italian national legislation (L.D. 152/2006 and M.D. 186/2006). In particular, for CDWs, reference was made to the codes generally most received by the treatment plants: EWC 170904 "mixed CDWs", 170504 "soil and stones" and 170101 "cement" (detailed data reporting most treated CDW flows are presented in subsection 3.1). In this paper, the technical, chemical and leaching properties have been discussed in tabular format, but the release of contaminants has also been discussed by analyzing, with the box plot and violin plot methodology, two of the most critical parameters for CDW recovery: pH and total chromium.

2.2.4 Economic performance of the treatment plants

To identify the difference in economic performance between fixed and mobile systems, the data relating to energy consumption per tonne of RA produced were collected and compared.

2.3 Statistical analysis of chemical and leaching certificates

2.3.1 Box-plot and violin plot methodology

The box-plot method was applied for the processing of statistical data. This method is a useful way to visualize differences among samples or groups and it provides a lot of statistical information, including median, range and outliers, which help to better understand sample characteristics.

The box-plot divides the data sample into quartiles, as shown in Figure 1. The central part of the box-plot consists of a "box", which goes from the first quartile (Q1 – 25° percentile) to the third quartile (Q3 - 75° percentile). Inside the box there is the median of the data set (Q2 - 50th percentile). Half of the data values are less than the median and the other half are greater than the median. Approximately 50 percent of all data fall inside the box (interquartile range IQR).

The lower and upper extremes, graphically represented by a line that goes out from the box to more or less 1.5 IQR (whiskers), represent the maximum and minimum values of the data set that are not outliers or extreme values. The whiskers show the range between these two extremes. A larger range indicates a wider distribution, i.e. more scattered data.

Points outside the whisker boundaries are anomalous values considered as outliers and/or extreme values. These values are very small or large and can affect the overall observation made from the data series. Outliers are all the values that lie from 1.5 to 3 times IQR (marked with circles beyond the whiskers), whilst extreme values lie more than 3 times IQR (marked with asterisks beyond the whiskers).

In the present study, the box-plots were used to graphically reproduce the variability and the dispersion of the various pollutant concentrations present within the data population with reference to the leaching tests performed on CDWs and RAs.

For better data representation, the same data were also elaborated and graphically represented by means of the violin plots. Unlike the box-plot, however similar, the violin plot allows to detect, with its "violin" shape, the density variation of the analyzed data, as shown in Figure 2.

3. RESULTS AND DISCUSSION

3.1 CDW and RA flows

Results from flow data elaboration are presented in Table 3. In the reference period, about 1.3 Mt of CDWs were managed by the three treatment plants. The results show that TP1 managed and treated the largest amount of CDWs equal to over 60% of all CDWs managed by the 3 treatment plants, while TP2 and TP3 manage approximately 20% respectively.

The average recovery efficiency, calculated as the ratio between the quantities of RAs produced and CDWs treated, varies considerably according to the technology adopted by the plant. The fixed plant (TP2), equipped with performing technologies, has the highest recovery efficiency

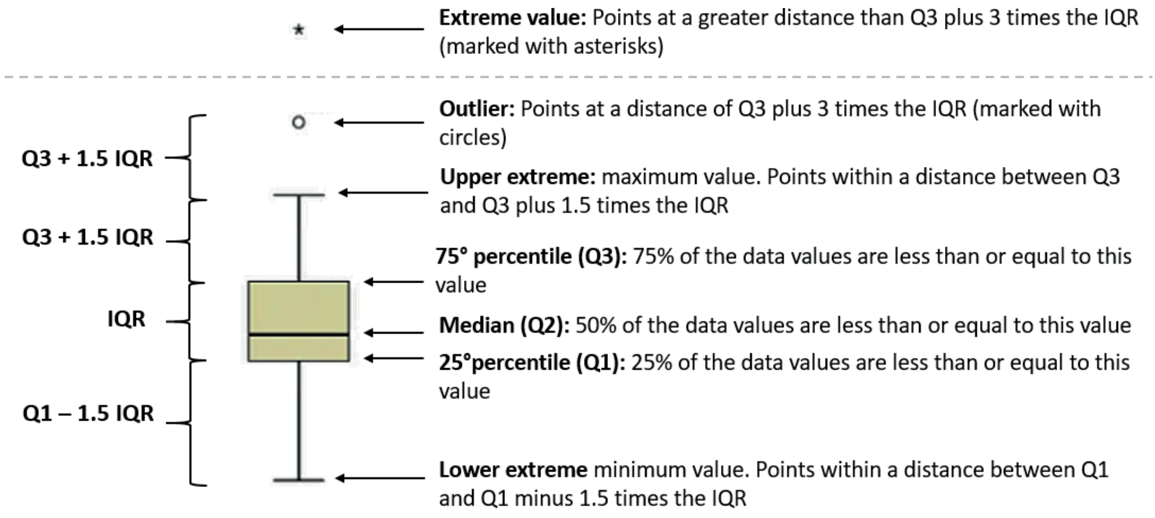


FIGURE 1: Structure of the box-plot chart.

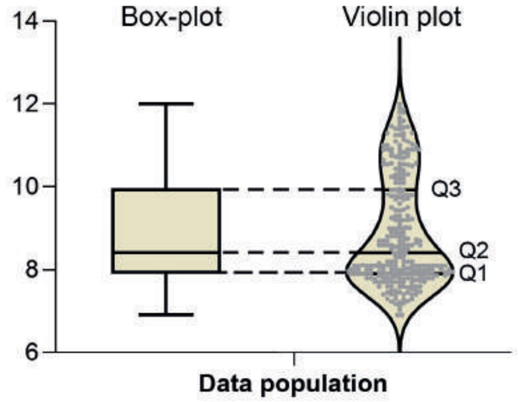


FIGURE 2: Structure of the violin plot chart.

of 95%. On the other hand, mobile plants (TP1 and TP3), equipped with less performing technologies compared to fixed plants, have lower efficiencies of 62% and 70% respectively.

As reported in Figure 3, the mixed construction and demolition waste (EWC 170904) is the main flow managed by the TPs, accounting for approximately 50%-80%. In accordance with the national situation, the CDWs delivered to the plants mainly derive from microdemolition operations of the residential building stock. In these activities, selective demolition is almost absent and, therefore, the CDWs produced are highly heterogeneous and classified as mixed construction and demolition wastes.

Soil and stones (EWC 170504) are the second most managed CDW stream, with about 11%-36%. This type of waste mainly derives from excavation works (i.e. removal of rocks and/or soils for new building construction or for existing building extension).

Finally, cement (EWC 170101) is the third most managed and treated flow, equal to 7%-14% of the total CDW amount. It mainly comes from the demolition of prefabricated industrial buildings.

At the same time, mixture of concrete, bricks, tiles and ceramics (EWC 170107), bituminous mixtures (EWC 170302) and track ballast (EWC 170508) are the least managed flows (less than 3%). All the other authorized EWC codes are delivered to TPs in quantities slightly greater than 0%.

TABLE 3: CDWs managed and RAs produced by the three TPs (from 2013 to 2018) and TP's recovery efficiency.

Treatment plant		Total CDWs managed (t)	RAs produced (t)	TP recovery efficiency (%)
TP1	TP1$_{,1}$	486,295	309,905	62
	TP1$_{,2}$	233,059	158,633	
	TP1$_{,3}$	98,167	49,342	
	TP1$_{,4}$	6,369	-	
TP2		238,650	231,340	95
TP3		229,915	160,580	70
Total		1292,455	909,8	

3.2 CDW treatment chains

3.2.1 CDW acceptance and management criteria

At the entrance to each treatment plant (TP1, TP2 and TP3), CDWs are subjected to acceptance procedures carried out by the following consecutive operations:

- Acquisition of the Waste Identification Form (called FIR in Italy): according to L.D. 152/2006, the CDW owner is obliged to trace the flows of CDWs produced in all transport phases, from the production site to the treatment plant. In this form the owner identifies the amount of CDWs produced, their EWC codes, the demolition building site and all the data of breaker, transporter and recipient (treatment plant).
- First visual inspection of incoming CDWs on trucks (by digital cameras) in order to verify the conformity of the material supplied to the plant with what was declared in the waste identification form.
- Assessment of CDW non-hazardousness by acquiring the chemical characterization analysis from the CDW owner. This operation is carried out so that the treatment plants are authorized to receive only non-hazardous CDWs.

The chemical parameters that must be analyzed by the producer are identified by accredited laboratories in relation to the type of CDWs and the local context in which the plant operates. In particular, for mixed construction and demolition waste (EWC 170904) the presence of asbestos must be detected.

Similarly, for CDWs identified by EWC codes 170302 "bituminous mixtures" (received by TP3) and 170508 "track ballast" (received by TP2), the PAH content and the ophiolite stone content (<0.1% by mass) must be respectively

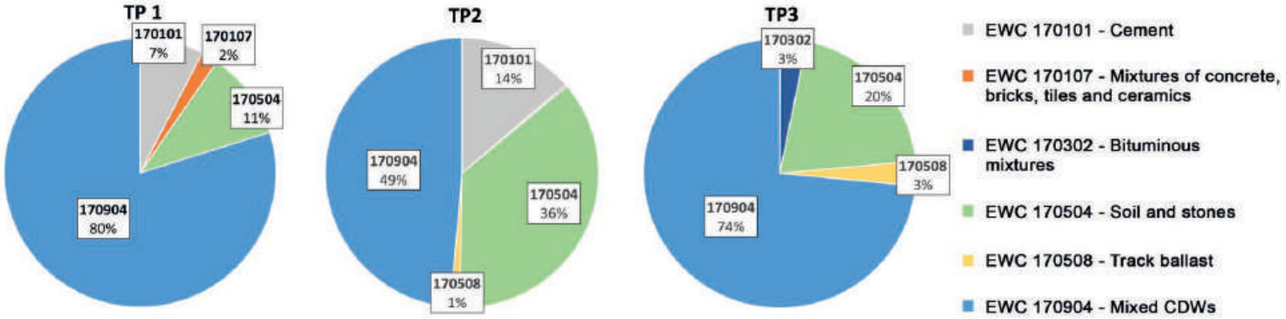


FIGURE 3: Type of CDWs mostly managed by the three TPs in the reference period (2013-2018).

detected. In particular, the ophiolite stone is a green rock, generally used for the construction of roads and railway sub-base layers, which can naturally contain asbestos fibers.

All these preliminary operations must be performed by the treatment plant for each CDW reception.

Only in the case that CDWs come from private micro-demolition (limited to EWC 170904) or from a well-defined and known technological cycle (sole producer), can the analytical non-hazardousness evaluation be performed by the treatment plant, respectively, every 6 months or when 500 m^3 of CDWs are reached. If non-compliance is determined on the heaps, the material must be sent to authorized disposal plants.

In any case, if the material does not comply with all the three above-mentioned conditions, the load is rejected. Conversely, the CDWs that are accepted must be weighed and stored on paved areas, according to EWC codes (R13 operation). This operation is mandatory and required by the plant environmental authorization to guarantee the traceability of the managed CDW flows. In this way, the inspection body in case of environmental non-compliance can trace the load of non-compliant material.

During the discharge in the storage areas, an additional visual quality control is performed to remove any unwanted coarse materials that cannot be treated by the system. Finally, all the CDWs stored using R13 recovery operation must end up in the final treatment process within 6 months of acceptance.

3.2.2 CDW treatment process

After the storage, the CDWs are sent to the mechanical treatment process, as shown in Figure 4. All 3 treatment plants have a single treatment line from which mixed recycled aggregates (MRAs) are obtained.

In the pre-treatment phase, the CDWs are fed to the treatment system by means of a crawler excavator, and the different EWC codes are mixed together. As confirmed by Borghi et al. (2018), the most common practice applied in the region is to mix them to obtain MRAs. This process is implemented since the market demand is exclusively directed to low-level applications (road and geotechnical ap-

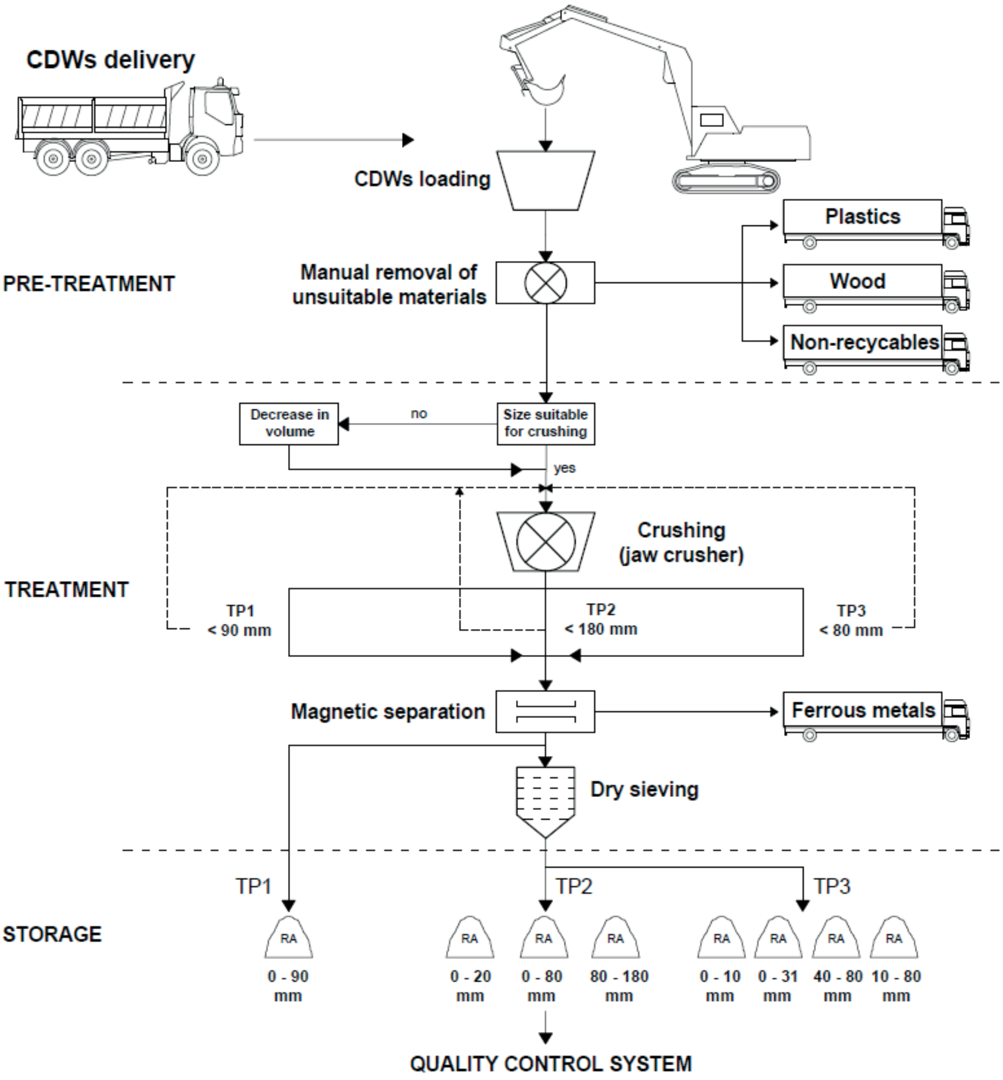


FIGURE 4: CDW treatment flow sheet.

plications) where MRAs are required. Treatment plants are therefore not incentivized to implement their own technologies for better-quality RA production which would require high economic costs. Moreover, this homogenization process is linked to the need to guarantee the environmental compliance of the RAs produced by reducing the pollutant concentrations. As shown in paragraph 3.3.2, some EWC codes contain high pollutant concentrations which, if treated individually, can generate RAs that do not comply with national environmental requirements (e.g. EWC 170101 "cement" releases high amounts of chromium or EWC 170802 "gypsum" releases sulphates).

At the same time, all the impurities (plastics, wood, glass, non recyclables fractions, etc.) are manually removed by operators and collected in roll-off containers to obtain final quality RAs. These wastes must be identified by EWC code 19 and then destined to authorized recovery or disposal plants.

In the subsequent treatment phase, according to the treatment plant analysed, the material is crushed by a jaw crusher to different maximum grain sizes (D):

- D<90 mm in TP1;
- D<180 mm in TP2;
- D<80 mm in TP3.

The oversize fraction (higher than 90 mm, 180 mm and 80 mm) is reprocessed in the crusher to further decrease the particle volume. Instead, the large CDW elements (Figure 5) that cannot be directly sent to the crushing phase are previously reduced in size using hammers and demolition grapples.

After crushing, in order to separate and remove all ferrous metals, the material is sent to the deferrization phase consisting of a powerful electromagnetic iron remover placed perpendicular to the treated material flow. The removed ferrous metals, coded with EWC 191001 "iron and steel waste", are deposited in a roll-off container and subsequently destined to authorized recovery plants.

After volumetric reduction and deferrization, the materials processed in TP1 are stored as RAs, while the materials processed in TP2 and TP3 are sent to the sieving phase. In particular, before being sent to the screening phase, as specified by the granted authorization, the material processed in TP2 must be subjected to leaching test (every 15 m^3), according to EN 12457-2. If the material complies with the national environmental regulatory limits (M.D. 186/2006), it can be sent to the sieving phase. Otherwise, it must be sent to authorized disposal plants.

FIGURE 5: CDW large elements with unsuitable size for direct crushing.

A dry sieve, with one or several sizes according to the end-user's demand, is used for granulometric selection and different type of RAs (all-in, fine, medium, and coarse fractions) are obtained in the 3 treatment plants, as shown in Table 4. In particular, to avoid the formation of dust during the sieving phase, water sprinklers and nebulizers are used.

TABLE 4: Type and grain size of RAs produced by the 3 treatment plants.

Treatment plant	RA type	Grain size (mm)	Uses
TP1	All-in	0 -90	Road and railway embankments and sub-base layers, civil/industrial yard foundation and sub-base layers, backfillings, remodeling works
TP2	Fine	0-20	Pipe coverings, environmental restoration
	All-in	0-80	Road and railway embankments and sub-base layers, civil/industrial yard foundation and sub-base layers, backfillings, remodeling works
	Highly coarse	80-180	Drainage systems, reinforcement of embankment layers (built on earthy, sandy and clayey ground)
TP3	Fine	0-10	Pipe coverings, environmental restoration
	Medium	0-31	Road and railway embankments and sub-base layers, civil/industrial yard foundation and sub-base layers, backfillings, remodeling works
	Coarse	40-80	
	Medium-coarse	10-80 (1)	

(1) *grain size obtained by mixing medium (0-31 mm) and coarse (40-80 mm) RAs.*

The fine RAs (0-10 mm and 0-20 mm) are generally used for pipe coverings and environmental restoration works (i.e revitalization of depleted quarries and/or landfills).

The medium and coarse RAs (0-31 mm, 10-80 mm and 40-80 mm), as well as the all-in fractions (0-80 mm and 0-90 mm) are mainly used for the construction of road and railway foundations, embankments and sub-base layers, the construction of civil and industrial yard foundations and sub-base layers, as well as for backfilling applications and remodelling works.

Finally, the highly coarse RAs (80-180 mm) are typically used for drainage layers and for the reinforcement of embankment layers built on earthy, sandy and clayey soils.

In the case that the end-user requests a selected grain size, RAs are reprocessed to obtain the required particle size.

As stated before, the RAs produced by the 3 TPs are MRAs currently used in low-grade applications.

In 2019, TP1 experimentally produced recycled concrete aggregates (RCAs) for internal use in concrete production. Unfortunately, at the regional and national level, there is no market demand for RCAs and, therefore, the company has stopped its production without future application developments.

Finally, according to the particle size, the RAs produced are stored in heaps located on natural draining surfaces. In particular, to avoid the dispersion of dust, an anemometer is positioned above the RA heaps to activate the wetting system if the wind speed exceeds 3 m/s. In case of wind above 10 m/s, the treatment process must be stopped.

3.2.3 RA quality control procedures

At the end of the entire treatment line, the RAs produced by the 3 TPs are subjected to a technical and environmental quality control system.

Since there are no national End of Waste criteria for CDWs in Italy, the RAs produced must be subjected to leaching test according to EN 10802:2013 (sampling procedure) and EN 12457-2:2004 (d<4mm; L/S=10 L/kg; contact time=24 h; demineralized water). Subsequently, the environmental compliance must be assessed with respect to the regulatory limit values of M.D. 186/2006. Based on the respective authorization, the TPs analyzed must verify the RA ecocompatibility on heaps of 1,000 m^3 (TP1), 15 m^3 (TP2) and 3,000 m^3 (TP3), for each grain size produced.

For both domestically and abroad marketing, RAs are subsequently subjected to EC marking according to EN 13242:2008 for their use in civil engineering work and road construction. If the RAs produced by the treatment process do not comply with the above technical standards, the material must be reprocessed or, if this operation is economically or technically unsustainable, sent to authorized recovery/disposal plants.

As required by the plant authorization, in addition to EC marking and leaching test, the RAs produced by TP2 and used for backfilling or remodeling applications must also be subjected to chemical characterization test and must fall whithin the limits indicated in column A or B, Table 1, Part IV, Title V, Annex 5 of the Italian Environmental Code (L.D. 152/2006). This legislation defines the Contamination Threshold Concentrations (CSC) in relation to soil use. Column A concerns green public areas and sites for residential use, while column B refers to commercial and industrial sites.

3.2.4 Economic performance evaluation of CDW treatment plants

Fixed and mobile treatment plants run on different types of power. As confirmed by studies in the literature (Borghi et al., 2018), TP1 and TP3 (mobile plants) are diesel-powered, while TP2 (stationary plant) is grid connected. Generally, the fixed treatment plants have a high operational efficiency and have more advanced and performing technologies than mobile plants (e.g. optimized crushing steps, more powerful magnets, vibrating screens for different grain size production). These plants commonly provide better quality RAs. Mobile treatment plants, typically diesel-fuelled, have the great advantage of lower operating and transport costs but they usually do not apply technologically advanced treatments (simple crushing steps followed by ferrous metal separation; the sieving phase is optionally adopted). These plants generally deliver low-quality RAs classified as all-in aggregates (i.e. mixture of coarse and fine aggregates).

To evaluate the economic performance of the treatment plants, Table 5 shows the diesel/energy consumption per tonne of RAs produced. As evidenced by the results, the stationary plant uses less energy than mobile plants. As shown in Figure 6, TP2 exploits 57% -71% less energy than TP1 and TP3, respectively.

This difference in consumption strongly affects the technical-economic performance of mobile plants which, without a clear market demand for RAs, are not incentivized to improve the quality of the RAs produced by implementing new advanced treatment technologies. But this is not only a critical issue for mobile systems as even the fixed system, without a market demand that requires quality RAs, is not motivated to implement its own technologies.

A more refined and advanced treatment process (more crushing steps, greater particle size separation, etc.) requires very high economic costs (in the processing, logistics and handling phases) which, to date, in Lombardy and Italy are not compensated by the market demand which still favors the use of natural aggregates.

3.3 Environmental and technical compatibility of CDWs and RAs

Besides the analysis on CDW and RA flows and the treatment chains currently available in the Porvince of Brescia, the present work includes a statistical evaluation of the environmental performance (chemical and leaching

TABLE 5: TP consumption per tonne of RAs produced.

Treatment plant	Typology	Power supply	Consumption	
			L/t$_{RAproduced}$	€/t$_{RAproduced}$
TP1	Mobile	Diesel	0.225	0.292
TP2	Fixed	Electricity	-	0.125
TP3	Mobile	Diesel	0.333	0.432

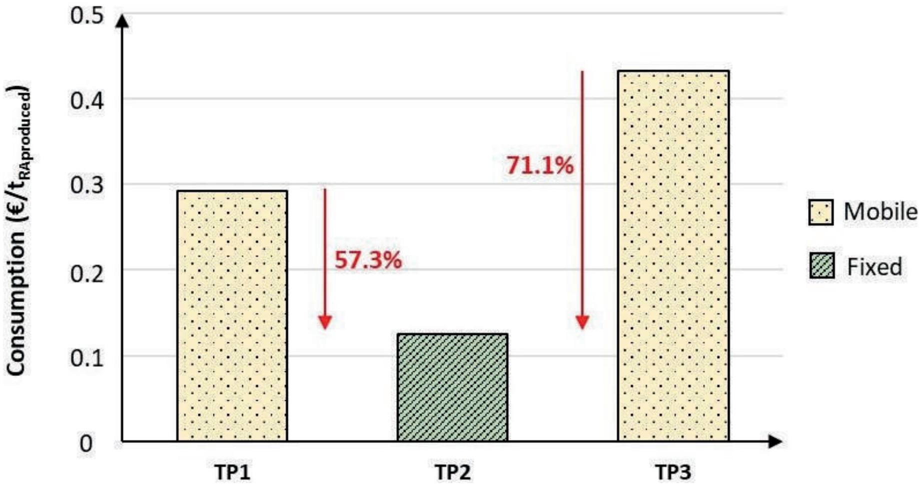


FIGURE 6: TP consumption per tonne of RAs produced.

properties) of both CDWs and RAs based on the quality control procedures carried out upstream and downstream of the treatment processes, as described above.

In this paper, as stated before, technical, chemical and leaching properties have been discussed in tabular format but the release of contaminants was also discussed by analyzing, with the box and violin plot methodology, the pH and total chromium.

3.3.1 Chemical properties

Table 6 shows the chemical composition of CDWs and RAs. Data obtained from the chemical certificates were reported on an average, minimum and maximum value basis and compared to the contamination threshold concentrations (CSC) listed in column A "green public areas and sites for residential use" and column B "commercial and industrial sites" of Table 1 of the L.D 152/2006.

According to Chen & Zhou (2020), the pH of CDWs is generally alkaline, with an average value of 8.5 and a maximum value of about 12. The highest pH values were mainly detected in EWC 170101 and EWC 170904 which have an intrinsically high content of cement. Aluminium is the most abundant element both in CDWs and RAs with an average content of 15.65 g/kg TS and 9.44 g/kg TS, respectively. These results agree with the data published by Butera et al. (2014) who detected a high aluminum content in CDWs equal to about 40 g/kg TS.

As regards CDWs, manganese and sulphates are minor non-critical elements with an average content of 831.7 mg/kg TS and 89.63 mg/kg TS, respectively. Mercury, tin and thallium are critical elements for the use in public and residential sites (average contents higher than 1 mg/kg TS), together with beryllium which showed an average content higher than 2 mg/kg TS. In particular, the maximum content of these elements was found in EWC 170904.

Total chromium, nickel, lead, copper, vanadium, and zinc are not critical parameters in terms of average concentrations, but the detected maximum values (between 350 and 5,300 mg/kg TS) are higher than the regulatory limits imposed by both column A and B of L.D. 152/2006. In particular, maximum values of total chromium and copper were detected in EWC 170904, while maximum values of nickel, lead, vanadium and zinc were detected in EWC 170504.

BTEX hydrocarbons (sum of benzene, toluene, ethylbenzene and xylene) are present in an average total concentration equal to 2.04 mg/kg TS exceeding the limit of column A (1.6 mg/kg TS). Specifically, benzene and xylene, with an average content of 0.53 mg/kg TS and 0.55 mg/kg TS respectively, are the critical compounds.

PCBs were detected in higher concentrations (range 0-3 mg/kg TS) than the regulatory limit of column A equal to 0.06 mg/kg TS. The light and heavy hydrocarbons content averaged 10 mg/kg TS and 850 mg/kg TS respectively and were above the regulatory limits of column A and B. In particular, the maximum value of heavy hydrocarbon (345,000 mg/kg TS) was detected in EWC 170302 "bituminus mixtures".

Regarding RAs, tin and benzene are the only critical elements with an average content higher than 1 mg/kg TS and 0.1 mg/kg TS (limits of column A), respectively.

For both CDWs and RAs, arsenic, antimony, barium, cadmium and selenium are present in average concentrations between 0.5 and 70 mg/kg TS, below the regulatory limits.

3.3.2 Leaching behaviour

As previously mentioned, the leaching behavior of CDWs and RAs was evaluated according to the compliance test imposed by UNI EN 12457-2 and compared with the regulatory limits of M.D. 186/2006 for unbound recovery. As for the chemical composition, the data obtained from leaching certificates were reported on an average, minimum and maximum value basis.

Comparing the CDW average leaching concentrations to the legal limits, mercury is the only critical parameter with an average concentration greater than 1 μg/L (Table 7). At the same time, the average concentrations of all the other parameters comply with the regulatory limits for both CDWs and RAs. Differently, the maximum values of

TABLE 6: Chemical composition of CDWs and RAs (pollutant contents reported in the average and minimum-maximum range) (TS: total solid).

Parameters	U.M.	CDWs			RAs			Limit values (L.D. 152/2006)	
		Min	Max	Mean	Min	Max	Mean	Column A	Column B
pH	-	2.90	11.95	**8.43**	-	-	**-**	-	-
Dry residue at 105°C	(%)	19.90	100	**93.70**	83.10	99.70	**95.87**	-	-
Aluminum	(mg/kg TS)	83.90	57248	**15651.8**	0.00	9442	**9442**	-	-
Antimony	(mg/kg TS)	0.01	102.48	**5.05**	0.30	10	**2.55**	10	30
Arsenic	(mg/kg TS)	0.05	82.90	**8.25**	0.30	15	**6.91**	20	50
Barium	(mg/kg TS)	1	482.50	**65.86**	-	-	**-**	-	-
Beryllium	(mg/kg TS)	0.01	20	**2.20**	0.20	10	**1.13**	2	10
Cadmium	(mg/kg TS)	0.01	22.60	**1.89**	0.10	10	**0.42**	2	15
Cobalt	(mg/kg TS)	0.02	105	**6.23**	0.50	10	**2.25**	20	250
Total chromium	(mg/kg TS)	0.10	7410.20	**53.49**	1.00	187.40	**31.90**	150	800
Exavalent chromium	(mg/kg TS)	0.10	50	**1.73**	0.10	10	**0.34**	2	15
Mercury	(mg/kg TS)	0	20	**1.55**	0.10	10	**0.37**	1	5
Molybdenum	(mg/kg TS)	0.10	20	**1.69**	-	-	**-**	-	-
Nickel	(mg/kg TS)	0.10	851	**20.95**	1	84.9	**15.94**	120	500
Lead	(mg/kg TS)	0.10	1589	**43.90**	6	98.66	**37.57**	100	1000
Copper	(mg/kg TS)	1	1490	**37.71**	1	100	**34.73**	120	600
Selenium	(mg/kg TS)	0	20	**1.33**	0.30	2.40	**0.79**	3	15
Tin	(mg/kg TS)	0.01	51.67	**3.66**	0.45	10	**1.37**	1	350
Thallium	(mg/kg TS)	0.01	20	**1.66**	0.50	1	**0.96**	1	10
Tellurium	(mg/kg TS)	0.05	10	**2.00**	-	-	**-**	-	-
Vanadium	(mg/kg TS)	0.5	355.80	**67.72**	0	39	**19.20**	90	250
Zinc	(mg/kg TS)	1	5288	**109.75**	13.90	145	**81.67**	150	1500
Manganese	(mg/kg TS)	0.10	2010	**831.70**	-	-	**-**	-	-
Cyanide	(mg/kg TS)	0.02	2	**0.89**	1	1	**1.00**	1	100
Sulfates	(mg/kg TS)	5	160	**89.83**	-	-	**-**	-	-
Nitrates	(mg/kg TS)	1.40	54	**15.78**	-	-	**-**	-	-
Chlorides	(mg/kg TS)	5.10	52.50	**15.78**	-	-	**-**	-	-
Benzene	(mg/kg TS)	0.003	10	**0.53**	0.01	0.02	**0.19**	0.10	2
Ethylbenzene	(mg/kg TS)	0.003	10	**0.48**	0.01	0.02	**0.19**	0.50	50
Toluene	(mg/kg TS)	0.003	10	**0.48**	0.01	0.02	**0.19**	0.50	50
Xylene	(mg/kg TS)	0.003	10	**0.55**	0.01	0.02	**0.19**	0.50	50
PAHs	(mg/kg TS)	0.00865	10	**0.75**	0.01	1	**0.09**	10	100
PCBs	(mg/kg TS)	0	2.77	**0.45**	0.01	0.01	**0.01**	0.06	5
Light hydrocarbons (C≤12)	(mg/kg TS)	0.01	285.20	**10.22**	-	-	**-**	10	250
Heavy hydrocarbons (C≥12)	(mg/kg TS)	0.50	345000	**850.65**	10	101	**26.36**	50	750
Hydrocarbons C10-C40	(mg/kg TS)	1	19583	**218.82**	20	20	**20.00**	-	-
Asbestos	(mg/kg TS)	0	<1000	**343.65**	<100	<1000	**118.37**	1000	1000

released contaminants often do not comply with the regulatory limits. In fact, nitrates, sulphates, copper, total chromium, lead, selenium, mercury, and COD were detected in higher concentrations than the limits imposed by M.D. 186/2006 in both CDWs and RAs.

Higher maximum concentrations of fluorides, chlorides, nickel, vanadium, and cadmium were also detected in CDWs.

Figure 7 shows the results of the statistical analysis developed on pH and total chromium. The analysis was performed by evaluating: (a) the release of contaminants from CDWs and RAs and (b) the release of contaminants according to the three selected EWC codes (EWC 170101, EWC 170504 and EWC 170904). In order to identify the critical pollutants for recovery and final use, the variability range of pollutant concentrations was assessed and com-

TABLE 7: Leaching behaviour of CDWs and RAs (pollutant releases reported in the average and minimum-maximum range).

Parameters	U.M.	CDWs			RAs			Limit values (M.D. 186/2006)
		Min	Max	Mean	Min	Max	Mean	
pH	-	2.20	12.20	**8.61**	7.03	12	**8.89**	5.5-12
Nitrates	(mg/L)	0.05	95.20	**5.09**	0.10	73.30	**10.28**	50
Fluorides	(mg/L)	0.01	7.50	**0.44**	0.01	1.21	**0.27**	1.5
Sulphates	(mg/L)	0.40	1613	**103.47**	1	615	**55.76**	250
Chlorides	(mg/L)	0.08	531	**14.93**	1	57.70	**10.99**	100
Cyanide	(µg/L)	0	50	**10.72**	0.05	50	**13.77**	50
Barium	(mg/L)	0	1	**0.09**	0.01	0.50	**0.08**	1
Copper	(mg/L)	0	0.22	**0.01**	0.0002	1	**0.02**	0.05
Zinc	(mg/L)	0.0001	2.80	**0.17**	0.001	1.64	**0.12**	3
Berillium	(µg/L)	0	10	**1.70**	0.10	10	**2.23**	10
Cobalt	(µg/L)	0	67	**5.67**	0.10	30	**5.49**	250
Nickel	(µg/L)	0.01	190	**6.47**	0.10	10	**3.82**	10
Vanadium	(µg/L)	0	643.50	**20.20**	0.015	94	**15.52**	250
Arsenic	(µg/L)	0.12	0.13	**7.55**	0.10	41	**5.63**	50
Cadmium	(µg/L)	0	10	**1.36**	0.10	5	**1.95**	5
Total chromium	(µg/L)	0.01	1361	**19.83**	1	183	**18.97**	50
Lead	(µg/L)	0.02	5111	**32.97**	0.10	79	**5.76**	50
Selenium	(µg/L)	0	84	**3.99**	0.10	10	**3.85**	10
Mercury	(µg/L)	0	50	**1.01**	0.10	1.62	**0.60**	1
Asbestos	(mg/L)	0	30	**3.98**	0	30	**2.61**	30
COD	(mg/L)	1	443	**25.48**	5	43.30	**18.35**	30

pared with the regulatory limits.

According to the box-plot results, the pH of both CDWs and RAs was generally alkaline with 50% of values ranging from 7.5 to 10. As shown by the violin plots (Figure 8), which reproduce the real distribution of data density, the pH values showed a peak also in the range from 11 to over 12, slightly exceeding the national regulatory limit.

As recently studies reported (Coudray et al., 2017; Gupta et al., 2018; Ng & Engelsen, 2018; Saca et al., 2017), this strong alkalinity is mainly due to the cement and concrete fractions contained in both CDWs and RAs. In agreement with these considerations, as shown in Figure 7 (b), EWC 170101 "cement" had the highest pH values with a median of 11.3 and highly left-skewed data.

Lower pH values were observed for mixed CDWs (EWC 170904) and soil and stones (EWC 170504) leachates (median values of 8.78 and 8.11 respectively). As stated by Butera et al. (2014) this could be ascribed to the presence of bricks and soil in the sample mixtures.

Total chromium is generally one of the critical compounds in CDW and RA leachates (Butera et al., 2014; Del Rey et al., 2015; Delay et al., 2007; Maia et al., 2018). According to the box and violin plot results, the highest chromium release was found in RAs in the range of 2.5 – 62.5 µg/L, exceeding the regulatory limit. As for the pH, the high chromium release was mainly due to the cement fractions contained in EWC 170101, in which over 50% of the total concentrations exceeded the recovery limit of 50 µg/L. On the other hand, as Galvín et al. (2014); Zhang et al. (2018) exposed, chromium has high leachability at alkaline pH (pH>10), which confirms the higher release compared to soil and stones (EWC 170504) and mixed CDWs (EWC 170904).

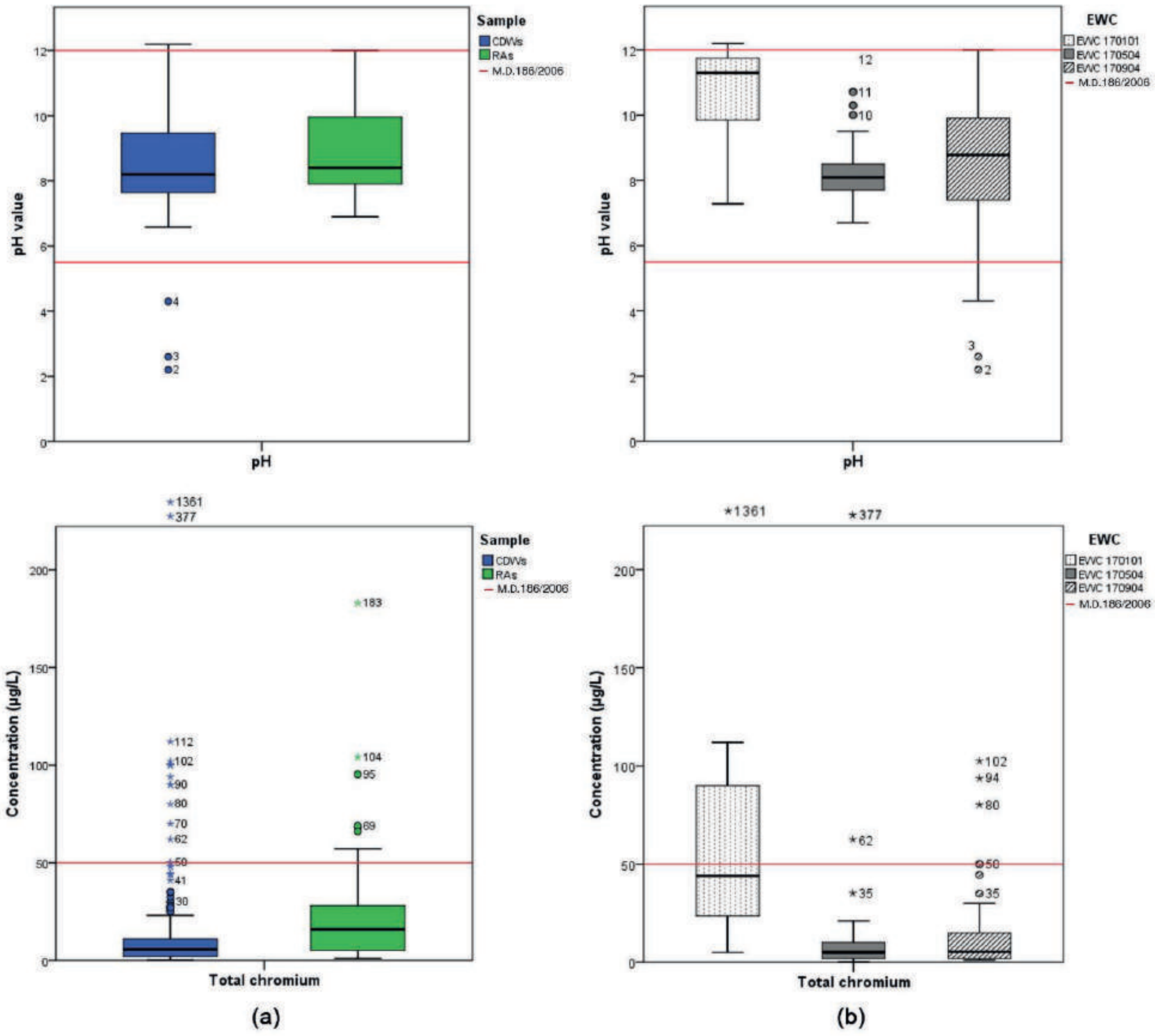
3.3.3 Technical performance of RAs

Table 8 reports the technical properties of RAs produced by the 3 treatment plants. As mentioned above, data refer to the EC marking tests performed on RAs (fine, medium and coarse) according to UNI EN 13242 for their use in civil engineering work and road construction.

According to the results, the RA particle density ranges from 2150 kg/m^3 to 2810 kg/m^3, with an average value of 2480 kg/m^3. In particular, the highest density values were detected in coarse RAs. In terms of resistance to fragmentation (Los Angeles), the values range from 29 to 40. Compared to natural aggregates, RAs are similar to friable rocks. Regarding water absorption (WA), WA of coarse recycled aggregates varies between 0.6% and 7%, while WA of fine RAs can reach values up to 9.6%. These data confirm the results obtained by other authors (Vieira et al., 2016). Finally, water-soluble chloride and acid-soluble sulphates show average concentrations of 0.066% and 0.4%, respectively.

3.4 Barriers and possible strategies for CDW recovery

Based on the analysis developed, this study has investigated the barriers to CDW recovery and the possible



(a) (b)

FIGURE 7: CDW treatment flow sheet.

strategies to overcome these hurdles. As shown in Figure 9, the main barriers identified are classified into 3 categories: economic, logistical/technical and regulatory/cultural.

The main obstacle concerns the propensity of end users to use, for high-grade applications, natural aggregates (cheaper and with high technical-environmental performance) rather than RAs which "derive from wastes". The lack of consumer awareness and consequently the commercial disadvantage for companies is therefore the main obstacle to recovery. In this context, the absence of a market demand for good quality RAs (e.g. RCAs) directs recycling plants to produce only mixed recycled aggregates. Thus, the implementation of advanced treatment technologies is not performed since the economic costs are too high and are not compensated by the market demand. This hurdle is recognized as a "not foreseen market opportunities" for which the company does not yet identify any added-value in further qualifying their products and services. Furthermore, in the regional and national territory, there are no economic incentives for recycling plants.

In terms of logistical and technical hurdles, also the sampling procedure prescribed by the EN 10802 for the evaluation of the RA leaching behaviour is considered not representative, as directly confirmed by companies. In particular, the quantity of sampled material, equal to a few grams, can not be considered representative of the heaps consisting of thousands of m^3. Similarly, the leaching test method defined by UNI EN 12457-2, which requires a maximum particles diameter equal to 4 mm, is not considered representative of the real RA use conditions (diameter >4mm).

To overcome these barriers, governmental and scientific research involvement are necessary to improve CDW recovery and RA end-use. Here, some levels for change are identified:

- Economic and policy instruments: increased recycling could be achieved by economically viable measures to improve waste management. In particular, the compet-

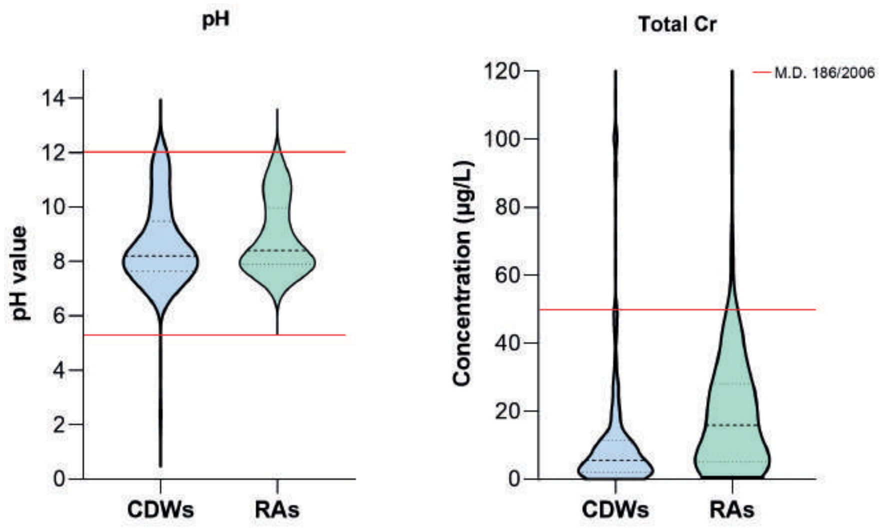


FIGURE 8: Statistical analysis: violin plots of pollutant concentrations (pH and total chromium) in CDWs and RAs.

itiveness of recycled materials could be increased by raising the price of natural aggregates and taxing the mining activity. In addition, economic incentives (tax credit) should be provided by national authorities for companies who produce recycled products.

- Education policies: recycled materials derived from wastes are still perceived to be inferior to natural ones. Policy interventions that spread awareness and knowledge of these new materials are therefore essential. In particular, public information and education campaigns should be launched and best practice should be promoted in order to clarify that the technical and environmental performances are comparable to those of natural aggregates. These actions would stimulate the market demand for RAs, even of better quality.
- Improvement of technologies: as above-mentioned, the increase in the use of recycled aggregates depends on their ability to be competitive with virgin raw materials in terms of both cost and quality. Enhancing the treatment process by implementing new advanced technologies may increase costs but the greater confidence in the use of RAs and the consequent market demand would allow to offset these costs. The research and development policies may impact the improvement of developed technologies and recycled materials produced. In particular, in the case studies analyzed, the commitment of the three companies is to evaluate the use of RCAs for more noble applications (i.e. concrete production). To achieve and promote this, they have launched research activities in collaboration with the

TABLE 8: Technical properties of RAs produced by the 3 TPs (values reported in the average and minimum-maximum range).

Property	**U.M.**	**RA**			**RA_{fine}**			**RA_{medium}**			**RA_{coarse}**		
		Min	**Max**	**Mean**	**Min**	**Max**	**Mean**	**Min**	**Max**	**Mean**	**Min**	**Max**	**Mean**
Particle size	d/D	0	180	-	0	10	-	0	31	-	0	180	-
Flakiness index	FI	18	35	**27**	-	-	-	-	-	-	18	35	**27**
Shape index	SI	20	40	**30**	-	-	-	20	40	**30**	20	40	**30**
Fines content	f	2	9	**6**	3	3	**3**	3	3	**3**	2	9	**6**
Sand equivalent value	SE	20	85	**53**	47	85	**66**	48	48	**48**	20	46	**33**
Methylene blue test	MB	0	4	**2**	0.3	0.6	**0.5**	-	-	-	2	3.9	**3**
Particle density	kg/m^3	2150	2810	**2480**	2150	2790	**2470**	2291	2756	**2524**	2282	2810	**2546**
Bulk density	kg/m^3	2321	2940	**2631**	2350	2786	**2568**	2446	2840	**2643**	2321	2940	**2631**
Los Angeles coefficient	LA	29	40	**35**	-	-	-	29	35	**32**	29	40	**35**
Micro-Deval coefficient	MDE	35	41	**38**	-	-	-	-	-	-	35	41	**38**
Water absorption	%WA	0.4	9.6	**5**	1	9.6	**5**	0.4	5.3	**3**	0.6	7	**4**
Freeze/thaw resistance	F	1	22	**12**	1	1	**1**	-	-	-	10	22	**16**
Alkali-silica reactivity	-	Non-reactive			Non-reactive			Non-reactive			Non-reactive		
Water-soluble chloride	%	0.0018	0.01	**0.006**	0.006	0.006	**0.006**	0.0018	0.01	**0.006**	-	-	-
Acid-soluble sulphate	AS	0.01	0.8	**0.4**	0.2	0.2	**0.2**	0.01	0.2	**0.1**	0.036	0.8	**0.4**
Total sulfur	S	0.01	1	**0.5**	0.01	1	**0.5**	0.01	1	**0.5**	0.14	1	**0.6**

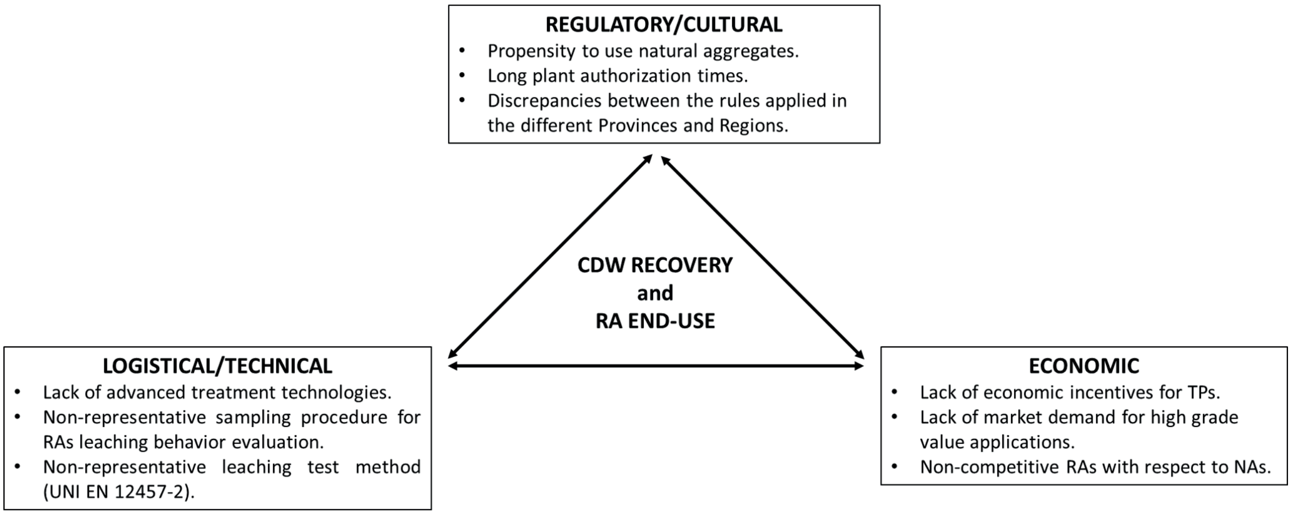


FIGURE 9: Barriers to CDWs recovery.

university to experimentally investigate the feasibility of RCAs use.

- Research activities: development of targeted research activities that re-evaluate the sampling methods and simulate the release of pollutants from RAs in conditions of on-site use (i.e. without altering the grain size).

4. CONCLUSIONS

In this study, the management and operational data of three CDW treatment plants located in the Province of Brescia (the Lombardy Region) were analyzed. The aim of the study was to understand and define the current CDW treatment chains adopted in the Region in order to highlight barriers that still limit CDW recovery and possible strategies to overcome these hurdles.

The following conclusions can be drawn as a result of the research:

- EWC 170904, EWC 170504 and EWC 170101 are the most managed CDW flows in the area. In particular, 50%-80% of the CDWs managed is represented by EWC 170904.
- Instead, EWC 170504 and EWC 170101 represent approximately 11%-36% and 7%-14% of the total CDWs managed.
- The average recovery efficiency of the treatment plants is strongly related to the technology adopted by the plants. The fixed plant, equipped with performing technologies, has the highest recovery efficiency of 95%. Instead, the mobile plants, equipped with less performing technologies, have lower efficiencies of 62% and 70%, respectively.

CDW treatment plants are located in natural aggregate excavation quarries which, over time, have adapted their work sites and technologies to the treatment and recovery of CDWs. The treatment techniques applied, are in fact borrowed from those already used to produce natural aggregates and consist of technologically interconnected phases of crushing, separation of ferrous metals and unwanted materials and dry sieving. The MRAs produced are of medium-low quality and are mostly used in road and geotechnical applications. In order to obtain high quality RAs, which can be used for high value applications, the selective demolition and the improvement of source separation is highly recommended. Upstream of the construction process, it is also essential to encourage the eco-design of products, in order to promote the industrialization of disassembly, the separation of single components and the recycling of end-of-life materials. Moreover, the current treatment technology implementation (e.g. washing systems for salt removal such as sulphates and chlorides, air classifiers for lighter fraction removal) through economic incentives provided to CDW treatment plants would allow for more efficient recycling and higher quality recycled materials.

Finally, to evaluate the potentially critical parameters for recovery and final use, the study included an analysis of the chemical and leaching properties of both CDWs and RAs.

The chemical composition results show that the pH of CDWs is generally alkaline and alluminium is the most abundant element both in CDWs and RAs. Comparing the average chemical contents to the legal limits of L.D. 152/2006, tin and benzene are the most critical elements for both CDWs and RAs with average contents higher than the limits of column A.

Comparing the leaching results with regulatory limit values of M.D. 186/2006 led to the following conclusions:

- the CDW and RA pH is generally alkaline and mainly related to the cement and concrete fractions contained in EWC 170101;
- mercury was identified as a critical compound in CDW leachates;
- total chromium was identified as a critical element both in CDWs (EWC 170101) and RAs in comparison with available regulatory limits. Also sulphates, with a maximum concentration of over 1600 mg/L, were identified as a critical parameter in CDWs and mainly releated to ceramic fractions contained in EWC 170904.

In this context, as emerged from the analysis developed, the environmental compatibility of these materials should be evaluated by adopting sampling procedures representative of the real quantities of materials analyzed (heaps of thousands of cubic meters) and leaching test methods representative of the real RA use conditions (d>4mm).

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CISA

VALORIZATION OF RARE EARTH ELEMENTS FROM END-OF-LIFE FLUORESCENT LAMPS: A CONTRIBUTION TO URBAN MINING

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ABSTRACT

In recent decades, Rare Earth Elements (REEs) have assumedfundamental place in the electrical and electronic (EE) industry as a result of the increasing interest in low-energy and intelligent technologies. Availability of those elements in limited area of the globe, the complexity of extraction processes and the high costs of their valorization negatively influence the supply chain to such an extent as to jeopardize the future offer of EE equipment. This issue is particularly acute in Europe where all REEs are imported, particularly from China that has a dominant position in the global market. To contrast this dependence and ensure a stable future demand, industrial stakeholders have embarked on an ambitious path aimed at recovering REEs from EEE waste. Indeed, the promotion of policies and measures for a circular economy has identified in urban mining the way to address this challenge.Cities are considered as a reserve of minerals, although applied research is still in its infancy and currently only 1% of REEs is recovered. Whilst handling waste, fluorescent lamps proves mainly challenging due to the presence of hazardous substances; however, they yield the highest purity rare-earth oxides. This article represents a preliminary multi-criteria analysis aimed at assessing the feasibility of launching an urban mining project based on the valorization of REEs from fluorescent lamps. The work is the result of the activities undertaken by the Italian WEEE company DISMECO in collaboration with the University of Bologna, paving the way for a more profitable circular economy for REEs.

1. INTRODUCTION

The well-known impacts generated by the linear economy on the environment, the problems deriving from resource scarcity, and the increasing awareness of critical climate change-related issues, have nurtured the rethinking of production and consumption patterns by adopting green processes and products (Beamon, 1999; Wong, 2012; Gadimi et al, 2019). Recycling and secondary raw materials can massively contribute to reducing carbon emissions, and therefore, to ensure a sustainable value chain and promote a more circular economy model (Zorpas, 2016). In a life cycle perspective, the valorization of the intrinsic value of materials at the end-of-life (EoL) has been a key element for reducing the manufacturing industry's carbon footprint. The life-cycle assessment (LCA) studies recently performed by Ibanescu et al. (2018) on five European WEEE management systems reveal a net negative total carbon footprint, thus showing the significant contribution of recycling in the WEEE (Waste from Electrical and Electronic Equipment) treatment options. Additionally, during the materials extraction and processing phase, recycling, and in particular upcycling, massively contributes to minimizing CO_2 emissions thanks to the reduction of virgin materials (Lieder et al., 2017; Nascimento et al., 2019; Riesener et al., 2019; Clarke et al., 2019).

1.1 The role of recycling in the EU high-tech industry

Recycling assumes higher relevance in Europe, where the reserves of oil, metals, and resources in general, are insufficient to meet the local demand. The presence of the so-called Rare Earth Elements (REEs) is confined to the Finnish country, holding the Cobalt market while the French region is the leading Hafnium supplier (European Commission, 2018). All the other materials are basically imported. In particular, Europe is highly dependent on China for the supply of "high-tech" metals such as Cobalt, Platinum, Rare Earths, and Titanium. Moreover, a residual business is controlled by the USA (especially for Beryllium and Helium) and Russia, which contains the business transactions

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for Cobalt and Scandium's reserve (European Commission, 2018). The economic aspect is relevant in the electrical and electronic (EE) sector, which is characterized by massive use of Gold, Copper, Silver, Titanium and REEs. In 2016, the EE industry registered a world market share of 14,6% (Oladele et al., 2009; Li et al., 2015). The main applications include refrigerators, illumination devices, mobile phones, tv, computers, and general high-tech equipment. Nowadays, the growing interest in low-carbon (solar panels, wind turbines and batteries etc.) and smart technologies (internet of things, cloud manufacturing, advanced automation etc.) has affected the demand for these materials that are expected to increase by a factor of 20 by 2030 (European Commission, 2016). Although the EE market represents an opportunity for a more competitive Europe, issues related to REEs supply are severe, especially for Europium (Eu), Neodymium (Nd), Terbium (Tb), Dysprosium (Dy), and Yttrium (Y) (Binnemans et al., 2013).

To ensure the survival of the Information and Communication Technologies (ICT) sector, the European (EU) Commission launched the European Raw Materials Initiative aimed at identifying those materials that have already reached or exceed the natural stocks (Baccini and Brunner, 2012; Bakker, 2012). The reasons why these materials are particularly critical are:

- the significant economic importance for key emerging sectors
- the increasing demand that is not ensured by the local availability and therefore contributing to supply risks, price volatility, and strict dependence from importing countries
- the lack of valid alternatives from the economic and technical point of view.

According to the variables mentioned above, the list is continuously monitored and updated in the EU Raw Materials Information System (Bio by Deloitte, 2015). The last analysis performed by UNEP reveals that less than 1% of REEs are globally recycled (UNEP, 2011). In Europe, even if the collection rate of household WEEE amounted to 492,532 tonnes in 2018, it was insufficient to meet the collection target established by the Directive 2012/19/EC entered into force on 13th of August 2012. The directive introduced stepwise higher collection targets to be applied from 2016 and 2019 (European Commission, 2012).

Consequently, major efforts are needed to be competitive in the global market. The circular economy covers a pivotal role in fostering this transition. The Circular Economy action plan, published by the Commission in 2015 and updated in 2019, represents the key innovative legislation in this field. The circular economy roadmap contains, among the most urgent sectors of intervention, actions aimed at closing the loop of electronics, ICT, and batteries industries (European Parliament and of the Council, 2015). Indeed, the Directive 2018/849/EC (2018b) broadens product types that fall in the compliance scheme and follows a separate collection process and special treatment.

1.2 Urban mining: an opportunity for a flourishing market of REEs in Europe

Among the circular strategies, recycling is the most exploited waste treatment (Price, 2000). While material valorization is well-established in some value chains such as plastics, glass and paper, the recycling of precious metals and REEs is still an embryonic process due to small contents and the difficulties in the extraction and the separation processes (Fellner et al., 2017). Problems are also related to the mis-collection of EE waste. To highlight the importance of monitoring and extracting value from materials in urban areas, the concept of urban mining has been introduced (Krook and Eklund, 2010). Even if a unique definition of urban mining doesn't exist, this emerging research trend aims to consider cities as potential reserves of metals and minerals to such a point that they are called urban mines (Sun et al., 2016; Ciacci et al., 2017; Bonoli et al., 2018).

From the urban mining perspective, producing vast quantities of wastes may represent a flourishing market for recyclers operating in the field of Copper, Gold, Silvers, and Critical Raw Materials (CRMs). As pointed out by the EU Commission, this practice may strongly contribute to the proper management of dangerous substances (European Commission, 2018). Moreover, in contrast to open burning and illegal treatment, mainly adapted in emerging countries, the process of disassembly, isolate and purify materials represents an excellent alternative to the linear economy. These operations are based on multiple value capturing principles, with positive economic, social, and environmental implications. However, implementing a systemic reuse system from urban infrastructures and buildings is challenging (Brunner, 2011). A comprehensive information dataset on flows and stocks is essential. It assumes higher importance if we consider that more than 40 million metric tons of WEEE are generated every year (Forti et al., 2020). As reported in the Waste Hierarchy and highlighted by Cossu and Williams (2015), urban mining should be contextualized in circular thinking. The life cycle approach should be adopted to extract the highest value from materials and ensure a competitive market for secondary materials. It follows that WEEE and relative materials become a source of revenue to be perceived as an opportunity for the growth of sustainable high-tech industry, the promotion of green jobs, and the adoption of advanced labor skills (Krooks and Baas, 2013). It is imperative especially when it comes to CRMs, that are sensitive to the supply chain's reliability. Among various applications, materials recovery from lamps represents one of the significant opportunities for experimentation.

No studies have been carried out yet on the recovery of REEs from small fluorescent lights used in Liquid-Crystal-Display (LCD) backlights or from phosphors used in white Light-Emitting-Diodes (LEDs) (Buchert et al., 2012). This work represents a preliminary multi-criteria analysis aimed at assessing the feasibility of launching an urban mining project based on the valorization of REEs from fluorescent lamps.

2. THE VALUE OF RARE EARTH ELEMENTS IN CIRCULAR ECONOMY

Rare Earth metals (and relative oxides) are widely used in EE applications. REEs are 17 elements, 15 of the Lantanoid series over than Scandium and Yttrium. REEs are categorized into "Heavy Rare Earths" and "Light Rare Earths." Lanthanum, Cerium, Praseodymium, Neodymium, Promethium, and Samarium are the Light Rare Earths while Yttrium, Europium, Gadolinium, Terbium, Dysprosium, Holmium, Erbium, Thulium, Ytterbium, and Lutetium can be considered Heavy Rare Earths (de Boer and Lammertsma, 2013). Despite the adjective "rare", which is mainly linked to the difficulties in the extraction process, REEs are widely present in the EEarth's crust. From a mineralogical point of view, REEs are considered "trace elements" because of their presence in some minerals that are not very abundant in common rocks (carbonates, oxides, phosphates, and halides). REEs are also characterized by a deficient concentrations in the reservoirs. They exist in a conglomerate form that implies complex and expensive extraction and separation processes. It follows that the economic exploitation of the deposits is profitable only in a few areas of the Earth, where the largest Rare Earths reserves are present, i.e. China, Brazil, Russia, India, Australia, Greenland, and the USA. Among them, China is the crucial player controlling more than 90% of the global market, thus achieving a substantial monopoly. The vital power of the Chinese EE industry on the global REEs market is strongly related to its business. Chinese-based companies have gained supremacy in the market since the 1980s thanks to their suddenness in reaching REEs growing demand through very low-price policy based on high volume supply and labor exploitation. This supremacy has never been jeopardized. Nowadays, all REEs' global reserves are estimated to be about 114 Mt, mainly located in China (Deboer and Lammertsma, 2013). China turns out to be the largest producer and exporter and consumer of Rare Earths, followed by USA and Japan.

To prevent possible future shortages without exceeding the extraction activity, EU countries, and specifically Italy, have to enhance a robust recycling activity, consisting mainly of WEEE proper management and valorization. In addition to self-sufficiency, recycling represents a valid option from an environmental point of view. However, many critical issues remain. The REEs processing is expensive, energy and resource-consuming: during the extraction and the mineral valorization phases, a large amount of water and chemicals is used, and at the same time, atmospheric releases are high and, vast amounts of solid waste and wastewater are generated. It has been estimated that the processing of a tonnage of Rare Earth metals involves the generation of about 2.000 tons of toxic waste. Furthermore, REEs are often associated with some radioactive materials, affecting the quality of the residual slags and emissions with risks for the ecosystems.

Despite the enormous environmental impact on the mining of REEs, the demand for REEs is growing. The industry is rapidly advancing with new and innovative electric devices and newly developed energy systems that increasingly need REEs' application on a large scale. In particular, Praseodymium, Neodymium and Dysprosium are widely applied in permanent magnets production (for wind turbines, electric cars, speakers, hard disks). Erbium, Holmium, and Gadolinium are commonly used in medical-surgical lasers. Terbium, Yttrium, and Europium are supplied for the lighting technology industry manufacture lamps and flat-screen luminophores. More generally, all REEs are used in a multitude of applications, including high-tech products, such as special steels and alloys, pigments, optical fiber and glass, rechargeable batteries, microwave devices, space and aircraft engines, components for satellites, catalysts for the petrochemical and polymer industry, catalytic converters, very high power lasers, superconductors, nuclear batteries, nuclear reactors, anticancer drugs, substances and sensors for Magnetic Resonance and Positron Emission Tomography diagnostics. They are also handled in electronic and IT components with high miniaturization, nanotechnologies, high-intensity lighting, solar cells, night vision devices.

The fluorescent lamp production can be considered the significant goods daily sold among the small electronic devices. As defined within the Directive 244/2009/EC (2009), fluorescent lamp means "a discharge lamp of the low-pressure Mercury type in which most of the light is emitted by one or several layers of phosphors excited by the ultraviolet radiation from the discharge". Fluorescent lamps are supplied either with or without integrated ballasts; Lanthanum, Terbium, Yttrium and Europium and their oxides are used as internal coating powders to filter the UV radiation generated by exciting the internal gas and resulting in a visible white light. In Figure 1 a typical fluorescent lamp and the lamps storage at the investigated plant.

FIGURE 1: A common fluorescent lamp and storage of lamps at investigated plant.

Fluorescent lamps were launched on the market at the end of the 1980s as an advantageous alternative to incandescent bulbs. The fluorescent lamps' benefits can be summarized in lower energy consumption, long life span, and superior light output. The demand robustly increased after the eco-design requirements established by the EU Directive 244/2009. The limited life span and the fast substitution have fostered a large volume of waste to be managed. Furthermore, because of some hazardous components, such as Mercury in tube light, a specific collection and treatment process is required. Therefore, the focus on this type of waste hasgrown over time due to the rising attention on environmental protection practices and the interest in recovering essential and strategic minerals, including REEs. This attention was also registered in the EU policy and decision-making process through the establishment of the Directive 2002/95/EC (the so-called RoHS 1 Directive) and the Directive 2011/65/EU (the so-called RoHS 2 Directive) that prohibited specific substances for EEEs marketed in the EU.

Fluorescent lamps can be tubular or compact. The so-called "neon tubes" can be linear or circular. They are identified with the following standard diameters: T12 (diameter 38 mm, the old fluorescent tubes), T8 (diameter 26 mm), the most used in indoor environments, and T5 (16 mm diameter), the new generation of fluorescent lamps, with reduced dimensions and electronic power supply, hence excellent energy efficiency. Light intensity and color mainly depend on the internal coating of fluorescent powders which can be obtained by phosphorus trioxide (triphosphorous) and phosphorus pentoxide (pentaphosphorus). Because of their composition, outdated fluorescent lamps represent an actual and precious urban reserve of materials. It is possible to recover glass, metals, minerals and especially heavy and light REE. Three of the rare earths generally included in lamps (Europium, Terbium, Yttrium) are considered CRMs (Binnemans, 2014). The kind of recycling process depends on the type of minerals. Some authors (Kumar, 2016) suggest adopting a chemical treatment such as the acid dissolution for oxides (i.e for Yttrium recovery). Following this process, the phosphorous powders from fluorescent lamps are treated using a leaching process and an extraction process with solvent called solvent extraction. The most efficient process (Innocenzi, 2017) consists of a multistage liquid-liquid extraction with cross-currents between the aqueous solution and an organic extracting solution in acid conditions by adding an oxalic acid. Yttrium is recovered as Oxalate Hydrate with a purity of around 97,5%. To avoid chemicals utilization, a new methodology for the recovery of Yttrium and Europium has been proposed (Van den Bogaert, 2015). The process is based on ultraviolet (UV) rays applied to aqueous solutions containing Europium and Yttrium in ionic form. UV light affects ions electrically, adding energy to the solution. As a result, a positive charge for the Europium ion is neutralized and if sulfate is added, Europium reacts with it, and the precipitate that is formed can be easily filtered, while Yttrium remains in the solution. The result is a 95% Europium extract and a remaining 98,5% Yttrium solution. The process results in greater efficiency than the liquid-liquid separation, with the additional benefit of preventing dangerous chemical by-products. Because of its larger economic value over all other elements recoverable from lamps, a particular attention deserves Terbium.

An experimental process suggests the possibility of extraction of REEs, as phosphates, especially aimed at maximizing the recovery of Terbium from fluorescent powders. Some dated studies (Merritt, 1990) suggest that RE phosphates, thermally pretreated in an alkaline solution, can be transformed into RE oxides. This process allows obtaining a lower acid concentration, with a hydrometallurgical solubilization treatment and the following segregation by using organic solvents. By identifying the optimal operating conditions (i.e temperature, solid concentration and reaction time of the leaching), the most effective extraction of rare earths can be obtained. In this case, the result was the separation of the initial solution into two mixtures, one with a high percentage of Yttrium (over 98%), with slight traces of other REEs, the different mixture containing several REEs, including Terbium, which has exceeded 70% by weight.

In this case, the possibility to recover REEs from "EoL" fluorescent lamps', internal coating powders has been explored. The research activity has been carried on at full-scale level, involving an Italian company specialized in WEEE recycling. Fluorescent lamps have been collected, treated, and analyzed to examine REEs content and the potential to recycle residual powders that are actually disposed of in the landfill. The work specifically aims to recover Terbium, Europium, and Yttrium oxides.

3. CASE STUDY: THE VALORIZATION OF TERBIUM, EUROPIUM AND YTTRIUM OXIDES FROM END-OF-LIFE FLUORESCENT LAMPS

3.1 Materials and method

3.1.1 Waste collection and characterization

The EEs recycling industry typically consists of waste collection and transport, dismantling, size reduction, sorting, and separation. The metallic concentrates are further subjected to smelting and refining (Hageluken, 2005). Efficient collection schemes have been set up in Europe (Binnemans and Jones, 2014). Exhausted or discarded lamps are considered hazardous waste because they contain MercuryMercury and must be collected separately from other waste in specific bins. Through the WEEE collection and transport chain, the lamps reach DISMECO company. Here, a dedicated system for all constituents, consisting of containing materials, carries out a mechanical separation. According to the type of lamps and the economic life service time, REEs occur in various ratios; therefore, quantity and quality can widely vary from product to product. It follows that identifying the materials, and therefore, the characterization of the waste is a time-consuming work. In the present study, a waste characterization procedure has been set up to identify the waste status and give the appropriate EU waste code. This activity allows identifying the materials in lamps and preparing a preliminary analysis of the potential separation efficiency (%), the recycling performance, and, finally, the percentage of waste destined to incineration or landfill.

To evaluate the separation and recovery efficiency, the following Equation (1) has been used per each material that is potentially valorizable:

- Recovery Efficiency (RE) = $(Wmi_{output} / Wmi_{input})*100$ (1)

  where:
  Wmi_{input} = the weight of material (mi) in input (kg)
  Wi_{output} = the weight of material (mi) in output (kg)

Simultaneously, the REE's complementary value, the Disposal Amount (DA, %) represents the amount of waste (in %) disposed of in an incinerator or landfill.

- Disposal Amount (DA) = 100 – RE (2)

3.1.2 End-of-life lamps treatment

The literature reports three different methods to treat fluorescent lamps:

- direct reuse of phosphorous mixture from lamps;
- recycling of the phosphor components through a physicochemical separation process;
- chemical methods to recover the REE content (Binnemans et al., 2013; Binnemans and Jones, 2014).

In this study, the latter approach has been used. Tube light lamps with external glass or a plastic protective tube must be removed before the mechanical processing. Other lamps (i.e. unusual formats, circular tubes, U-tubes, etc., or broken ones), require a pre-treatment process where additional waste, such as plastics, paper, or other packaging materials, have to be separated. Before mechanical treatment, lamps have to be dried as humidity hinders the functioning of the filters and decreases the system's capacity to separate the fluorescent powders from glass. A mechanical treatment system has been used for the present study, named CCS Unit (Compact Crush and Sieve Unit). It is composed of: a hammer mill, two shredders, three air-material separation towers, a magnetic separator, a glass-metal conveyor, a metal separator, a glass discharge conveyor, a cyclone filter, two filters self-cleaning dust, a pneumatic handling system, four carbon filters, a control panel (see Figure 1).

The fluorescent tubes are fed into the hammer mill (A) to be crushed. In a first separation tower (B), the coarser materials (glass and metals) are separated from the finest particles (dust) inside an air flux. Glass and metals are (C) divided employing a vibrating screen. After a secondary crushing (D) phase, materials are transported by an airflow to the second separation tower and discharged (E-F). Glass andmetals are sorted by the means of a magnetic separator (G). Metals are collected in a container, while glass is further crushed (H) and conveyed to the third separation tower (I). Finally, the glass is sent into the rotating feeder (K) and to the discharging conveyor (L), and collected in "big bags".

Fluorescent powders, carried by the air stream, are separated from a cyclone (M) and two filters (N) system where fine particles are captured and discharged into containers positioned under each filter. The air is treated inside a carbon filter (O) to capture residual Mercury vapors. All materials are transported to the processing plant by using a pneumatic system. A first gravity separation stage segregates the finest and lightest particles (powders) that continue in the flow. Afterward, coarse and heavy elements (glass, ferrous and non ferrous metals, Bakelite, etc.), after a size reduction with a hammer mill, are separated at the second stage of the gravity separation. Later, in other crushing and separation settings, powders are collected in "big bags".

Fluorescent powders containing Mercury vapors, are purified in three stages constituted by:

- a cyclone, where most of the dust is collected in a container;
- a couple of bag filters, where a separation dust-air occurs and fine particles are captured and discharged into containers while air is purified from the dust;
- activated carbon filters retaining Mercury vapors and avoiding their emissions into the atmosphere. Lamps are separated into different fractions represented by glass, ferrous metals, non-ferrous metals, fluorescent powders.

3.1.3 Preparation of the samples

The preparation of the sample consists of a first grinding phase to obtain a fine powder. Then, the powder is homogeneously mixed with a binder (wax, H_3BO_3) and pressed at 100 kN in 13 mm pellets or at 200 kN in 40 mm pellets. Then it is put on a sample holder and inserted into the machinery where a crystal geometrically disperses the emitted radiation (fluorescence). In that way, the different energies, i.e. the spectrum's different lines, can be detected according to the dispersion angle.

In the present study, two samples corresponding to two different batches of collected materials have been investigated. The powders have been extracted from the lamps to carry out a preliminary size analysis. Both samples, weighting 135 g, have been submitted to a particle size analysis performed by a common mechanical siever. It was chosen a column of six sieves, (according to technical Iso standards) having the following meshes dimensions: 500, 300, 212, 150, 106, 75 microns.

3.1.4 Fluorescent powder analysis

Existing methods for the recovery of the detected materials have been explored to evaluate the concentration of the REEs and the other main constituents. In this study, a Wavelength Dispersion X-Ray Fluorescence (WD XRF) instrumentation has been chosen. The. X-Ray Fluorescence (XRF) spectrophotometry is a non-destructive technique, which allows the identification of chemical elements in a sample on the basis of the X-ray fluorescence radiation emitted by the atoms following a high-energy X and gamma rays irradiation. In particular, in the Wavelength Dispersion XRF (WD XRF) technique, the emitted radiation is geometrically dispersed by a crystal. The different energies, i.e. the different spectrum lines, can be detected as a function of the dispersion angle. The WD XRF detects the different wavelengths of the radiation emitted by the material (excit-

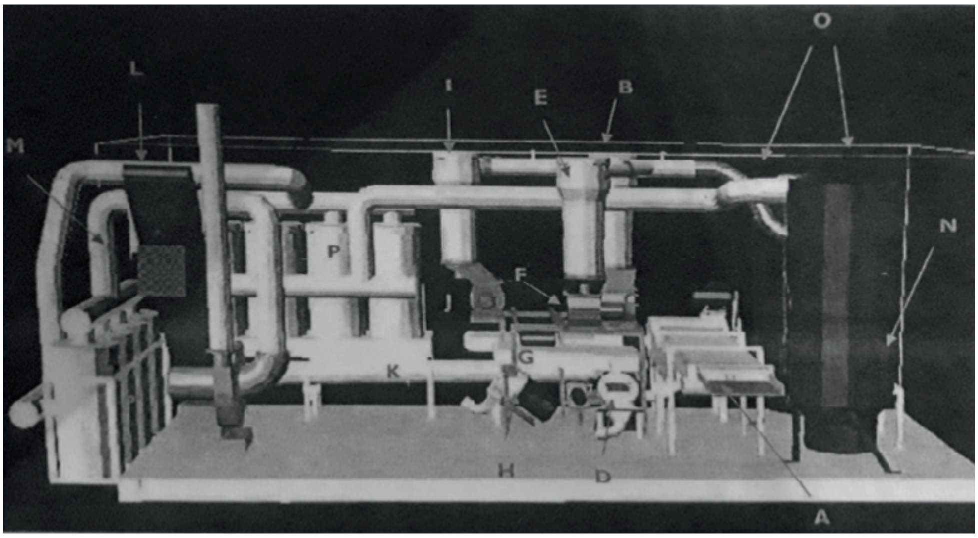

FIGURE 2: The CCS scheme.

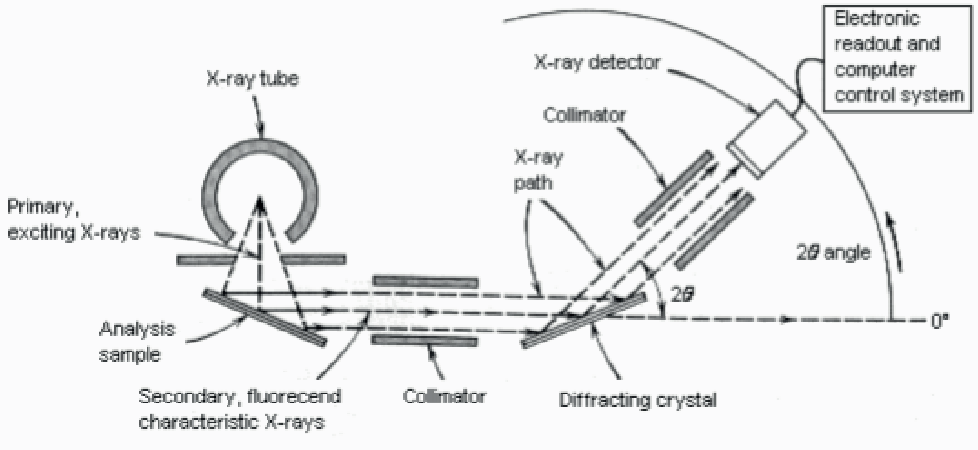

FIGURE 3: WD XRF instrumentation picture.

ed by the X-Rays) through the dispersion spectrum that it generates through a prismatic crystal. In Figure 2, the WD XRF instrumentation is shown.

The analysis was performed on two identical samples, weighting 7,60 g. each, and considering three size intervals: d<75 μm; d = 75÷150 μm; d = 150÷500 μm.

4. RESULTS

4.1 Preliminary feasibility analysis

Tests have been carried on defining the waste status and its classification according to the European waste code (see Table 1). Once the wastes have been codified, each material is weighted to measure the raw materials recovery efficiency. As shown in the table, the company carries out an almost total recovery for ferrous and non-ferrous metals, paper packaging and other lamps; a partial recovery occurs in cases of glass, plastics, and impure ferrous metals that will be sent to the energy recovery system. Fluorescent powders, on the other hand, are valorized through a physical-chemical treatment.

As described above, the following step is represented by the test analysis to identify and characterize the materials. Indeed, when an urban mining project is approached, the primary goal is to explore the potential extraction performance (Allwood et al., 2011). From an ordinary lamp, weighting about 200 g, it is possible to exploit about 137 g of glass, 20 g of plastics, 13 g of metals, and 5 g of powders.

The results of the size analysis are reported in Table 2, while Table 3 reports compound concentration at three different size ranges: d<75 μm, d = 75÷150 μm and d = 150÷500 μm for both samples. The particle size analysis was only functional to the subsequent detection of the REEs concentrations as a function of the samples' size. The data reveal that the types and amounts of elements found deviate slightly for the particle sizes d<75 μm and d = 75 ÷150 μm. The sample with size d=150÷500 μm presents a high glass content, which increases proportionally with the elements' size.

In the second sampling, a greater atmospheric humid-

TABLE 1: Test Recovery Efficiency.

EU Waste Code	**Description**	**Waste** (kg)	**Mass Balance** (%)	**Materials Recovery Efficiency (RE)** (%)	**Incineration Landfilling** (%)
191212	Powder	10.537	5,30	0	100
191202	Ferrous Metals	2.386	1,20	98	2
191203	Non Ferrous Metals	3.579	1,80	100	0
191205	Glass	136.078	68,45	20	80
191204	Plastics	19.741	9,93	40	60
160216	Ferrous and Non Ferrous metals	20.476	10,30	100	0
150101	Paper packaging	4.970	2,50	100	0
200121*/160214	Other lamps	1.034	0,52	100	0
	Total	198801	100		

TABLE 2: Size analysis results for the two tested samples.

Sieve Size (μm)	**Sample 1 Cumulative Passing (%)**	**Sample 2 Cumulative Passing (%)**
500	89,48	86,76
300	72,75	71,67
212	48,67	49,36
150	24,50	24,36
106	16,83	18,92
75	8,51	9,57
0	0	0

ity resulted in an over-discharging of dust due to adhesion to the coarse glass from the towers This was proved by the analyzed fine dust in terms of high SiO_2 percentage and a reduced presence of Rare Earth. It should be emphasized the result in terms of abatement of MercuryMercury which is always less than 0,02%, demonstrating the high efficiency of the treatment.

Regarding the presence of REEs, Yttrium Oxide (Y_2O_3), Lanthanum Oxide (La_2O_3), Cerium Oxide (CeO_2), Europium Oxide (Eu_2O_3), Therbium Oxide (Tb_4O_7) and Gadolinium Oxide (Gd_2O_3) have been found in both samples. The differences in % can be explained because of the high glass content at the second sample mainly located at the more coarse particle size class (500-150 μm).

The results show that the significant REEs percentage is contained in the finest powders. This trend is evident for all the oxides and both samples. The results relating to the most acceptable fraction (<75μm) remain substantially constant, providing helpful information for determining the dimensions in which to look for the REEs. However, significant amounts, although less, of REEs are also present at the other sizes. A first comparison between the two samples shows that the elements found and their quantities do not differ much, especially in sieving <75 μm. At the d = 150÷500 μm, the second sample shows a higher glass content, probably due to humidity, making it more challenging to separate the glass from the dust. This analysis allows forecasting the yearly volume of lamps to treats and the amount of materials that can be potentially exploitable. By assuming about 21.000 kg/year of fluorescent lamp powders that can be treated at the plant, Table 4 shows the total amount of REEs that can be recovered yearly.

4.2 Economic feasibility analysis

Once the materials have been registered in a database and weighted, it is necessary to estimate the economic benefits and the maximum value extracted from lamps to assess the actual economic feasibility (Krook, 2010; Cossu and Williams, 2015). Based on the results carried out by the preliminary analysis, a market and cost-benefit evaluation have been performed. At first, the current price of REEs in the international market has been overviewed. Updated in December 2019, all data are reported as RMB (the Chinese currency Renminbi) per kilos, as summarized in Table 5. As Deboer and Lammertsma (2013) noted, the prices of REEs have changed dramatically in the last years, reaching fluctuations of up to 10. After a peak registered in 2011-2012, the prices quickly decreased in the following years.

Considering the plant recovery rate and the volume of materials potentially treated every year (see Table 6), the revenue from this kind of urban mining exploitation can be estimated.

At the same time, costs should be addressed. To minimize initial investments, representing the highest economic output, the possibility of activating joint public/private collaboration mechanisms has to be explored.

5. DISCUSSION

To date, the recovery of REEs is still almost entirely unexplored at large-scale level since the techniques

TABLE 3: Compound concentration for the two tested samples (REEs in bold).

Compound Name	Sample 1			Sample 2		
	500-150 µm Concentration [%]	**150-75 µm** Concentration [%]	**<75 µm** Concentration [%]	**500-150 µm** Concentration [%]	**150-75 µm** Concentration [%]	**<75 µm** Concentration [%]
SiO_2	32,00	29,13	22,18	68,27	30,79	17,22
Al_2O_3	15,33	17,39	18,40	2,49	12,14	13,37
CaO	14,87	14,75	16,79	6,56	18,57	22,61
P_2O_5	11,35	12,52	14,16	0,18	14,61	16,38
Y_2O_3	**8,60**	**9,60**	**10,95**	**0,08**	**6,44**	**9,04**
Na_2O	6,37	5,82	4,72	16,14	7,05	4,74
SrO	1,77	2,05	2,90	0,08	0,43	0,67
BaO	1,54	1,37	1,24	0,57	1,55	2,13
CdO	1,32	1,29	1,21	0,68	1,01	1,61
La_2O_3	**1,03**	**1,15**	**1,45**	**-**	**0,50**	**1,28**
CeO_2	**1,02**	**1,17**	**1,44**	**0,28**	**0,64**	**1,42**
K_2O	0,89	0,77	0,56	1,32	0,75	0.54
Eu_2O_3	**0,71**	**0,83**	**0,99**	**-**	**0,45**	**0,73**
Tb_4O_7	**0,56**	**0,65**	**0,82**	**-**	**0,37**	**0,62**
Fe_2O_3	0,42	0,47	0,58	0,17	0,40	0,52
Gd_2O_3	**0,39**	**0,42**	**0,57**	**-**	**0,31**	**0,56**
PbO	0,38	0,33	0,21	0,31	0,58	0,24
GeO_2	0,33	0,39	0,55	0,32	0,30	0,05
SO_3	0,19	0,20	0,18	0,17	0,16	0,12
MnO	0,19	0,20	0,27	0,01	0,27	0,47
Cl	0,17	0,19	0,22	0,04	0,21	0,24
NiO	0,16	0,18	0,24	0,15	0,08	0,15
Sb_2O_3	0,14	0,12	0,16	0,04	0,21	0,40
TiO_2	0,07	0,07	0,07	0,06	0,08	0,06
WO_3	0,07	0,07	0,12	0,08	0,06	0,09
ZnO	0,05	0,05	0,08	<0.01	0,05	0,09
CuO	0,04	0,05	0,08	0,01	0,03	0,06
SnO_2	0,04	0,03	0,07	0,03	0,03	0,03
Cr_2O_3	0,02	0,02	0,01	<0.01	0,02	0,01
HgO	0,02	0,01	<0.01	<0.01	<0.01	0,02
Bi_2O_3	0,01	0,01	0,01	<0.01	<0.01	<0.01

TABLE 4: Potential yearly amount of REE (kg).

	Sample 1				Sample 2			
	d=500-150 µm	**d=150-75 µm**	**d<75 µm**	**Total**	**d=500-150 µm**	**d=150-75 µm**	**d<75 µm**	**Total**
Y_2O_3	1806,0	2016,0	2299,5	6121,5	16,8	1352,4	1898,4	3267,6
La_2O_3	216,3	241,5	304,5	762,3	-	105,0	268,8	373,8
CeO_2	214,2	245,7	302,4	762,3	58,8	134,4	298,2	491,4
Eu_2O_3	149,1	174,3	207,9	531,3	-	94,5	153,3	247,8
Tb_4O_7	117,6	136,5	174,3	428,4	-	77,7	130,2	207,9
Gd_2O_3	81,9	88,2	119,7	289,8	-	65,1	117,6	182,7

mentioned have remained at the experimental stage. As demonstrated by the preliminary feasibility analysis, the chemical treatment here described cancan extract considerable amounts of REEs from powders disposed of in the landfill. The quantities of Rare Earths recoverable from fluorescent lamps, as the analysis results have proved, are interesting to support an effective recycling process.

At the same time, limits and critical issues already exist. A summary f challenges are here listed:

- lack of product-centric approach;
- high costs for extraction processes;

TABLE 5: Price of Rare Earth Oxides (Updated in December 2019).

	Price [RMB/kg]	**Price** [€/kg]
Lanthanum Oxide	12,25	1,6
Cerium Oxide	12,25	1,6
Europium Oxide	215	28,0
Gadolinium Oxide	160	21,0
Terbium Oxide	3470	450,0
Yttrium Oxide	20	2,6

TABLE 6: Potential revenue from the REEs valorization).

	Total amount [kg]	**Revenue** [€/kg]	**Yearly revenue** [€/year]
Lanthanum Oxide	762,3	1,6	1219,68
Cerium Oxide	762,3	1,6	1219,68
Europium Oxide	531,3	28	14876,4
Gadolinium Oxide	289,8	21	6085,8
Terbium Oxide	428,4	450	192780
Yttrium Oxide	6121,5	2,6	15915,9

- high price volatility;
- generation of toxic by-products from the chemical techniques adopted to extract materials;
- relative costs to dispose of the toxic by-products;
- critical cost-benefits outputs because of the higher treatment costs compared to the market price of REEs
- fragmentation and lack of harmonization in the collection and treatment of fluorescent lamps at EoL;
- lack of incentives promoting research on separation of Mercury from fluorescent powders, making the latter more environmentally compatible and treatable at lower costs.

6. CONCLUSIONS

Given their properties and field of application, REEs have become fundamentally crucial for all industries manufacturing electronics and high-tech technologies. The growing interest in low-energy and intelligent technologies will massively affect REEs' market in the next ten years. However, as highlighted by the EU Commission, the EE industry is at risk due to the limited availability of primary resources. This critical issue is of the highest urgency for EU countries that import REEs from China as the primary producer, exporter, and consumer worldwide. REEs extraction, as well as transportation, have a significant impact from an environmental point of view. Additional effects have been registered at the economic and social level since China, taking advantage of the lack of environmental controls and regulations in exploitation and mining, leads the market as a monopoly.

To counteract this situation and ensure the future REEs supply, EU countries, driven by policies and regulations, have started to develop techniques for the recovery of REEs.

According to the type of electronic devices, materials and relative amounts can widely vary. Simultaneously, the limited presence of these materials is complicating the extraction process at such a point to be inconvenient from the economic point of view. In the case of lamps, the REEs recovery is not well-developed to date. The essential technologies would require long timeframes to exploit a functional industrial scale and provide an adequate supply source.

In the short term, the search for alternative substances for the development of technological appliances has not led to the discovery of valid substitutes: only REEs can guarantee, where used, high EE efficiency with drastic and indispensable dimensional reduction of components and appliances.

In the long run, since REEs are still used in the new LED generation, recycling should be robustly explored to translate material value into economic revenue and so foster the circularity of these materials. At the same time, additional efforts should be made in hazardous materials management: Mercury and radioactive elements (such as Uranium and Thorium) represent a pervasive presence in EE applications. By subjecting as much of WEEE as possible to REEs recovery and recycling processes, hazardous substance dispersion should decrease, at least within a controlled industrial environment.

The increasing awareness of resource scarcity and climate pressures have led industrial stakeholders to adopt pro-active behaviors aimed at reducing impacts and externalities in the environment. The EE industry has started a demanding route to prevent the supply of CRMs by implementing circular strategies: from eco-design to service-based system and finally, material recovery. All these actions promote on the one hand, the life service extension and in the other the value maintenance of the materials. The complexity in isolating and separating REEs from EEEs at EoL is challenging because of the minimum content. In Italy, about 200 companies are authorized to treat and valorize materials included in the WEEE stream. Among them, DISMECO srl, works on the processing of light-emitting devices. The substitution of fluorescent lamps with LED lamps has caused a rapid increase in the disposal of this lighting technology. The high volume of these products has boosted the company to think about a material valorization process to recover REEs. The sampling activitysampling activity results have encouraged the creation of collaboration forms with lamp designers and manufacturers in ordersampling activity results, enabling the creation of potential collaboration forms with lamp designers and manufacturers to adopt a product-centric approach able to optimize EoL performance. The establishment of Rare Earths' recovery centers would significantly reduce the technical and technological issues. This could also be translated into a positive economic value: either in terms of potential sales income of the substances obtained or as a stimulus to experimenting with advanced technologies. Indeed, even if fluorescent lamps have been substituted by LED lamps, their presence will remain in the WEEE stream for the next twenty years. Their proper treatment will feed the supply chain for the new lights that, in a systemic approach, should integrate additional strategies such as eco-design, assembly-disassembly, and finally, remanufacturing.

Despite the quantitative availability of fluorescent powders, the results of the analyses carried out in the present study do not highlight any truly significant contents of rare metals, such as to motivate a targeted recovery investment. The empirical analysis reveals high cost for material extraction and lack of substantial revenue from the REEs purchase. In the absence or pending further research progress based on the sustainable extraction process (at lower costs, with less use of chemical reagents and less generation of toxic by-products), the only possible alternative would be the use of economy of scale by developing integrated recycling routes. To manage higher volume, more harmonization in the collection scheme is necessary. After that, it is essential to boost cooperation and collaborations among the experts of the sectors and increase public entities' participation. Now that the COVID-19 pandemic has forced the European Community and nations to review the state aid criteria completely thoroughly, a new historical phase could open. Consequently, it would be likely possible to re-evaluate the support for companies working in sectors whose continuity is indispensable for the community, such as those protecting the environment and generators of work and income.

Therefore, this study, starting from a particular theme, explored technical and operational problems directly influencing possible industrial opportunities. Moreover, it had the ambition to highlight issues in a broader context, from local to the national level, up to the global one, intending to spread activities that increasingly support the circular economy and urban mining perspective.

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DEMONSTRATING EEE RECOVERY FOR REUSE IN A DISTINCT URBAN MINE: A CASE STUDY

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ABSTRACT

At the heart of the circular economy model is the reorientation of consumer behaviours away from disposing of items before they reach end of their functional life as a step towards resource efficiency and reduction of environmental impacts. One way to facilitate this change is to enable proactive redistribution of electrical and electronic equipment (EEE) with reuse value from urban spaces followed by high quality recycling at end-of-life. We have conducted the first assessment and critical evaluation of a model for the recovery of reusable EEE from a distinct urban mine (DUM) - in this case, a university. The Infrastructure, Service and Behaviour (ISB) model was used as a guide for interventions. Small EEE recovered from two Halls of Residence at the University of Southampton were characterised, visually inspected and sorted. From the items inspected visually, 97% was reusable and were donated to participating charities for redistribution via reuse/sale. The results show that an ISB Model system designed using choice architecture to recover reusable EEE from a DUM contributes strongly to extending products' lifetimes and promotes circular economy ambitions. The study provides strong evidence of a viable reuse-based recovery system for small EEE in a university DUM and with a potential for replicability at global scale. It is recommended that a carefully planned and tailored system based on the ISB model should be put in place in universities for the recovery and redistribution of reusable EEE (ReEEE) and that recycling is implemented only after practical options for reuse have been exhausted.

1. INTRODUCTION

Waste Electrical and Electronic Equipment (WEEE) is one of the fastest growing waste streams globally; over 53 million tonnes are generated annually (Forti et al., 2020; Shittu et al., 2021). In 2019, the world generated 53.6 million metric tonnes of WEEE, and only 17.4% of this was officially documented as properly collected and recycled (Forti et al., 2020). Further growth is expected with the rapid expansion of the digital economy, especially during the COVID-19 pandemic and its anticipated consequences, and the emergence of new technologies in areas such as artificial intelligence, biomedical engineering, renewable energy, space travel, e-textiles and smart agriculture (Shittu et al., 2021). A large proportion of this waste currently comprises personal and consumer electronics such as laptop computers, mobile 'phones and TVs. Around 20% of WEEE generated globally is recycled but the fate of much WEEE is undocumented (Balde et al., 2017; World Economic Forum, 2019; Forti et al., 2020); tracking the flows of end-of-life electrical and electronic equipment (EEE) is thus challenging. Europe currently has the highest reported WEEE recycling rate at 35%, due to the implementation of the WEEE Directive in European Union (EU) Member States (WEEE Forum, 2019; Forti et al., 2020; Shittu et al., 2021).

The increasing quantities generated have brought WEEE to global attention (Zhang et al., 2019). Studies of WEEE range from the ill-effects on health and environment of poorly-managed WEEE to global recycling practices. There is some, if limited, focus and emphasis on reusability of discarded EEE, aligned with the promotion of a circular economy. Distinct Urban Mining (Ongondo et al., 2015; Pierron et al., 2017; Parajuly et al., 2017; Wilkinson & Williams, 2019) offers much in this regard. As an example

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of a distinct urban mine, a university campus is analogous to a small city with the potential for a high concentration of discarded EEE (Ongondo et al., 2015), i.e. a distinct space that could be a hub for the recovery of valuable materials and products that would otherwise be thrown away (Cui & Zhang, 2008; Powell & Williams, 2019).

Consumerism is escalating as manufacturers create and promote new products, particularly as an outcome of the quest by producers for higher market share. At the same time, growing global affluence has resulted in an increase in "throw-away culture." Consumption and manufacture of short-lived or disposable items rather than durable products that can be repaired inevitably leads to high turnover of items by consumers as consumers opt for newer, often trend-driven items. The linear pattern of buy-use-discard results in the relentless generation of waste that puts immense strain on resources, as there is a constant need to source raw materials to manufacture products to meet increasing demands. Contemporary high-tech EEE such as home entertainment equipment is especially rich in metals and critical raw materials (e.g. rare earth metals and platinum group metals) and constitutes a marked portion of anthropogenic stocks (Massari & Ruberti, 2013; Golev et al., 2016; Williams, 2016). Demand for consumer electronics is increasing and device usage cycles are shortening, which results in the generation of large quantities of discarded items. This situation has led to an increase in resource efficiency-oriented plans and strategies in the UK and Europe. These range from strategies targeting sustainable consumption such as 'Resource Revolution' (WRAP, 2015) to those promoting reduction in waste generation via product reuse and reparability, such as the EU Circular Economy Action Plan (Circular, 2020). However, some manufacturers appear to dislike these initiatives; Apple Inc., for example, took legal action against a small independent repair shop to the Supreme Court in Norway in order to prevent refurbishment of Apple's iPhones, claiming that its trademark had been "unlawfully appropriated". Apple Inc. won the Norwegian Supreme Court case in June 2020, which could be considered an impediment to reuse. Indeed, this legal decision reinforces maintenance of the status quo (i.e. more recycling and better management of waste disposal) at the expense of societal objectives to reduce, through reuse, the amount of waste being generated and to minimise its potential for harm to human health and the environment. In this respect, this case also raises the importance of recycling specific items (such as small WEEE) and thus highlights the importance of data on product (waste) composition to facilitate end markets for recyclates.

2. END-OF-USE DECISIONS

Purchasing decisions are influenced by several factors including, but not limited to, purchasing power (money), lifestyle, peer pressure, contemporary fashion trends, advertising, etc. These decisions often influence the duration of product use (Cox et al., 2013). When a product is no longer wanted or needed (e.g. broken or deemed obsolete, old or out of fashion), the owner could make one of a number of decisions for the fate of the item; decision in this context refers to what is done to the product after it is deemed by the owner to have reached its end-of-life. The decision made could either continue a linear path of production (make-use-dispose) or that of a closed loop. Figure 1 illustrates potential end-of-use decisions and the relationship between each decision.

End-of-use decisions remain strongly influenced by targets relating to recycling. Whilst recycling provides a "feel-good" factor to the public, it does not result in decoupling of consumption and waste generation or the correct application of principles of the waste hierarchy and circular econo-

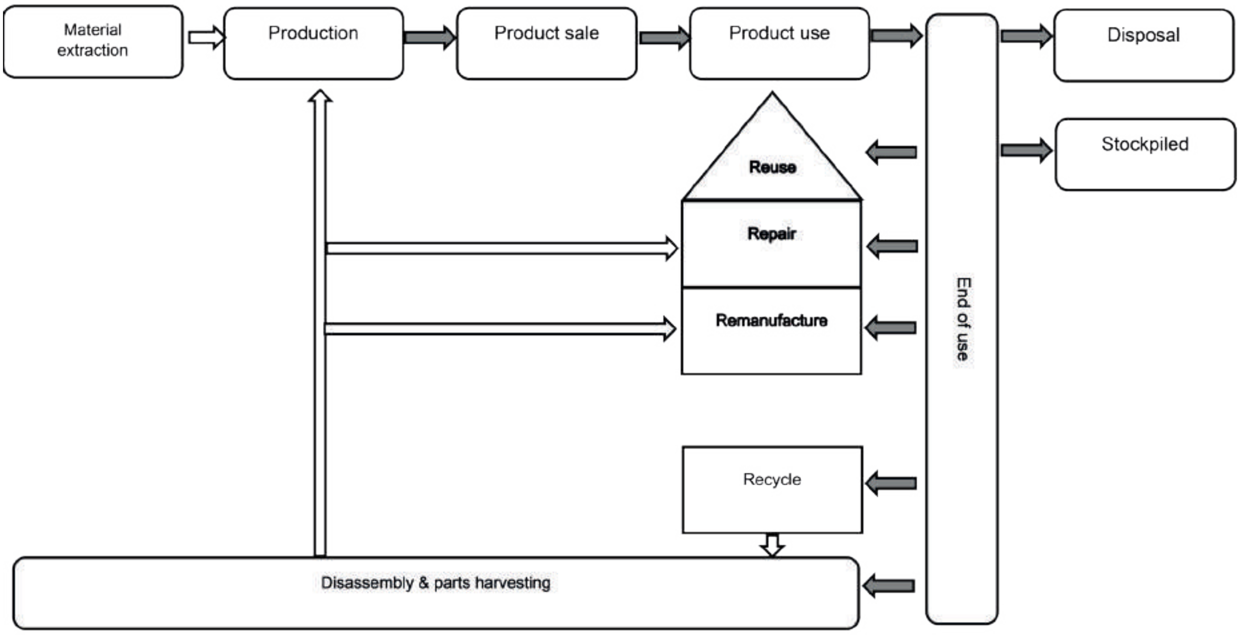


FIGURE 1: Generic product lifecycle with potential destinations after use. Solid arrows indicate product movement; unshaded arrows indicate materials movement.

my. The decision to dispose, stockpile (a back-up supply retained for future use if/when needed), hoard (stored items that exceed consumers' requirements for use or back-up), reuse or recycle is dependent on various factors including, but not limited to, perceived intrinsic product values, availability and convenience of reuse/recycling channels, ease of repair etc. Behaviour is an important influence in the decision-making at the end-of-use of a product. Decisions promoting a circular economy approach require product owners to behave in a certain manner – such as de-stockpiling/de-hoarding or buying pre-owned products – to facilitate actions consistent with the desired application of the waste hierarchy (Dunlap & Jones, 2002; Ongondo et al., 2015, Pierron et al., 2017). Despite the general unpredictability of human behaviour, several theories have been developed to provide insights into human behaviours and the factors influencing them (Darnton, 2008). The Theory of Planned Behaviour (TPB) and the Value-Belief-Norm (VBN) model, for example, have been applied to explain pro-environmental behaviour (Kaiser et al., 2005). Whilst pro-environmental decisions are likely to be influenced by intrinsic motivators such as beliefs, attitudes and norms (Kaiser et al., 2005), pro-environmental behaviours can also be influenced by choice architecture (Thaler et al., 2010), which involves the modification of situational factors to bring about a desired outcome. In their study of distinct urban mining potential of a UK university, for example, Pierron et al. (2017) investigated ownership, stockpiling and disposal of small EEE amongst students and concluded that choice architecture can be deployed to initiate specific and desired outcomes at a product's end of use.

Behavioural theories have been applied when seeking to develop interventions in environmental management. Such interventions are designed such that they address intrinsic and extrinsic barriers to change (Schultz et al., 1995; Timlett & Williams, 2011). Intrinsic motivation for individuals to engage in pro-environmental behaviours such as reuse and recycling can give rise to a naturally satisfying "warm-glow" effect. Such motivators include personal satisfaction, positive emotions and altruistic motives that benefit the well-being of others. Extrinsic motivators to engage in reuse and recycling may include a desire to conform to social and societal norms, enhancement of personal reputation, praise and financial rewards; several socio-psychological behavioural models have been proposed to explain waste-related behaviours (Williams, 2015). Behavioural change is also influenced by situational factors such as income and infrastructure, although the strength of these factors upon major decisions and actions may be limited (Stern, 2000). Consequently, behavioural change interventions require measures addressing both internal (psychological; intrinsic and extrinsic) and external (situational) factors. Models incorporating both intrinsic and extrinsic factors to explain behaviour in the context of waste and resource management have merit. The Infrastructure, Service, Behaviour (ISB) model, for example, adopts this approach for planning interventions to maximise resource efficiency via consideration of situational and psychological variables (Timlett and Williams, 2011), and has been previously applied to demonstrate the impact of introducing specific and guided interventions in waste and resource management.

2.1 Higher Education Institutions as circular economy hubs

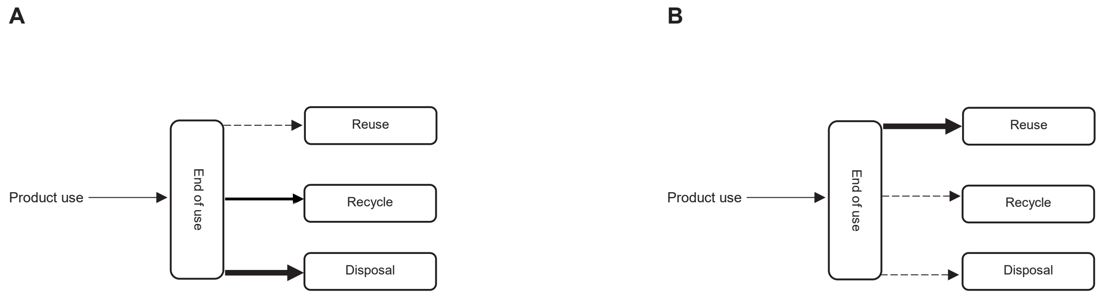
Higher Education Institutions (HEIs) are viewed as beacons of positive change and promoters of environmental sustainability (Martin and Samels, 2012; Vagnoni and Cavicchi, 2015). This reputation is achieved via knowledge creation and dissemination as well as commitment to sustainable initiatives and policies (Zhang et al., 2011; Tangwanichagapong et al., 2017), ranging from construction of 'green' buildings to carbon-neutral transportation systems and sustainable waste management systems with emphasis on reuse, recycling and resource conservation. With regard to waste management, one step towards achieving sustainability is to consider a HEI to constitute a distinct urban mine (Ongondo et al., 2015). HEIs can be viewed as small cities and provide a microcosm of the settlements within which they are situated. People within these HEI environments, like regular towns or cities, are consumers of goods and services, which make these urban spaces ideal for studying and trialling new initiatives before being implemented at broader scale.

In recent years, resource recovery from waste has been in focus in HEIs. There is growing emphasis on diversion of materials from landfill and circular economy thinking based upon application of the waste hierarchy. In a university urban mine, there is potential to divert potentially reusable items during periods when students vacate Halls of Residence accommodation and thereby contribute to efforts to use resources more effectively. One of the categories of items recoverable during such periods is (W)EEE. With WEEE collection rates currently low in most countries and stockpiling of WEEE common, valuable and critical raw materials within these items are potentially lost. WEEE generated globally in 2019 was estimated to contain £50 billion worth of secondary raw materials (Forti et al., 2020). The loss of such critical raw materials as well as good, functional reusable products emphasizes the need to adopt circular economy approaches, as these will have a positive impact on the future management of (W)EEE. To achieve this aim, an understanding of distinct urban mines is required which will help enhance recovery of reusable EEE as well as resource recovery from WEEE via recycling, leading overall to improved WEEE management. However, for an urban mine to be considered viable, there must be detailed data and information concerning its attributes such as location, size, concentration of materials and resources to be prospected, and products flows.

HEIs in the UK typically provide accommodation for first year students as well as international students enrolled on foundation, pre-sessional and postgraduate courses. According to HESA (2020), approximately 15% of all enrolled students in UK HEIs in were in university accommodation during the 2018/19 academic year (>300,000 students). An academic year is typically made up of 3 terms each of around 10 weeks; the beginning of each term is accompanied by a turnover of students synchronised with teaching schedules. The first term generally comes with the highest level of enrolment in late September, while the summer term sees most student departures in June or July. These turnover periods usually result in a high number of departures from student accommodation. The university-maintained facilities thus encounter two (or sometimes three) annual "move-outs" during which students vacate their accommodation (Williams and Powell, 2019); undergraduates move out in early summer and postgraduates (and sometimes pre-sessional language) students move out during early autumn. These periods are usually associated with some products reaching their end of use. The end-of-use decision made by the owner will be largely dependent on the availability of appropriate (situational) factors. While a departing student is likely to take with them some of their personal possessions, there is always a high likelihood that some products will be discarded, some of which will retain functionality and be reusable. This pattern typically results in the generation of large quantities of discarded items, ranging from bedding, textiles and bric-a-brac to electronics and kitchenware. These regular and predictable surges in the generation of discarded items often lead to littering of streets around student dwellings and tension with the local community. There have been a few attempts by HEIs to alleviate this issue. In their review of HEIs' reuse schemes, Williams and Powell (2019) highlighted a number of schemes in UK HEIs conceived to deal better with move-outs: recovering reusable items results in diversion from landfill, aid to charitable causes and improves relationships with residents and authorities. In several of the schemes reviewed, items deemed in good condition and reusable were donated to charity organisations, sale of which generates income to support the actions of the charities.

The potential recycling value of WEEE is well established in terms of, for example, the potential value of material recycling (Chancerel and Rotter, 2009) and as a secondary metal resource (Oguchi et al., 2011). The concept and potential of urban mining for WEEE within distinct urban spaces (i.e. universities) has been established for WEEE (Ongondo et al., 2015); the collection of both WEEE and end-of-use EEE could be enhanced by the application of choice architecture (Pierron et al., 2017). Relatively few studies, however, have explored the potential for recovery of reusable EEE within distinct urban spaces. Wilkinson and Williams (2020) evaluated the ownership and hoarding levels of home entertainment EEE within a DUM and found a high level of hoarding of devices that can be potentially recovered; the potential for the recovery of reusable items from students in a HEI has been demonstrated but not yet for EEE (Williams and Powell, 2019).

The potential for enhancing reuse in this context can be illustrated by comparing two scenarios, with and without opportunities for EEE reuse. Under a scenario in which HEIs provide limited opportunity for the recovery of reusable EEE discarded by students upon departure from university accommodation (Figure 2A), such items are either destined for recycling or commingled with residual waste. Consequently, discarded items with reuse value would be landfilled or recycled: while recycling is a preferred to landfill as an outcome, recycling does not exploit the full value of a still-functional product. A scenario in which opportunities for the recovery of reusable EEE are provided (Figure 2B) requires a targeted intervention and desired behavioural response. Using the ISB model (Timlett and Williams, 2011), suitable interventions can be implemented. A perfect scenario (Figure 2B) for a reuse-based system is one where infrastructure (I), service (S) and behaviour (B) all contribute towards achieving 100% reuse.



A

I: little or no infrastructure to facilitate product reuse; products likely to be comingled with residual waste or hibernated

S: no ancillary services for recovery of reusable items

B: willing participants however little service and/or infrastructure likely to inhibit participation in product reuse

OUTCOME: Recovery for reuse rate = 0%

B

I: provision of infrastructure to facilitate product reuse e.g. personal collection bags, temporary storage area

S: availability of ancillary services for recovery of reusable items

B: awareness campaign in form of adverts, notices, themed events etc. to facilitate behaviour change

OUTCOME: Recovery for reuse rate = 100%

FIGURE 2: Schematic illustration of extreme end-of-use scenarios. Infrastructure (Aspects of the built environment such as buildings, storage bins for recycling); Services (Protocols or systems that enable patterns of behaviour e.g. weekly collection of dry recyclables); Behaviour (How a person undertakes recycling/reuse) (Timlett and Williams, 2011). Panel A illustrates the baseline scenario before intervention. The likely destination for reusable EEE in this scenario is residual waste which goes to landfill with some recycling occurring; B shows the shift to recovery of products for reuse after introduction of an ISB-based recovery protocol aimed at achieving 100% recovery for reuse. Thickness of arrows indicates likelihood of product destination.

In this study, we demonstrate the implementation of a reuse-based EEE recovery system conceptualised using the ISB model for the recovery of functional reusable EEE in a university DUM. This is the first assessment and critical evaluation of the potential for recovery and redistribution of reusable EEE from a specific stream in a university urban mine using a reuse-based recovery system. The study makes the case for product reuse as the priority, targeted, most beneficial end-of-use option for sound products as opposed to an inadvertent and unconscious drift towards options that result in giving precedence to product recycling within a DUM. This study presents therefore an evaluation of the potential of a DUM for the recovery of reusable EEE as a priority, with focus on recycling only after practical options for reuse have been fully applied and exhausted. We present reuse as the preferable outcome for end-of-use EEE, thereby keeping products in the system for as long as possible.

The outcomes of the study are as follows:

- Design and implementation of a reuse-based recovery protocol using the ISB model (Timlett and Williams, 2011);
- Estimation and potential value/revenue projection from products recovered for reuse (product and material value);
- Appraisal of collection and recovery protocols; and
- Recommendations on improvements to collection and preparation for reuse of EEE and recycling of WEEE from a DUM.

3. METHODS

The study was centred on an assessment of reuse potential in a university urban mine, with focus was on students' Halls of Residence (HoR). HoR experience turnovers of student residents at specific periods, which provide a unique opportunity to investigate the potential for recovery of reusable EEE when students move out of their accommodation. Also, due to their mixed occupancy, HoR present an ideal study area to assess recovery of items from different groups of students (e.g. undergraduates/postgraduates; home/international domiciled).

The study was conducted in three phases: pre-collection, collection and sorting, and post-collection (Table 1). The pre-collection activities took place in the months leading to the end of summer term of the 2018/19 academic year while the collection phase took place during June, July and September 2019.

3.1 Study area

The study took place at the University of Southampton, a large multi-campus university located in the city of Southampton, UK. It has a student population of 24,625 (HESA, 2020), most of whom are based at the main campus in Highfield. The university currently has eight institution-owned HoR offering an array of room types and sizes (University of Southampton, 2019). For this project, two HoR were selected with contrasting populations of student residents (Table 2). As the study set out to encompass a varied and comprehensive mix of students for representativeness, these halls provided representative samples with regard to predominant student groups they accommodate and size and layout.

Mayflower Halls is a large student complex located near Southampton city centre, some 3 km from the main campus. It has a room capacity of 1,105. The complex provides accommodation to students at all levels of study (foundation, undergraduate and postgraduate). By contrast, City Gateway is relatively small, located 1.6 km from the main campus and is exclusively for postgraduate and mature students (21 years or older at the start of their studies). These two HoR vary from layout to types of students accommodated which provides ideal contrast and justifies their selection for the study.

TABLE 1: Project methodology outline including phases and activities.

Study Phase	Activities
Pre-collection	Protocol development Meetings with representatives of residential services and selected charities Recruitment and protocol briefing of project volunteers
Collection and Sorting	Items drop and transportation to central sorting location Product inspection and data collection Product redistribution
Post-collection	Analysis of reuse potential Material composition analysis (using secondary data) Process analysis

TABLE 2: Details of selected Halls of Residence (University of Southampton Residential Services, 2019).

Hall of Residence	Maximum capacity	Room types	Dominant student type
City Gateway	364	Single rooms (334) Studio apartments (14) 1-bed flats (12) 2-bed flats (4)	Postgraduate students
Mayflower	1105	Single rooms (1031) Studio apartments (54) 1-bed flats (20)	Undergraduate; pre-sessional students

3.2 Protocol development

The study was based on the assessment of outtakes from two HoR (Mayflower and City Gateway Halls; Table 2) in the University of Southampton using the ISB model (Timlett and Williams, 2011). Increased rate of reuse/recycling, or indeed any pro-environmental behaviour, requires a balance of situational (infrastructural, service) and psychological (behavioural) factors (see Figure 2). In this context, infrastructure is defined as aspects of the built environment such as buildings, storage bins for recycling; service refers to protocols or systems that enable a pattern of behaviour e.g. weekly collection of dry recyclates; and behaviour refers to how a person undertakes recycling/reuse, influenced by the intrinsic and extrinsic factors discussed above. The methods adopted for this project are guided by the WEEE characterisation study by Parajuly and Wenzel (2017) to evaluate the reuse value and recycling potential of collected household WEEE in Denmark.

This study formed a part of wider initiatives at the University of Southampton to improve institutional resource management (Zhang et al., 2011; Ongondo and Williams, 2011; Pierron et al., 2017; Robinson et al., 2015, 2018), including the collection of reusable items (clothes, homeware, and furniture) for donation to selected charities (Powell & Williams, 2019). The University of Southampton has committed to an evidence-based cleaner, greener and healthier future, bringing students, staff and local residents together to improve sustainability across our estate (University of Southampton, 2020). In the days leading up to the move-out periods, each collection room at the selected HoR was provided with red plastic bags to enable the deposition of functional but unwanted EEE. Students were instructed to deposit filled bags at designated areas in their accommodation complex. Pamphlets containing information on what was suitable for donation were provided as well as strategically deployed posters and TV screens that displayed relevant information.

There were three survey periods, which were tailored to coincide with the students' departure dates at the HoR in June, July and September 2019. The June period covered students on 38-week accommodation contracts, which is usually the choice for undergraduate students. The majority of students leaving accommodation at this time are final-year undergraduates who have completed their studies, or continuing students seeking accommodation elsewhere. The July period covered students on 40-week contracts whilst the September period involved a mixture of postgraduate (Masters) students and "pre-sessional" students improving their English language skills at a summer school before commencing their subject studies.

3.3 Participating charities

The UK charity sector is large and varied, comprising over 160,000 charities with an income of approximately £39 billion (NCVO, 2014). There are currently several schemes and collaborations between UK HEIs and charities involving student donations (Williams and Powell, 2019). The donations help to support causes ranging from education advancement and poverty alleviation to funding medical research. The British Heart Foundation collaborates with over 80 UK universities (Williams and Powell, 2019) via its 'Pack for Good' campaign and the revenue generated from donations helps to fund research into the cure and treatment of heart conditions (BHF, 2020). This charity collects, amongst other items, donated EEE which are sold in their outlets nationwide. The BHF was one of three charities involved in the project (Table 3). Debra is also a national charity which supports epidermolysis bullosa research. Scratch is a regional charity based in Southampton which provides relief effort within the city and surrounding areas and caters for the needs of deprived communities by redistributing reusable items donated such as furniture and small EEE to those in need.

3.4 Product collection and transportation

Collection and transfer of donated items occurred over a period of 4-5 days during which the donated bags were transferred to a central location (Wessex Lane Complex). The logistics (collection, transport and sorting) were planned to cater to the needs of the HoR studied. Due to the varied layout and sizes of both halls, transport arrangements differed slightly. For Mayflower Halls, the initial storage area (where students dropped bags) was a large bicycle shed, which acted as a central location for storage before the bags were moved to the sorting area. The arrangement at City Gateway was slightly different due to the lack of a large storage area. Instead, two vehicles were stationed at the hall, which were used to collect the items before transporting to the central sorting area, at which the team of volunteers inspected each bag and sorted items accordingly.

The set-up (Figure 3) included a dedicated skip to hold unsorted items, gazebos to shelter project crew and equipment and a 1100L storage container (Figure 3A) to hold sorted items awaiting collection by participating charities.

3.5 Sorting and product characterisation

A crew of volunteers was recruited to sort the donated EEE over five days in June, July and September 2019. The contents of each bag were visually inspected, weighed using digital scales and graded according to their physical

TABLE 3: Charities involved in the project.

Charity	Mission	Coverage	Annual Income (£'000)
British Heart Foundation	Support for cardiovascular research	National	138,000 (2018)[1]
Debra	Funding Epidermolysis Bullosa research	National	16,138 (2018)[2]
Scratch	Poverty relief	Southampton/Hampshire	N/A

[1] *https://www.bhf.org.uk/what-we-do/where-your-money-goes*
[2] *https://www.debra.org.uk/downloads/trustees-annual-report-2018.pdf*

FIGURE 3: A. 1100L storage container used for storage; B. Storage container with bagged contents; C. volunteers sorting and inspecting donated items; D. EEE donation examples: printer and computer display monitor; E. EEE donation examples: ICT devices; F. EEE donation examples: small kitchen appliances.

condition (Table 4). Functionality testing of items was not carried out as it was beyond the scope of the study.

Items that were rejected ("broken"; Table 4) were collected separately for recycling. Each donated bag was also weighed, and numbers of commingled items were recorded to evaluate the success rate of the scheme.

The following information was collected for each inspected item:

- Device type;
- Device category (small kitchen appliances (SKA), small home appliances (SHA), personal care appliances (PCA) and information and communication technology devices (ICT/AV);
- Product brand (where identifiable);
- Product condition (visual inspection of products was carried out and each item was given a grade based on its

TABLE 4: Grading system for product sorting. Product rating determined the fate of each item after sorting with products rated "Good" and "Reusable" recovered for reuse while those rated "Broken" were put aside for recycling. Note that products rated 'Good' and 'Reusable' are ultimately reusable and the distinction between the two ratings is based on the physical condition of item.

Condition	**Definitions and examples**
Good	Good as new; saleable (e.g. a printer with all accessories present; kettle with plug in tact)
Reusable	Requires repair/component replacement or upgrade (e.g. kettle with a broken plug; LED TV without remote control)
Broken	Broken/parts missing (e.g. laptop with shattered screen)

physical condition (Good, Reusable or Broken; Table 4)
- Product weight (in kg). Products that could not be weighed were assigned weights of similarly sized variants/models.

3.6 Potential for product reuse

The potential reusability of EEE is largely dependent on the physical condition and functionality of the items collected. Items rated as "Good" (Table 4) are likely to be valued higher due to their condition. Values will also be dependent on other factors such as an item's model, demand and platform/vendor. Estimation of the resale value was based on average prices of commonly-traded EEE, which were obtained from online pre-owned goods trading platforms (www.preloved.com and www.gumtree.com). Prevalent items in the recovery stream were selected for this analysis. In order to account for price variations for each item, 10 price listings were randomly drawn from the aforementioned online platforms to estimate the average price of each item as well as the standard error to account for product price range.

3.7 Material composition analysis

In parallel with the evaluation of product reusability and redistribution, a material composition analysis was carried out to estimate the value of materials contained within the EEE collected. This provided a comparison between reuse and recycling scenarios for recovered items. EEE are known to be a rich source of materials such as ferrous and non-ferrous metals (WRAP, 2012; Meloni, 2020). Products that occurred most commonly in the items collected were selected for material composition analysis. For this analysis, secondary data (average material composition of common household EEE) were used (Parajuly and Wenzel, 2017). Minor materials accounting for less than 5% of an item's weight were excluded in the analysis.

4. RESULTS AND DISCUSSION

4.1 Students' Departures from Halls of Residence

A total of 1,885 student departures were recorded during the study period (Table 5). Mayflower Hall saw significantly more departures in June than City Gateway. The reason for this is partly due to size difference: Mayflower Hall accommodates more students (Table 2). City Gateway Hall houses mostly postgraduate students many of whom opt for longer letting contracts (51 weeks) that end in September. July showed the lowest overall departures (199) whilst September saw the highest (1058).

Seventy percent of overall departures during the study period were international students. This contrasts with the demographic profile of the entire university (30% international students, 70% home students) (HESA, 2020). September saw the highest international student departures (53% of total departures) while the lowest count of international student departures was in July (0.07% of total departures). This profile indicates a high proportion of postgraduate international students' departures in this period, many of whom tend to stay longer due to the duration (a full year) of their degree programmes. Pre-sessional students' departures are also numerous at this period. Mayflower Hall saw a high number of home students' departures in June (71% of Mayflower departures) while a single home student departure from City Gateway was recorded in the same period.

4.2 Collection output

In total 128 bags were collected and sorted (Table 6). From these, 447 electrical and electronic items with total weight of 447.67 kg were inspected and sorted. This equates to approximately 0.24 kg of donated items per departing student. The June collection saw the highest number of bags (77), despite having the second highest number of departures (661). June also accounted for the highest number of donated items of EEE (234) with a total weight of 242.37 kg. This constituted 52% of the number of all items collected and 54% by weight.

With 52 items received, July saw the lowest number of donated items by number and weight (67 kg). This corresponds with the number of departures as the fewest departures (67) at this time. However, July saw the highest collection per student departure at 0.52 kg/student compared with 0.39 kg/student and 0.19 kg/student for June and September, respectively. The overall average collection rate was 0.24 kg/student.

TABLE 5: Student departures for Mayflower and City Gateway Halls in 2019.

Hall of Residence	Month	Home (UK) students	International students	Total
Mayflower	June	437	177	614
	July	30	77	107
	September	14	795	809
City Gateway	June	1	13	14
	July	40	52	92
	September	41	208	249

TABLE 6: EEE donation bag collections from Mayflower and Gateway halls of residence in 2019.

Month	Number of donation bags collected
June	77
July	19
September	32
Total	**128**

TABLE 7: Numbers of items collected during surveys in 2019 and their condition (see Table 4).

Month	Number of items	Good	Reusable	Broken
June	234	101	119	14
July	52	21	28	3
September	161	106	55	0
Total	**447**	**228**	**202**	**17**

TABLE 8: Weights of items collected during surveys in 2019 and their condition (see Table 4).

Month	Weight (kg)	Good	Reusable	Broken
June	242.4	100.7	132.2	9.5
July	55.3	22.4	30.8	2.1
September	150	90.8	59.2	0
Total	**447.7**	**213.9**	**222.2**	**11.6**

A summary of the items inspections and their condition is presented (Tables 7 & 8). Of the 234 items inspected in June, 101 items were rated as “Good” (43%) while 14 items were rated as “Broken” (6%). A similar trend was recorded in July with 21 out of 52 items rated as ”Good” (40%) while 54% of items inspected were rated as “Reusable”. September saw the highest percentage of items rated as “Good” (66%; 106 out of 161 items inspected).

The EEE collected were categorised into: SHA, SKA, ICT and PCA based on the internationally recognised categorisation framework by Forti et al. (2018). The volumes collected for each category varied, however, a greater volume of SHA was collected in June (40%) and September (35%) than other categories. This category includes items such as desk lamps, fans and extension cables. June also saw a high proportion of SKA items (37%). However, the highest proportion of SKA was recorded in July (Figure 4). Approximately 61% of all items collected during this period were SKA. Regarding ICT, 41 and 43 devices were collected in June and September respectively, including some high-value devices in good condition: three LED TVs and six printers. The September collection also included higher-value ICT items: six printers (two Good and four Reusable) and six laptops (five Good and one Reusable).

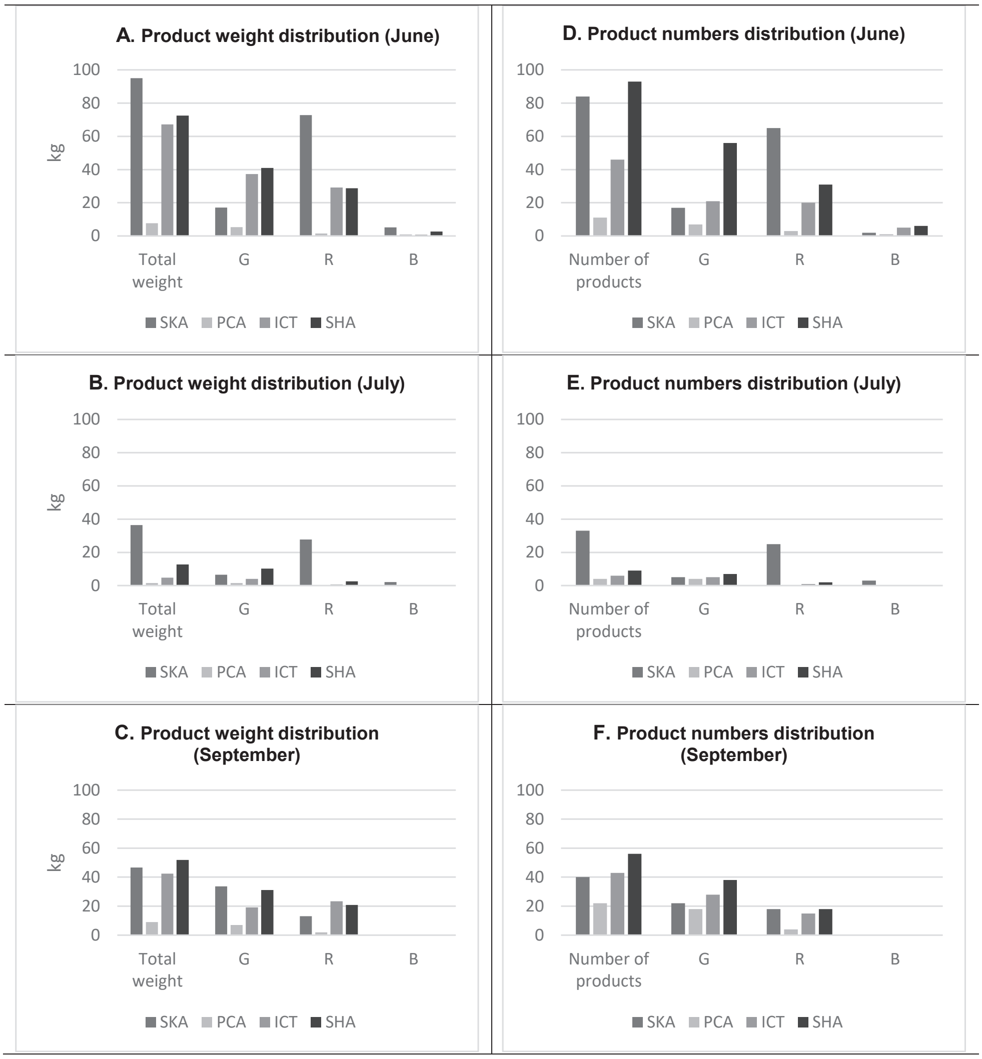


FIGURE 4: Distributions of collected and assessed items and associated grading (see Table 4).

4.3 Reuse potential

A total of 430 out of 447 items collected during the project were either rated as ”Good” or “Reusable”. For each category, over 90% of the items inspected were rated either as “Good” or “Reusable” (Table 9). SKA and SHA products constituted most of the items collected (152 items per category), each with reusability of 97% and 96% respectively. Though the total number of items collected in PCA category was the lowest, this category had the highest reuse rate (Good or Reusable items) at just over 97%. Hair grooming products such as hair dryers and stylers featured prominently in this category.

The resale value was estimated for selected items collected during the study. The potential resale value of the items selected is presented in Table 10.

The resale value of an item is dependent on its physical condition and functionality. The resale values (Table 10) show a range of values of similar items from reuse platforms. From the analysis, the items were estimated to be worth between £2584 and £3758. We note that this estimate is for a subsample of items collected (~36%) and the items collected and assessed for this were from only two HoR in a single HEI (University of Southampton).

4.4 Material composition

A material composition analysis was carried out to estimate the quantities of materials contained in the items collected. EEE are a rich source of materials such as ferrous metals, non-ferrous metals and plastics (WRAP, 2012; Meloni, 2020). A selection of products was analysed based on their prevalence in the stream of items collected (Tables 11 and 12). Products rated as “Broken” were also included for the material composition analysis.

Using compositional analysis data (Table 11), the material composition of a selected group of EEE from the collection was estimated (Table 12). These five products accounted for 36% of the 447 items collected and sorted. From the analysis, 162.6 kg of these four materials were recovered (Fe, Cu, Al and plastics) and constituted 94% of the total weight of these five products. Table 13 presents the value of metals in the prevalent items evaluated. The residual weight is for materials with trace quantities which were not considered in the analysis. Plastics form the bulk of material component (55%) which is expected as small EEE are typically made of over 15 different plastic polymers (Martinho et al., 2012). Copper is the least abundant material fraction in the EEE sampled though at $5,763/tonne (LME, 2020), it is the most valuable material per unit weight.

4.5 Variation in products collected

The study demonstrates the substantial potential for recovering small EEE for reuse and recycling from students departing from university HoR. With 447 items weighing approximately 450 kg (Tables 7 and 8) collected from just two HoR in one academic year, there is a huge potential for

TABLE 9: Product grading by category. Items in each category assessed and graded using grading system adopted (see Table 4). Reusability (%) is the proportion of assessed items that were reusable (products rated ‘Good’ or ‘Reusable’).

Product category	Total	Good or Reusable	Reusability (%)	Broken
SKA	157	152	96.8	14
PCA	37	36	97.2	3
ICT	95	90	94.7	0
SHA	158	152	96.2	17

TABLE 10: Estimated product reuse value of select items collected. Items selected featured heavily in stream assessed. Resale value estimates are based on average price of similar products on online resale platforms; ± standard error of the mean

Item	Average price (pre-owned) (£)	Number of Good or Reusable items collected)	Estimated potential resale value (£)
Iron	9.70 ± 2.00	14	108 – 164
Kettle	9.40 ± 0.80	74	636 – 755
Lamp	21.20 ± 5.00	59	956 – 1546
Toaster	8.80 ± 1.20	39	257 – 390
Printer	51.30 ± 9.50	12	502 – 730
Hair dryer	7.45 ± 1.20	20	125 – 173
Total		**218**	**2584 - 3758**

TABLE 11: EEE average material composition (%). Source data from Parajuly and Wenzel (2017).

Product	Iron (%)	Copper (%)	Aluminium (%)	Plastic (%)
Iron	21.6	7.1	19.3	51.0
Kettle	4.7	5.6	22.0	62.2
Toaster	36.8	3.3	27.3	30.4
Printer	26.1	1.8	0	60.2
Hair dryer	15.7	15.3	0	63.5

TABLE 12: Number and average material composition (kg) of select products collected.

Product	Number collected	Total weight (kg)	Iron (kg)	Copper (kg)	Aluminium (kg)	Plastic (kg)
Iron	14	10.3	2.22	0.73	1.99	5.25
Kettle	77	52.5	2.47	2.94	11.55	32.56
Toaster	39	49.1	18.07	1.62	13.40	14.93
Printer	12	52.2	13.62	0.94	0	31.42
Hair dryer	21	9.4	1.48	1.44	0	5.97
Total	**163**	**173.5**	**37.86**	**7.67**	**26.94**	**90.13**

TABLE 13: Material value of subsample of items (see Tables 11 and 12). Metal values based on London Metal Exchange average prices per tonne at three months forward as of 18/6/2020 (LME, 2020).

Metal	Weight (kg)	£/tonne*	Material value (£)
Fe	37.86	212	80.3
Cu	7.67	4,591	35.2
Al	26.94	1,274	34.3
Total	**72.47**		**149.8**

**Metal values converted to £ sterling using OANDA currency converter (Oanda, 2020)*

the recovery of reusable from this stream if more HoR are involved. The study observed the peak month for collection to be June. This is despite more students departing in September (Table 5). The higher proportion of 'Good' items recorded in September may be indicative of better information assimilation by the students on type and condition of items suitable for donation as the students that moved out at this time had more time to prepare.

Small kitchen appliances and small household appliances constituted the bulk of overall EEE recovered (157 and 158 items respectively out of a total of 447 items). This suggests that such items are purchased or brought in by students moving into halls at high numbers. While the HoR studied provide basic kitchen and household items for shared use, the high level of recovery of items in these categories suggests that students opt to bring in or purchase their own. This may be more common amongst international students (70% of total departures) who may have brought with them items like kettles, sandwich makers; some of the product brands were from outside the UK. While it is difficult to attribute items donated to individual students, it is safe to assume, due to large percentage of international students in the sample population, that departing international students are more likely to donate items deemed excess, making them a potentially viable group to target for a reuse scheme (Williams and Powell, 2019).

The present study observed a high level of compliance from students regarding the condition of items collected; only a small fraction of items collected did not meet criteria as specified in the guidance and were deemed unsuitable for reuse. The majority of these unsuitable items were received in June (Table 14) and were mostly SHA. The level of compliance resulted in a contamination rate of less than 5% during each month of collection (with an overall contamination rate of 2.6%). These products, while not available for reuse, retain resource value and material recovery can occur via recycling.

The HoR studied provide facilities for WEEE recycling in the form of storage bins and yards which may have contributed to the low rates of contamination since students already have an option to recycle broken/non-functional EEE. Studies have shown that without such facilities, such items are likely to be disposed of in general waste (Ongondo & Williams, 2011; Pierron et al., 2017). Pierron et al. (2017) also observed high likelihood of end-of-use PCA disposal as opposed to being recycled. This may explain the low prevalence of PCA collection observed during this study.

4.6 Reuse and recycling potential

The UK has over 150 HEIs and each HEI, in principle, is potentially an urban mine rich in items and resources that can be reintroduced into the circular economy (Pierron et al., 2017). According to the Higher Education Statistics Agency (2020), 351,605 students live in university-owned HoR/accommodation (2018/2019 data) across the UK. Assuming a reusable EEE recovery rate of 0.24 kg/student as observed in this study, there is a UK-wide potential for recovery of 84 tonnes of EEE for reuse annually. This potential value excludes other students living outside HoR who constitute the majority; of the over 2 million students

TABLE 14: Contamination rate of EEE collection (Contamination rate: proportion of contaminants in total items collected i.e. contaminants (kg)/total items (kg) as %. Contaminants in this study comprise items rated 'Broken').

Month	Total weight (kg)	Weight (Good & Reusable) (kg)	Weight (Broken) (kg)	Contamination rate (%)
June	242.4	232.9	9.5	4.1
July	55.3	53.2	2.1	4.0
September	150	150	0	0
Overall	447.67	435.87	11.6	2.6

enrolled at universities during 2018/19 academic year, only 17.5% students resided in HoR (HESA, 2020).

The observed subsample of items (Table 10) was estimated to yield potentially an estimated resale value of £2,600-£3,700. This equates to £1.30-£1.90 per student (from a student population of 1,885). Assuming that this figure is broadly representative of UK university students, this represents a monetary value of up £485,000-£690,000 for all students living in UK university accommodation nationwide in the 2018/19 academic year. Scaling this up for total student population, assuming the same monetary value per student, and that students in other accommodation undergo periodic clear-out, the student population at University of Southampton (24,625 students) and UK universities (2,383,970 students), the monetary potential is up to £46,000 and £4,500,000 respectively (using a value of £1.90 per student). On the other hand, the material value of the same subsample was estimated to be £27,942 for a student population of 361,605 and £189,453 for entire student population in the UK. These data suggest a high potential for reuse as these estimates cover a subset of all items recovered and assessed. This potential could be higher considering that the products, apart from their reuse value, also possess material value which can be exploited via recycling at their end of life. While resale value is likely to plummet with each product usage cycle, the materials contained (especially metals) will retain their value.

4.7 Process analysis

Schemes involving donation of unwanted and/or end of use items with reuse value is commonplace in UK HEIs. Schemes such as 'Shift your Stuff' organised by the Student Union at the University of Southampton have been planned to coincide with the departure periods of students and encourage the donation any unwanted items when they move out. Schemes run in the past by Students' Union collected items such as clothing, homeware and other bric-a-brac but excluded collection of reusable EEE. The present study is the first of its kind that specifically studied the outcome of source-segregated EEE donations. Comingling with broken EEE was minimal as observed in the contamination rate (Table 12), indicating that information provided was largely understood. As stated previously, the study was preceded by a period of information disseminated using different media, including strategically-located posters and pamphlets (Figure 5).

The study had three major components; collection/recovery of EEE from departing students, sorting and characterisation of the items received and the redistribution which involved select charities (Figure 6). Interventions made for the collection/recovery component included provision of bags as well as temporary storage areas to hold the EEE. With this, the protocol differed between the two HoR studied. Mayflower Halls has a large sheltered bicycle shed which doubled as a temporary storage (Infrastructure aspect of ISB model) hold for the EEE collected. The situation was different at City Gateway Hall, which lacks an adequate storage area; instead, the collected items were stored temporarily in vans. This is an example of infrastructure differences which required different interventions as postulated by the ISB model (Timlett & Williams, 2011).

The EEE collected went through a process of screening and sorting. The activities could not be feasibly carried out at the sites of collection (HoR) for logistical reasons. While Mayflower Halls complex has a storage space which, in principle, could have been used for product sorting, City Gateway lacks such a facility and the small number of volunteers recruited for the project were necessarily co-located. As a result, all items collected were moved to another location where sorting took place (Figure 2).

The study recorded a high reuse rate overall (Table 9). The assessment of products received (Table 4) was necessarily based on visual inspection. Several items received required cleaning while some others were missing minor components. However, a more robust assessment such as functionality testing was not carried out as it was beyond the scope of the project. Whilst there is merit in undertaking robust functionality tests (Parajuly and Wenzel, 2017), addition of a testing stage to the protocol would incur additional resources and costs. It is probable that a functionality test would have resulted in a higher rejection/contamination rate. An idealised scenario would involve recovery and

What goes in my red bag?

Kettles, Toasters, Blenders, Rice cookers, Humidifiers, Fans, Heaters, Coffee machines, Grills/ appliances, TVs, Laptops, PC equipment, Fairy lights, Irons, Hairdryers, Printers, Lamps & lights

What can't go in?

If it's broken, we can't take it! Also only items that have the BSI kitemark and or the CE logo on the plug can be donated.

Broken Electrical Items - You can dispose of any damaged or broken waste electrical items by taking them to the WEEE (waste electrical and electronic equipment) bin at your site.

Electrical Items

Please donate these kinds of items here:

Kettles
Toasters
Blenders
Rice Cookers
Humidifiers
Fans
Heaters
Coffee Machines
Grills/appliances
TVs
Laptops
PC Equipment
Fairy Lights
Irons
Hairdryers
Printers
Lamps/Lights

Is there anything I can't donate here?

We are unable to reuse broken electricals, items without a CE marking / BSI kitemark or badly scratched items so please don't leave them here.

IMPORTANT - Please think about whether your item can actually be used before leaving it here. If it's too old or it is soiled you should dispose of it in the general waste. If you are unsure, please read the information below.

✓ My item is clean, has no clear signs of wear and is 'as new' / My item is no longer 'as new' but is in full working order and could be resold. Please donate it here if it has either a CE marking or BSI Kitemark on the plug

● My item is pretty scruffy, it has a few minor defects but I would still feel okay giving it to someone in need. Be realistic, only donate if it is reusable otherwise this can be a waste of a charity's time

✗ My item is broken, has a lot of scratches or has a defect that means it can't easily be reused. Please do not donate it here. Recycle / bin it as appropriate.

ECOBOARD

FIGURE 5: Some examples of media used to influence behaviour change by providing information on the project (Source: University of Southampton Residential Services, 2019).

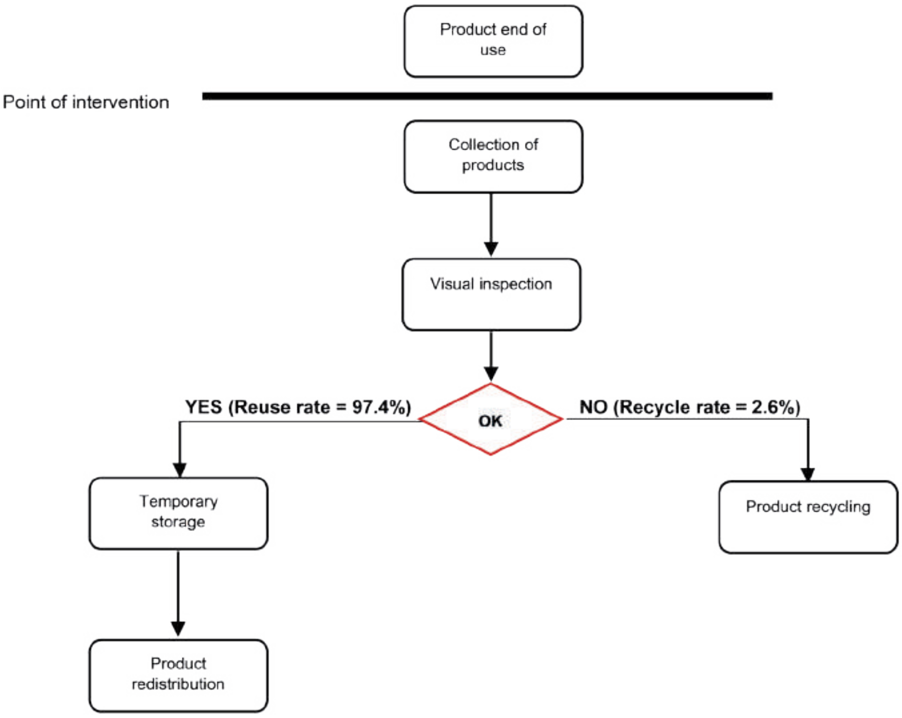


FIGURE 6: Project protocol and processes including the interventions made using ISB model (I: temporary storage; S: collection and inspection of products; B: products are redirected and made to pass through the system which recovers items with residual reuse value).

movement of products and materials within a closed loop (Figure 7) together with ancillary services for product testing, repair/upgrade, storage and redistribution. However, a recovery system based on reuse can still accommodate items not deemed for this purpose (reuse) as such items still possess some value and can be recycled.

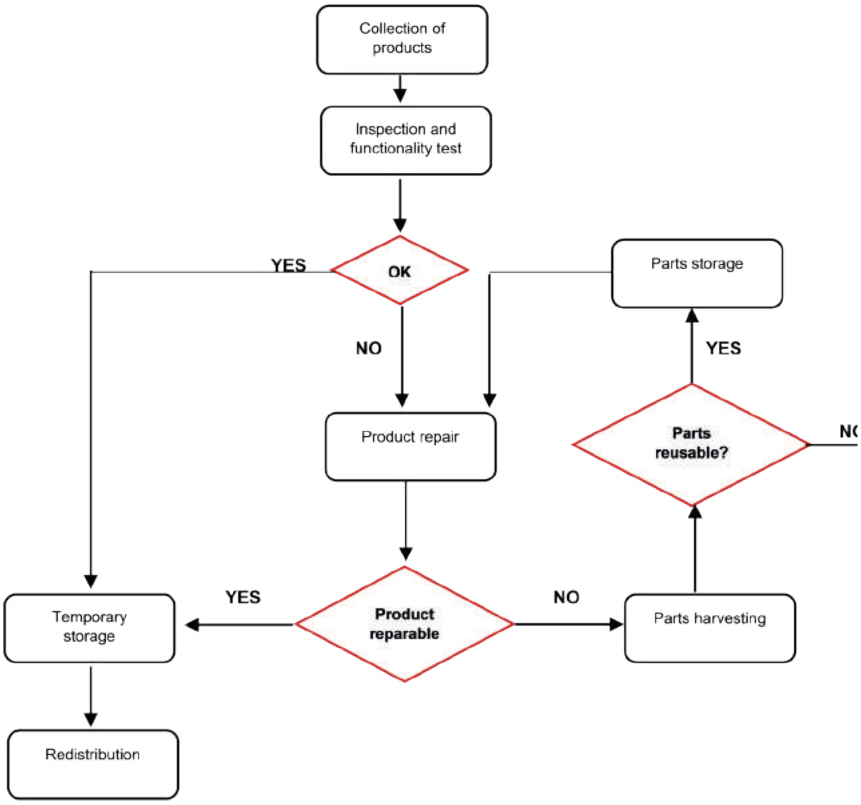


FIGURE 7: A reuse-based collection and redistribution protocol for EEE recovery in a university DUM including the relevant ancillary processes.

TABLE 15: Cross-sectoral benefits of a reuse-based EEE recovery system.

Stakeholder	Benefit	Benefit category
Charities/NGOs or other recipients	Receipt of good quality reusable items at little cost; redistribution of items	Economic; Social
Universities/HEIs	Lower disposal expenditure; enhancement of 'green' credentials	Economic; Environmental
Students	'Warm glow' (intrinsic factor) of contributing towards environmental sustainability	Psychological
Waste/environmental manager	Carbon savings, resource conservation; circularity	Environmental; Economic

For this study, the involvement of charities proved effective in the redistribution and reuse of products collected. As previously noted, there are national charities that have partnered with HEIs in schemes involving product recovery and redistribution, diverting reusable products from landfill or recycling. Future collaborations in such schemes, as demonstrated in this project, will yield positive environmental, social and economic impacts. Examples of such impacts are highlighted in Table 15.

These benefits are universally derivable irrespective of location. While this study was undertaken in the UK, adoption of a similar strategy in similar settings (i.e. HEIs) elsewhere will potentially yield similar outcomes. The UK, due to its well-established charity sector, provides a platform through which recovered reusable products can be redistributed for resale/reuse. Similar organisations elsewhere in form of NGOs (non-governmental organisations) are potential benefactors and can perform similar roles. A key factor of the reuse-based recovery system is the transient nature of university/HEI student population. Due to periodic turnover of students, there will be predictable and repeated opportunities for recovery of reusable EEE from departing students or those changing accommodation. While the frequency of this annual turnover within an academic year may vary from country to country, this unique factor allows for global replicability of the reuse-based EEE recovery system.

4.8 Future perspectives

Circularity provides a means to keep products (including EEE) and materials in use for extended periods and presents an opportunity to reduce negative environmental impacts. A successful implementation of the circular economy model will be dependent on factors such as product design and reverse logistics as well as having an enabling environment (Meloni, 2020). Product reuse can become a mainstay, with adequate interventions, from production to end of use. Products designed with ease of disassembly and/or repair are likely to be in a closed loop longer as such products can retain functionality for longer periods, potentially changing owners during their lifetime. Going forward, interventions such as using modular designs would become crucial for incorporating circular economy principles in product design.

Reverse logistics is another crucial element of the circular economy model. As this study has shown, providing the means to recover and redistribute reusable items can provide economic, societal and environmental gains (Table 14). With organisations such as charities available to absorb and aid redistribution of such items, they provide solutions within a circular economy by either selling or donating to potential new owners. As noted by Meloni (2020), movement of products between different categories of users, e.g. high-end users to lower/emerging users, ensures that products can be made available to cater to the needs and constraints of different users during their lifetime. While such movements have been labelled as 'dumping,' particularly when involving product movement from developed to developing countries, providing a high-quality reuse standard for pre-owned items would ensure that such products undergo rigorous testing and certification before being moved on to new owners.

Attitudes towards pre-owned EEE are a potential barrier to reuse. Improving perceptions of reuse could be a significant step towards circularity. Again, a high-quality reuse standardisation and certification provides an opportunity to alter perceptions. This post-use quality assurance system such as that used in studies such as Hickey et al. (2014) and Dietrich (2014) involves testing, upgrading and certifying pre-owned EEE before redistribution to new owners. A system such as this provides a 'seal of quality' which will help alter perception on pre-owned but functional EEE.

5. CONCLUSIONS

This study clearly provides evidence that a reuse-based recovery system for small EEE significantly increases the urban mining potential of a university DUM while creating an avenue to provide a platform for extending the life time of small EEE. While there were logistical challenges, the project demonstrates a workable proof-of-concept for a reuse-based recovery and redistribution system within a university DUM.

The study presented product reuse as a priority for end-of-use EEE for sound but unwanted products within a DUM as opposed to an inadvertent and unconscious drift towards less desirable options. It provides an important insight into the significant potential for recovering reusable small EEE from a DUM, in this case, students within a university urban mine. The study presents, for the first time, data on recovery of reusable small EEE from departing students from university HoR. Through applying a protocol informed by the ISB model (Timlett & Williams, 2011), the transfer of items from donors (students) to beneficiaries (participating charities) has been demonstrated to support the case for reuse as the preferred end-of-use decision for products with good functionality and, in so doing, result in positive environmental, economic and social impacts. As this was a demonstration project, the potential for impact if replicated nationwide is highly significant in terms of increased diversion from landfill, resource efficiency, materials recovery at end-of-life, reduction of adverse environmental effects, and social and economic benefits. This study also highlights the role individual young people can play when they act in concert for societal benefit and the global replicability of the reuse-based system as a viable route to circularity of EEE.

This project was aided by a communication campaign, which elicited a positive behavioural response from the students, as well as provision of necessary infrastructure and service. This demonstrates that the ISB model can bring about desired changes in addressing waste management issues. It is recommended that the adoption of similar systems in universities and other HEIs within the UK and globally has tremendous potential to divert several tonnes of reusable EEE from landfill as well as providing social and economic benefits.

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DETECTING CONTAMINANTS IN POST-CONSUMER PLASTIC PACKAGING WASTE BY A NIR HYPERSPECTRAL IMAGING-BASED CASCADE DETECTION APPROACH

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ABSTRACT

Recycling of post-consumer packaging wastes involves a complex chain of activities, usually based on three main stages, that is: i) collection from households or recovery from Municipal solid waste (MSW), ii) sorting and, finally, iii) mechanical recycling. Among these activities contaminants detection and separation play a preeminent role. The utilization of a Near InfraRed (NIR) – HyperSpectral Imaging (HSI) based methods, along with chemometrics and machine learning techniques, can fulfill both the two previously mentioned goals. In this paper, the HSI-based sorting logics, to apply, to implement, to set up and to perform an automatic separation of paper, cardboard, plastics and multilayer packaging are investigated. The built PLS-DA-based cascading classification model allows to recognize polymeric fragments from cellulosic ones and to identify multi-layer materials (i.e. laminated plastic and laminated cardboard). The misclassified fragments are constituted by laminated plastics. The set up cascade model reached in prediction a Recognition and a Reliability of 0.933. The proposed NIR-HSI-based approach can represent an optimal, reliable and low-cost answer to systematically identify impurities and composite materials inside plastic waste streams.

1. INTRODUCTION

Post-consumer plastic packaging waste is one of the primary sources to recover polymers (Bonifazi et al., 2015a). The recycling of this waste involves different complex activities, involving the collection from households or the recovery from Municipal Solid Waste (MSW) of plastic packages and the further sorting and mechanical recycling to washed milled goods (Jansen et al., 2015).

The complexity of the processing phases is linked to the heterogeneity of this waste stream, mainly due to the presence of contaminants and cellulose fragments, and secondly to the presence of multi-layer materials. Those materials, for their characteristics, are difficult to separate by classical methods (i.e. gravimetric separation), thus ending up in the recovered plastics products.

The properties of waste materials, their characteristics, and physical-chemical attributes can be investigated by using sensing devices that are able to acquire objects' spectral characteristics attributable to a specific wavelength range. The further processing of these spectra can allow the identification of the object spectral signatures and their classification. This approach is currently used to set-up and develop on-line procedures aimed to recognize different materials occurring in a waste stream as they result after specific processing/selection actions (Bonifazi et al., 2009). In recent years, industrial operations concerning automated sorting of plastic packaging strongly increased thanks to technology improvement, especially that based on Near InfraRed-HyperSpectral Imaging (NIR-HSI) (Hollestein et al., 2015).

HSI, in fact, represents an attractive solution to characterize, to control and to perform the quality control of different materials in many sectors, i.e. cultural heritage (Capobianco et Al., 2015; Agresti et Al., 2013), agricultural/food industry (Serranti et Al., 2018a; Serranti et Al. 2018b; Kumaravelu and Gopal, 2015; Tsuchikawa and Kobori, 2015), in the pharmaceutical and chemical industry (Roggo et Al. 2007; Larrechi et Al. 2003), and more generally in analytical science (Pasquini, 2003). In recent decades, HSI has emerged in the recycling sector (Bonifazi et Al. 2015b; Serranti et Al. 2015; Serranti et Al. 2013; Palmieri et Al. 2014; Hu et al., 2013; Ulrici et al., 2013; Serranti et Al., 2011).

In this paper, a Near InfraRed (NIR)-based hyperspectral imaging (HSI) method to perform real-time identification of

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different materials occurring in a post-consumer packaging waste stream, such as the polymeric fraction, the cellulose-based fraction and the multi-layers material fraction (i.e. laminated plastics and laminated cardboard) resulting from a recycling process, is presented.

The possibility to utilize a cascade classification model based on a two-step Partial Least Squares – Discriminant Analysis (PLS-DA) to identify cellulose-based fragments from polymer/plastic-based fragments, thus recognizing laminated plastic from polymer/plastic-based fraction and laminated cardboard (i.e. Tetra Brik) from the cellulose-based fraction, is explored (Figure 1).

2. MATERIALS AND METHODS

2.1 Analyzed materials and sampling

The materials were sampled from sorted output resulting from a material recovery facility (MRF) of post-consumer household plastic packaging waste, located in the Lazio region (Italy). Samples were collected from a waste bale composed of post-consumer packaging fragments, resulting from manual and semi-automatic sorting (oversize of a drum sieve separator). Ideally, the output of the sorting line should not contain paper-based material. However, the quality analysis performed on the 2D output of the facility highlighted the presence of paper-based packaging and similar material (i.e. laminated card): about 3.5% in weight of the analyzed material consists of paper-based packaging fragments. Coning and quartering method was used to reduce the sample size to collect. The amount of material collected was about 5 kg (i.e. 426 fragments). 81 particles were then selected from this pool in order to represent the typical fragments usually belonging to this fraction. For calculating the sample size (i.e. number of investigated particles) was used the simplified Yamane's (1967) formula:

$$n = \frac{N}{1+N(e^2)} \qquad (1)$$

where n is the number of sampled particles, N is the total number of particles constituting the collected material, and e represents the level of precision. A 90% confidence level was assumed for this equation. The fragment size ranges from 2 to 10 cm.

Paper, cardboard and plastic. In order to build a model able to recognize paper-based fragments from polymer-based particles, 40 mixed plastic polymer-based and paper-based particles were used as a training set (Figure 2): 24 plastic/polymer-based particles (including laminated plastic) and 16 paper-based particles (including laminated card and paper).

A total of 30 particles were used as a validation set, subclustered in two sets: "Validation set 1" and "Validation set 2" (Figure 3). Sixteen particles were included in "Validation set 1": 11 plastic/polymer-based particles (including laminated plastic) and 5 paper-based particles (including laminated card and paper). While 14 particles were included in "Validation set 2": 9 plastic/polymer-based particles (including laminated plastic) and 5 paper-based particles (including laminated card and paper).

Laminated card. In order to recognize laminated from other paper-based particles, 5 paper-based particles to be included in the calibration set were additionally analyzed (Figure 4).

Laminated plastic. To set-up a model able to discriminate laminated plastic from other polymer-based particles, 6 laminated plastic fragments were included in the calibration set (Figure 5).

2.2 HyperSpectral Imaging system and data handling

HyperSpectral Imaging (HSI) is a technique, based on a specific sensing architecture, that allows to digitally collect

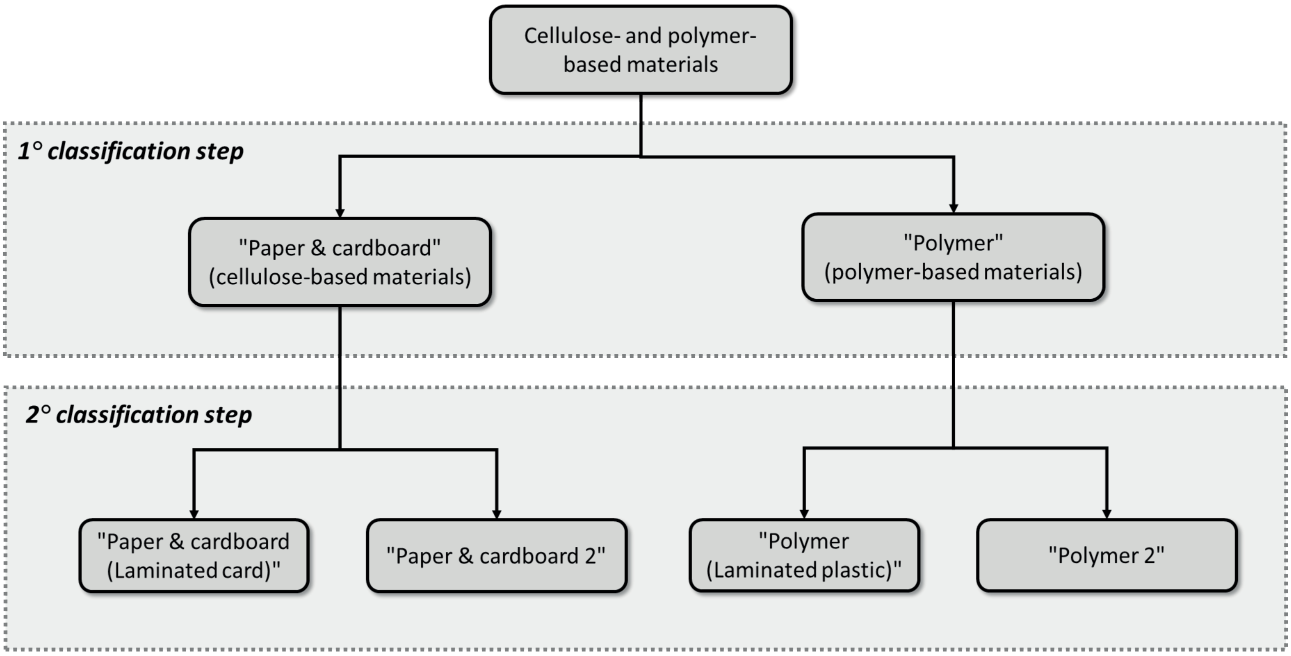


FIGURE 1: Decision tree describing the two-stage cascaded classifier applied to a post-consumer packaging waste stream to perform product classification.

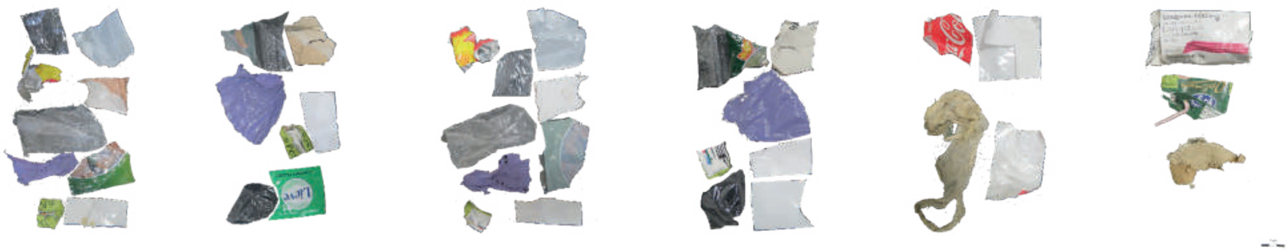
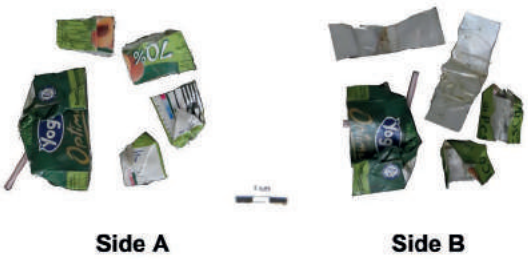


FIGURE 2: Digital images of particles included in the calibration set to build a model able to recognize paper-based fragments from polymer-based particles.

a

b

FIGURE 3: Digital images of particles included into the validation set: "Validation set 1" (a) and "Validation set 2" (b).



Side A

Side B

FIGURE 4: Digital images of laminated card particles. Side A: 5 fragments with the plastic/paper/metal/plastic side exposed and Side B: 4 fragments with the plastic/metal/paper/plastic side exposed.

Side A

Side B

FIGURE 5: Digital images of laminated plastic particles. Side A shows the fragments with the plastic/metal side exposed, while side B the metal/plastic side.

the spectrum associated with each pixel of the collected image. The acquired information is arranged in a three-dimensional (x, y, λ) dataset, called hyperspectral cube or "hypercube".

The hyperspectral device utilized in this study works in the NIR range (1000-1700 nm). The sensing unit is constituted by a SpecIm's NIR spectral camera consisting of an ImSpector N17E imaging spectrograph, developed by SpecIm™ Oy, with a spectral sampling/pixel of 2.6 nm coupled with a temperature-stabilized InGaAs photodiode array (320 × 240 pixels in image frame), positioned above a light source (Serranti and Bonifazi, 2010; DV Optics S.r.l., 2008). The illuminant system consists of a diffused light cylinder architecture, embedding five halogen bulbs. The equipment, connected to a PC, is mounted on a conveyor belt (width = 26 cm and length = 160 cm) that can operate at variable speed (variable between 0 mm/s and 50 mm/s).

The calibration of the spectrograph was performed by recording an image with the lens of the camera completely closed, dark image acquisition (D_i), and by measuring the "white reference image" (W_i) on a standardized white Spectralon® ceramic material. The dark reference was used to remove the effect of the CCD (charge-coupled device) dark current. After this calibration stage, the spectra image (R_{0_i}) is acquired and the reflectance (R_i) of the acquired image is then computed, by using the equation:

$$R_i = \frac{R_{0_i} - D_i}{W_i - D_i} \tag{2}$$

The calibration procedure was performed via Spectral Scanner™ v.2.3 software.

2.2.1 Data acquisition

Hyperspectral image acquisition was carried out setting the speed of the conveyor belt equal to 10 mm/s (for 1000 frames). The 40 particles utilized to develop a model able for discriminating paper-based fragments from polymer-based particles (i.e. training set), were acquired in 6 hyperspectral images. To set-up the model to discriminate laminated card (i.e. Tetra Brik package fragments) from other cellulose materials, 5 laminated particles were acquired in 2 hyperspectral images from 2 sides: one image showing the plastic/paper/metal/plastic side (Side A) and the other image showing the plastic/metal/paper/plastic side (Side b) (Figure 4). To set up the model to identify laminated plastic fragments from other polymer-based particles, 6 laminated plastic fragments were acquired from 2 sides in 4 hyperspectral images: 2 images with the particles' plastic/metal side exposed (Side A) and 2 images with particles' metal/plastic side exposed (Side B) (Figure 5). The data set to validate the models was constituted by 16 particles included in "Validation set 1", scanned in 2 hyperspectral images, and 14 particles in "Validation set 2" also acquired in 2 hyperspectral images.

Acquired hyperspectral images ".sif" were converted into ".hdr" files and imported into MATLAB® environment (MATLAB R2018a ver. 9.4) as dataset objects. Imported datasets were thus analyzed using Eigenvector Research's PLS_toolbox (ver. 8.7) and MIA_toolbox (ver. 3.0) in MATLAB® environment.

2.2.2 Data pre-processing and explorative analysis

The spectral range of each acquired hyperspectral image was firstly reduced from 950-1700 nm to 1000-1650 nm, excluding the noisiest parts of the spectra. Background removal was performed for all the hyperspectral images. Object silhouettes were cropped, and the background was excluded via the PLS_toolbox 'Image_Flatfield' function.

Regions of Interest (ROIs) were selected on the training set images and mean raw reflectance spectra were extracted. More in detail, 39 ROIs were selected from the 6 hyperspectral images of the "Training set": 23 ROIs of polymer/plastic-based particles and 16 ROIs of paper-based particles ("Paper & cardboard") as shown in Figure 6.

Spectra of the 39 ROIs were thus extracted and con-

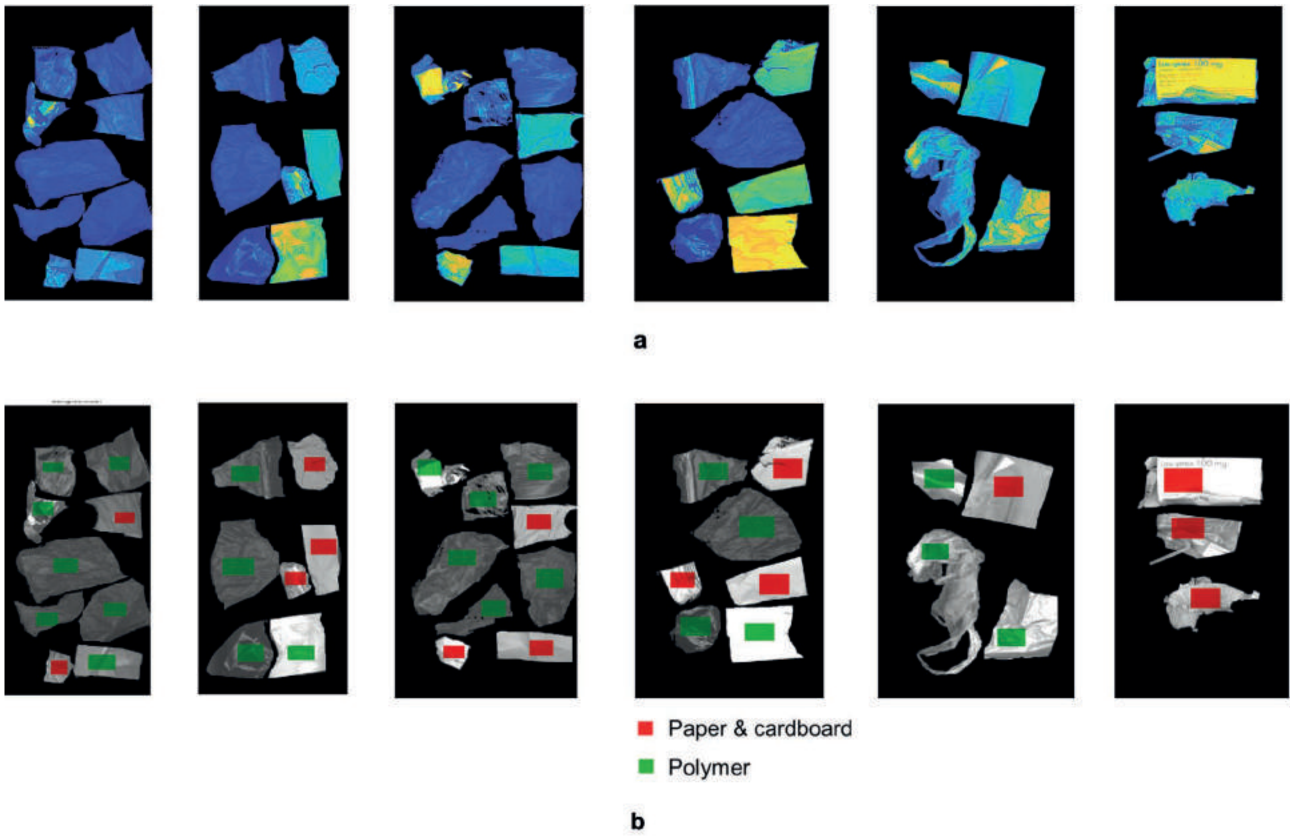


FIGURE 6: "Calibration set": hyperspectral images (a) and selected ROIs of "Paper & cardboard" and "Polymer" (b). Red: Paper & cardboard, green: Polymer.

catenated to create a unique dataset (for the classes “Polymer” and “Paper & cardboard”). The model calibration to discriminate laminated card (i.e. Tetra Brik package fragments) from other cellulose materials (Figure 7) was carried out by extracting 15 ROIs from 2 acquired hyperspectral images (8 ROIs from side A and 7 ROIs from side B: side A is the plastic/paper/metal/plastic side, while side B is the plastic/metal/paper/plastic side). Spectra from extracted ROIs were used to create a new dataset for class “Paper & cardboard (laminated card)”.

Figure 8 shows the 4 concatenated hyperspectral images depicting laminated plastic particles included in the calibration set and the extracted 30 ROIs (14 ROIs on side A and 16 ROIs on side B: side A is the plastic/metal side, while side B is the metal/plastic). Spectra from extracted ROIs were used to create a new dataset for class “Polymer (Laminated plastic)”. While the hyperspectral images of the “Validation set 1” were only concatenated together, as shown in Figure 9. The same procedure was applied to “Validation set 2”.

A combination of pre-processing algorithms (Rinnan et al., 2009) was applied to spectral data used to train each step of the cascading classifier. Data pre-processing is not only necessary to implement both scattering corrections and noise removal procedures, but also to enhance differences occurring among clusters of data classes. Different pre-processing strategies were sequentially applied. The one giving the better response in terms of data decomposition

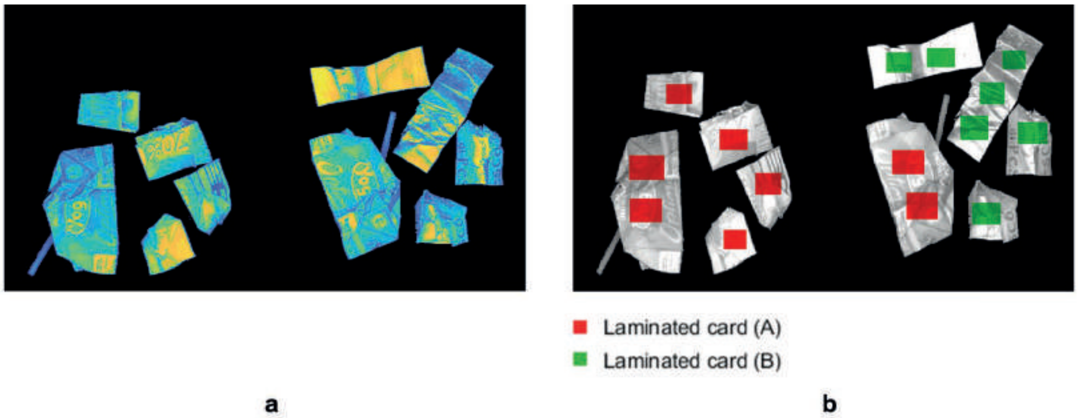


FIGURE 7: Laminated card particles (side A is the plastic/paper/metal/plastic side, while side B is the plastic/metal/paper/plastic side): a) concatenated hyperspectral images of the particles and b) ROIs selected on the hyperspectral images (Red: laminated card - side A, green: laminated card - side B).

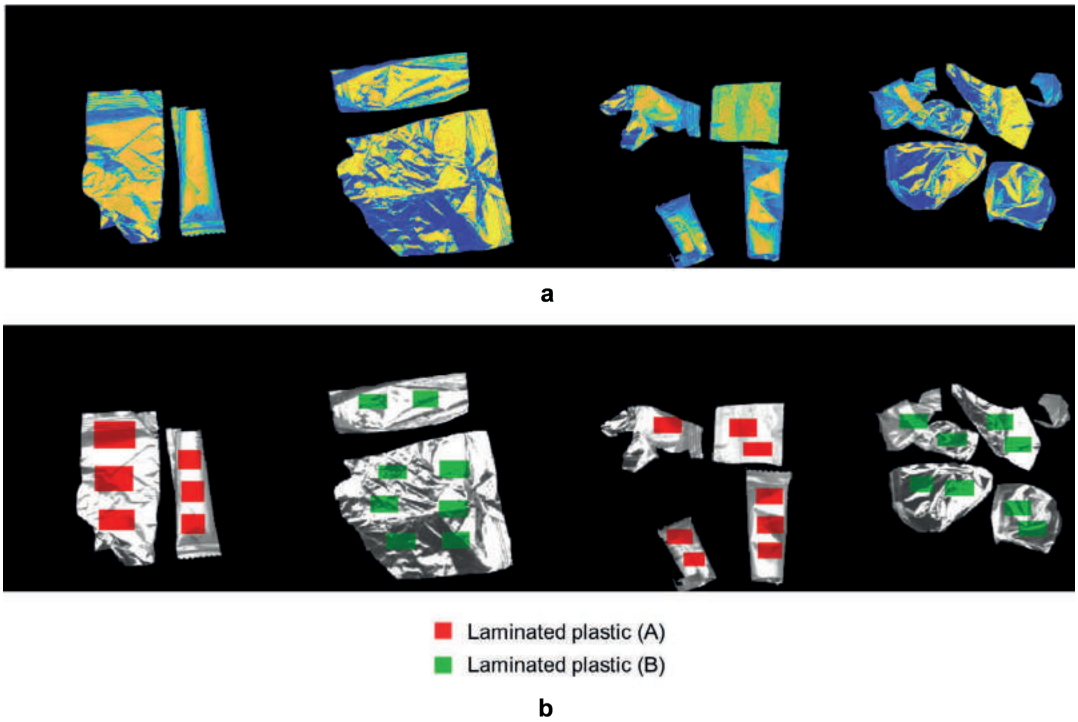


FIGURE 8: Laminated plastic particles (side A is the plastic/metal side, while side B is the metal/plastic): a) concatenated hyperspectral images of the particles and b) ROIs selected on the hyperspectral images (Red: laminated plastic - side A, green: laminated plastic - side B).

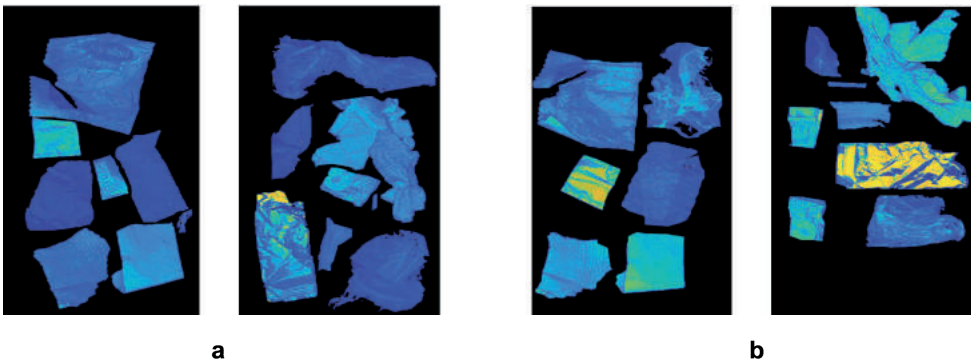


FIGURE 9: Hyperspectral images of "Validation set 1" (a) and "Validation set 2" (b).

was thus chosen, that is: Standard Normal Variate (SNV), Smoothing and Mean Center (MC) (Savitzky and Golay, 1964; Rinnan et al., 2009; Eigenvector, 2017; Eigenvector, 2018).

SNV consists of a scatter-corrective pre-processing method and it is one of the most widely used pre-processing techniques in NIR spectroscopy (Rinnan et al., 2009; Eigenvector, 2017). The smoothing algorithm is a low-pass filter used to remove high-frequency noise from samples. This pre-processing, based on the Savitzky-Golay algorithm, (Savitzky and Golay, 1964) is often used on spectra. Smoothing operates separately on each row of the spectra matrix acting on adjacent variables. Adjacent variables, containing similar information, are averaged together without significant loss of the spectral information. Finally, MC pre-process algorithm centers columns to have a zero mean (Eigenvector, 2018).

Principal Component Analysis (PCA), a well-known unsupervised pattern recognition technique, was chosen to perform the exploratory analysis of decomposed spectral data, according to sample type and for excluding outliers from the datasets (Wold et al. 1987; Rivsik, 2007; Wise et al., 2008).

2.3 Cascade Model based on Partial Least Squares – Discriminant Analysis (PLS-DA)

A two-step cascade model based on Partial Least Squares-Discriminant Analysis (PLS-DA) (Figure 1) was built and validated. PLS-DA is a supervised pattern recognition technique consisting of a multivariate inverse least squares discrimination method. Partial Least Squares (PLS) regression is used to develop a model able to predict the class of each sample (i.e. pixel) under study (Wise et al., 2008).

The first step of the cascade classification model was performed to identify cellulose-based fragments ("Paper & cardboard") from polymer/plastic-based fragments ("Polymer"). The second step of the classification was performed in order to discriminate Laminated card ("Paper & cardboard (Laminated card)") from the other cellulose-based particles ("Paper & cardboard 2") and to identify laminated plastic fragments ("Polymer (Laminated plastic)") from other polymer-based fragments ("Polymer 2"). The first model was calibrated with the classes "Paper & cardboard" and "Polymer" by using the spectra extracted from ROIs selected on the calibration set (Figures 10a and 10b). Venetian-blinds, as a cross-validation method, were utilized to perform model tuning and to choose "the right grade of complexity" (Eigenvector, 2016): 3 Latent Variables (LVs) were used. The model was validated on "Validation set 1" and "Validation set 2", to assess the ability of new "unknown" sample prediction.

The second step of the cascading model was calibrated using:

i) the "Paper & cardboard (Laminated card)" belonging to the selected ROIs, as shown in Figures 10c and 10d, to perform the discrimination between "Paper & cardboard (Laminated card)" and "Paper & cardboard 2" and
ii) the "Polymer (Laminated plastic)" belonging to the selected ROIs, as shown in Figures 10c and 10d, to perform the discrimination between "Polymer (Laminated plastic)" and "Polymer 2".

In the first case, 5 LVs were used. The model was validated on "Validation set 1" and "Validation set 2", with the polymer-based particles removed from the analysis. In the second case, 5 Latent variables (LVs) were used. The model was validated on "Validation set 1" and "Validation set 2", with the cellulose-based particles removed from the analysis.

The confusion matrix, adopting a pixels-based logic, was considered to evaluate classifier performance in each of its steps and the commonly used performance metrics calculated from it. Sensitivity, Specificity, Precision, Accuracy, Misclassification Error and Class Error were calculated from the confusion matrix (Ballabio and Todeschini, 2009; Fawcett, 2006), which are:

$$Sensitivity = \frac{TP}{TP+FN} \tag{3}$$

$$Specificity = \frac{TN}{FP+TN} = 1 - Sensitivity \tag{4}$$

$$Precision = \frac{TP}{TP+FP} \tag{5}$$

$$Accuracy = 1 - \frac{TP+FN}{TP+TN+FP+FN} = \frac{TP+TN}{TP+TN+FP+FN} \tag{6}$$

$$Misclassification\ Error\ (ERR) = 1 - Accuracy \tag{7}$$

$$Class\ Error = 1 - \left(\frac{Sensitivity + Specificity}{2}\right) \tag{8}$$

where: *TP* (True Positive) is a positive instance that is classified as positive; *FN* (False Negative) is a positive instance

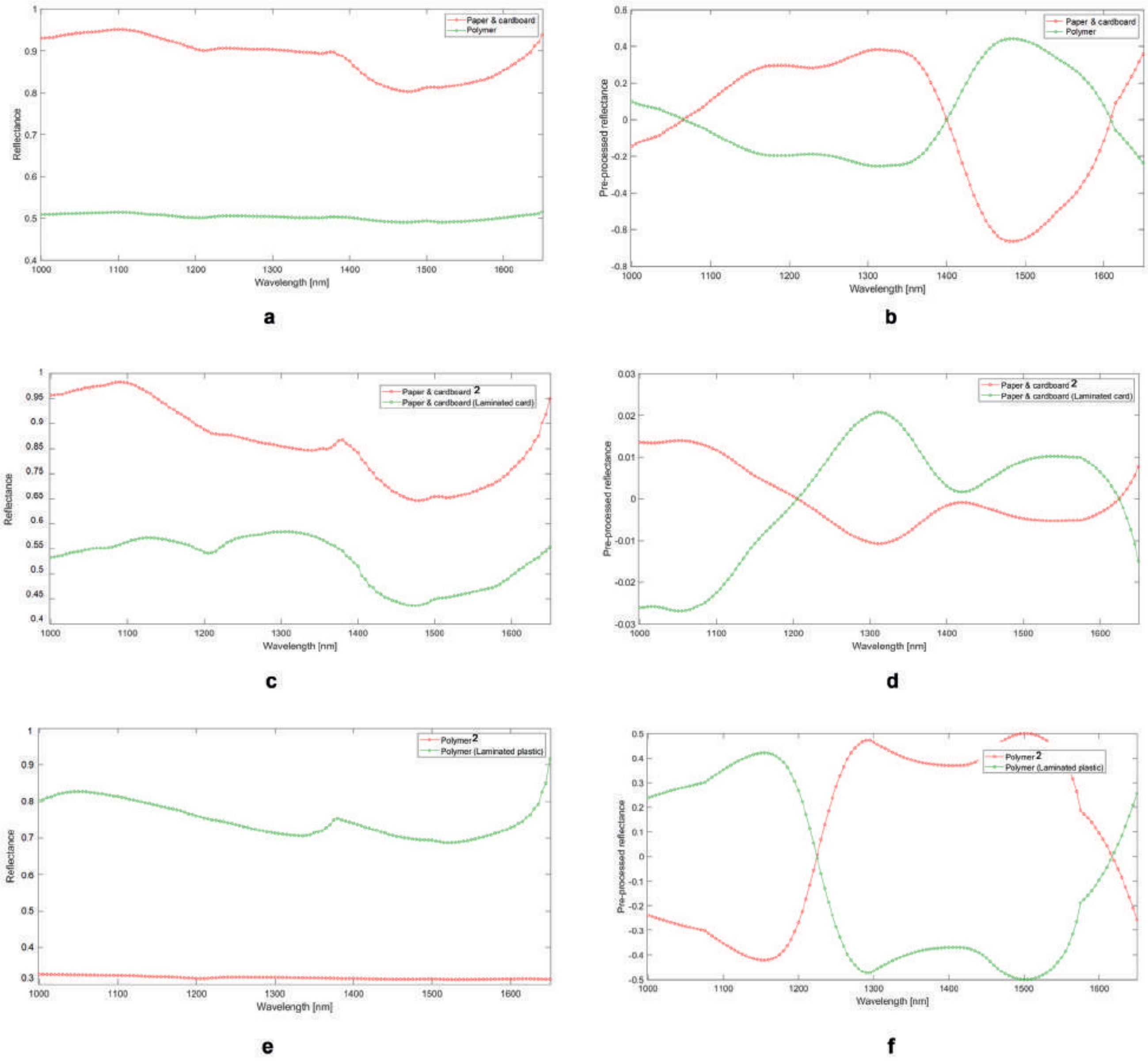


FIGURE 10: "Paper & cardboard" - "Polymer" calibration set: grand-average of raw reflectance spectra (a) and pre-processed spectra (b). "Paper & cardboard 2" and "Paper & cardboard (Laminated card)" calibration set: grand-average of raw reflectance spectra (c) and pre-processed spectra (d). "Polymer 2" and "Polymer (Laminated plastic)": grand-average of raw reflectance spectra (e) and pre-processed spectra (f).

that is classified as negative; *TN* (True Negative) is a negative instance that is a classified as negative and *FP* (False Positive) is a negative instance that is classified as positive.

While, in an object-based logic, considering 50% of the pixels in each object as discriminating threshold, the overall performance of the cascade model was computed in terms of Recognition (i.e. Accuracy), Error and Reliability rates, according to Eq. 9, Eq. 10 and Eq. 11 (Silva et Al. 2017).

$$Recognition = \frac{N_{Cor}}{N} \quad (9)$$

$$Error = \frac{N_{Mis}}{N} \quad (10)$$

$$Reliability = \frac{N_{Cor}}{N_{Mis}+N_{cor}} \quad (11)$$

being *N* the total number of samples, while N_{Cor} and N_{Mis} are the total of well-recognized and misrecognized samples, respectively.

3. RESULTS AND DISCUSSION

3.1 Cascade classification model

3.1.1 "Paper & cardboard" - "Polymer" model

"Paper & cardboard" - "Polymer" Classification model performance details for Calibration, Cross-validation and Validation phases are reported in Table 1. As can be seen in Figure 11, the validated model was able to recognize all the particles in "Validation set 1" and 13 out of 14 particles in "Validation set 2" (93% of the total in the set: a "Polymer" plastic (i.e. a laminated plastic fragment circled in blue) was misclassified for over 50% of pixels as "Paper & cardboard".

TABLE 1: "Paper & cardboard" - "Polymer" classification model details for Calibration, Cross-validation and Validation phases: Sensitivity, Specificity, Misclassification Error, Precision, accuracy and Class Error.

	Class	Sensitivity	Specificity	Misclassification Error	Precision	Accuracy	Class Error
Calibration	Paper & cardboard	1.000	0.970	0.021	0.938	0.980	0.015
	Polymer	0.970	1.000	0.021	1.000	0.980	0.015
Cross-validation	Paper & cardboard	1.000	0.970	0.021	0.938	0.980	0.015
	Polymer	0.970	1.000	0.021	1.000	0.980	0.015
Validation set 1	Paper & cardboard	0.952	0.993	0.020	0.985	0.980	0.028
	Polymer	0.993	0.952	0.020	0.978	0.980	0.028
Validation set 2	Paper & cardboard	0.958	0.896	0.083	0.829	0.917	0.073
	Polymer	0.896	0.958	0.083	0.976	0.917	0.073

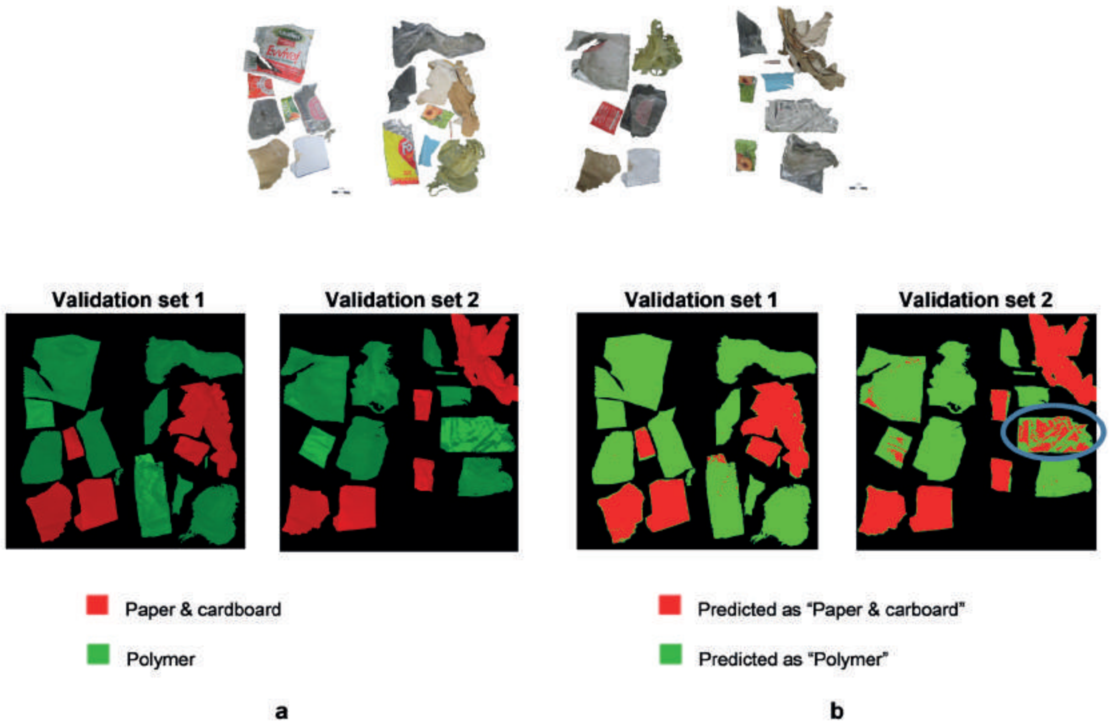


FIGURE 11: The validation set 1 and set 2 - "Paper & cardboard" and "Polymer" classification: actual classes (a) and classes predicted by the PLS-DA model. By comparing the actual classes to the prediction map, it can be seen that all the fragments were correctly classified by the two-class PLS-DA model in "Validation set 1". While, In "validation set 2", a comparison between the actual classes and the PLS-DA prediction map highlights the misclassification of a particle (circled in blue), a laminated plastic fragment, predicted for more than 50% of the pixels as "Paper & cardboard".

3.1.2 "Paper & cardboard 2" - "Paper & cardboard (Laminated card)" model

"Paper & cardboard 2" - "Paper & cardboard (Laminated card)" achieved good classification model performance as reported in Table 2. As can be seen in Figure 12, the validated model was able to recognize all the particles in "Validation set 1" and in "Validation set 2".

3.1.3 "Polymer 2" - "Polymer (Laminated plastic)" model

"Polymer 2" - "Polymer (Laminated plastic)" classification model performance details for calibration, cross-validation and validation phases are reported in Table 3. The validated model was able to recognize 10/11 particles in "Validation set 1" (91% of the total in the set, as can be seen in Figure 13): a "Polymer (Laminated plastic)" fragment (i.e. a laminated plastic fragment circled in blue, as seen in Figure 13), was misclassified for over 50% of pixels as "Polymer 2" class. However, the model can predict all particle class labels in "Validation set 2".

3.1.4 Overall performances of the cascade identification process

The "Paper & cardboard" - "Polymer" classification model can discriminate 29 out of 30 particles (97% total particles of the two validation sets), reaching a Recognition and a Reliability of 0.967. In the second step of the cascade classification, the "Paper & cardboard 2" - "Paper & cardboard (Laminated card)" model is able to discriminate all

TABLE 2: "Paper & cardboard 2" - "Paper & cardboard (Laminated card)" classification model details for Calibration, Cross-validation and Validation phases: Sensitivity, Specificity, Misclassification Error, Precision, accuracy and Class Error.

	Class	Sensitivity	Specificity	Misclassification Error	Precision	Accuracy	Class Error
Calibration	Paper & cardboard 2	0.882	0.937	0.099	0.965	0.901	0.090
	Paper & cardboard (Laminated card)	0.937	0.882	0.099	0.804	0.901	0.090
Cross-validation	Paper & cardboard 2	0.881	0.937	0.100	0.965	0.901	0.091
	Paper & cardboard (Laminated card)	0.937	0.881	0.100	0.804	0.901	0.091
Validation set 1	Paper & cardboard 2	0.983	0.887	0.031	0.982	0.969	0.065
	Paper & cardboard (Laminated card)	0.887	0.983	0.031	0.894	0.969	0.065
Validation set 2	Paper & cardboard 2	0.997	0.954	0.009	0.992	0.991	0.024
	Paper & cardboard (Laminated card)	0.954	0.997	0.009	0.983	0.991	0.024

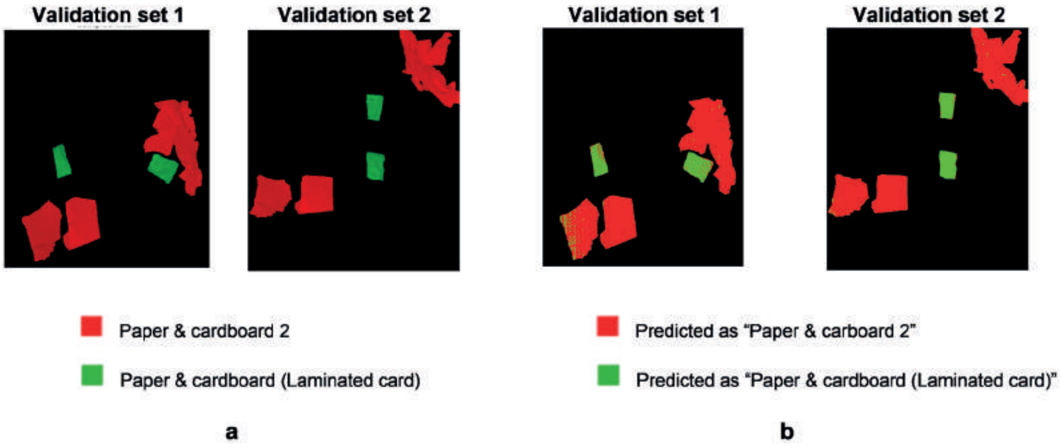


FIGURE 11: The validation set 1 and set 2 - "Paper & cardboard 2" - "Paper & cardboard (Laminated card)" classification: actual classes (a) and classes predicted by the PLS-DA model. Fragments were correctly classified by the two-class PLS-DA model as comparing the actual classes to the prediction map.

the particles included into the two validation sets (10 out of 10), thus reaching a Recognition and a Reliability of 1. While "Polymer 2" - "Polymer (Laminated plastic)" model is able to correctly classify 19 out of 20 of the particles included in the validation sets, reaching a Recognition and a Reliability of 0.950. The misclassified fragments are laminated plastics, this misclassification is due to their high reflection related to the exposed surfaces wrinkling of the bent packaging fragments and linked to scattering phenomena.

Following a pixel-based logic, the most reflective zones and shadow zones of the bent fragments are sometimes misclassified. To sum up, the cascade model, by having misclassified 2 out of 30 particles in total, reached in prediction a Recognition and a Reliability of 0.933, with an Error equal to 0.066.

4. CONCLUSIONS

A PLS-DA-based cascading classification model able to recognize polymeric fragments from cellulosic ones and to identify multi-layer materials (i.e. laminated plastic and laminated cardboard) was built and validated. In the first step of the cascade classification model, finalized to discriminate cellulose-based from polymer/plastic-based fragments 29 out of 30 objects were recognized. In the second step, the laminated card and other cellulose-based particles were all correctly recognized and only one fragment of laminated plastic was misclassified performing discrimination of laminated plastic fragments from other polymer/plastic-based fragments. The set up cascade model reached in prediction an overall Recognition and Reliability of 0.933.

A NIR-HSI-based approach can represent an optimal, reliable and low-cost answer to systematically identify impurities and composite materials inside plastic waste streams. The proposed approach, if fully implemented, can be utilized as a control strategy for continuous monitoring or as an analytical core for sorting materials in post-consumer plastic packaging waste characterized by the presence of paper, cardboard and multilayer packaging.

Results showed as the application of hyperspectral imaging, in the near-infrared range (1000–1700 nm), applied

TABLE 3: The "Polymer 2" - "Polymer (Laminated plastic)" classification model details for Calibration, Cross-validation and Validation phases: Sensitivity, Specificity, Misclassification Error, Precision, accuracy and Class Error.

	Class	Sensitivity	Specificity	Misclassification Error	Precision	Accuracy	Class Error
Calibration	Polymer 2	0.930	0.839	0.107	0.894	0.893	0.116
	Polymer (Laminated plastic)	0.839	0.930	0.107	0.891	0.893	0.116
Cross-validation	Polymer 2	0.930	0.839	0.107	0.894	0.893	0.116
	Polymer (Laminated plastic)	0.839	0.930	0.107	0.891	0.893	0.116
Validation set 1	Polymer 2	1.000	0.304	0.130	0.863	0.870	0.348
	Polymer (Laminated plastic)	0.304	1.000	0.130	0.998	0.870	0.348
Validation set 2	Polymer 2	0.990	0.621	0.093	0.900	0.907	0.194
	Polymer (Laminated plastic)	0.621	0.990	0.093	0.948	0.907	0.194

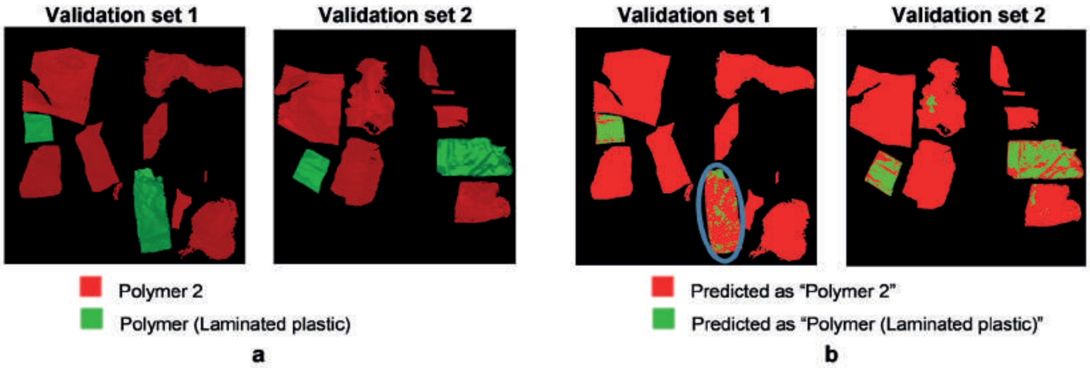


FIGURE 13: The validation set 1 and set 2 - "Polymer 2" - "Polymer (Laminated plastic)" classification: actual classes (a) and classes predicted by the PLS-DA model. In Validation set 1, a comparison between the actual classes and the PLS-DA prediction map highlights the misclassification of a particle (circled in blue), a laminated plastic fragment, predicted for more than 50% of the pixels as "Polymer 2". While in Validation set 2, all the fragments were correctly classified by the two-class PLS-DA model; however, some pixels are misclassified.

on a packaging waste stream, opens the door to future innovations in the field of resources and recycling, that is the possibility to develop sensing architectures to recognize plastic and/or 2D pollutants (i.e paper-based packaging and laminated card fragments), in order to be used not only as a sorting engine but also as an analytical core to perform quality control on products and/or byproducts coming from different manufacturing stages.

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APPENDIX

The detailed information regarding the sample sets selected to perform model calibration and validation are reported in Figure A1. Each fragment is labeled and their characteristics, for the different sets, are reported in Table A1.

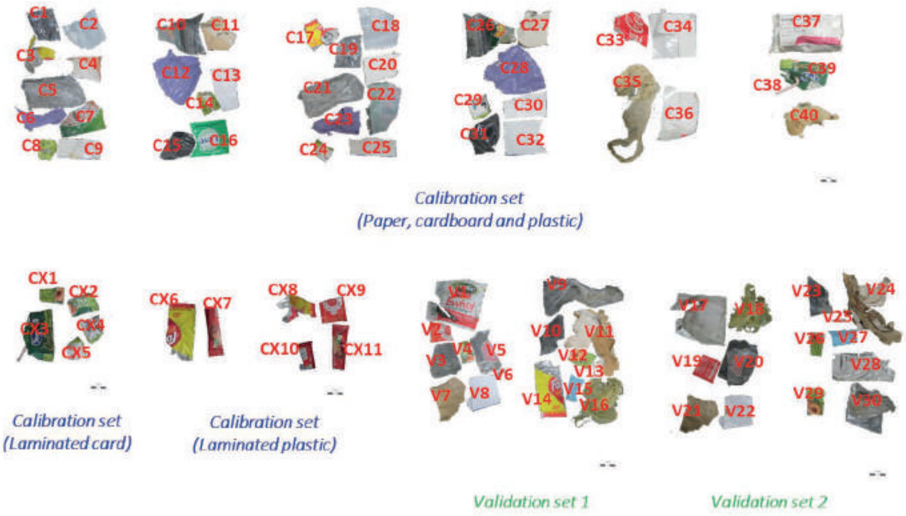

FIGURE A1: Reference image of the sample sets. For each fragment is reported the sample identification label.

TABLE A1: Characteristics of the sample sets used in this study.

Sample set	Sample ID	Fragment/sample definition	Sample class	Lamination
Calibration set (Paper, cardboard and plastic)	C1	Transparent plastic cup	Plastic	
	C2	White plastic cup	Plastic	
	C3	Laminated plastic potato chip bag	Plastic	Laminated plastic
	C4	Pizza cardboard carton	Paper and cardboard	
	C5	Transparent plastic film	Plastic	
	C6	Plastic bag	Plastic	
	C7	Plastic peanut bag	Plastic	
	C8	Tetra Brik juice carton	Paper and cardboard	Laminated cardboard
	C9	Laminated paper envelope	Paper and cardboard	Laminated card
	C10	Plastic peanut bag	Plastic	
	C11	Cardboard carton	Paper and cardboard	
	C12	Plastic bag	Plastic	
	C13	Paper	Paper and cardboard	
	C14	Tetra Brik juice carton	Paper and cardboard	Laminated cardboard
	C15	Transparent plastic bottle	Plastic	
	C16	Plastic bottle label	Plastic	
	C17	Laminated plastic potato chip bag	Plastic	Laminated plastic
	C18	White plastic cup	Plastic	
	C19	Transparent plastic cup	Plastic	
	C20	Cardboard carton	Paper and cardboard	
	C21	Transparent plastic film	Plastic	
	C22	Plastic peanut bag	Plastic	Laminated plastic
	C23	Plastic bag	Plastic	
	C24	Tetra Brik juice carton	Paper and cardboard	Laminated cardboard
	C25	Laminated paper envelope	Paper and cardboard	Laminated card
	C26	Plastic peanuts bag	Plastic	
	C27	Cardboard carton	Paper and cardboard	
	C28	Plastic bag	Plastic	
	C29	Tetra Brik juice carton	Paper and cardboard	
	C30	Paper	Paper and cardboard	
	C31	Transparent plastic bottle	Plastic	
	C32	Plastic bottle label	Plastic	
	C33	Plastic bottle label	Plastic	
	C34	Laminated paper - plastic envelope	Paper and cardboard	Laminated card
	C35	Plastic bag	Plastic	
	C36	Plastic bottle label	Plastic	
	C37	Paper box	Paper and cardboard	
	C38	Tetra Brik juice carton	Paper and cardboard	Laminated cardboard
	C39	Plastic straw	Plastic	
	C40	Cardboard carton	Paper and cardboard	
Calibration set (Laminated card)	CX1	Tetra Brik juice carton	Paper and cardboard	Laminated cardboard
	CX2	Tetra Brik juice carton	Paper and cardboard	Laminated cardboard
	CX3	Tetra Brik juice carton	Paper and cardboard	Laminated cardboard
	CX4	Tetra Brik juice carton	Paper and cardboard	Laminated cardboard
	CX5	Tetra Brik juice carton	Paper and cardboard	Laminated cardboard

Calibration set (Laminated plastic)	CX6	Laminated plastic potato chip bag	Plastic	Laminated plastic
	CX7	Laminated plastic snack bag	Plastic	Laminated plastic
	CX8	Laminated plastic potato chip bag	Plastic	Laminated plastic
	CX9	Laminated plastic ketchup sachet	Plastic	Laminated plastic
	CX10	Laminated plastic snack bag	Plastic	Laminated plastic
	CX11	Laminated plastic snack bag	Plastic	Laminated plastic
Validation set 1	V1	Sausage plastic bag	Plastic	
	V2	Laminated plastic ketchup sachet	Plastic	Laminated plastic
	V3	Transparent plastic film	Plastic	
	V4	Tetra Brik juice carton	Paper and cardboard	Laminated cardboard
	V5	Plastic bottle label	Plastic	
	V6	Plastic bag	Plastic	
	V7	Cardboard carton	Paper and cardboard	
	V8	Paper	Paper and cardboard	
	V9	Transparent plastic film	Plastic	
	V10	Transparent plastic bag	Plastic	
	V11	Cardboard carton	Paper and cardboard	
	V12	Tetra Brik juice carton	Paper and cardboard	Laminated cardboard
	V13	Plastic straw	Plastic	
	V14	Laminated plastic potato chip bag	Plastic	Laminated plastic
	V15	Plastic plate	Plastic	
	V16	Plastic bag	Plastic	
Validation set 2	V17	Sausage plastic bag	Plastic	
	V18	Plastic bag	Plastic	
	V19	Laminated plastic ketchup sachet	Plastic	Laminated plastic
	V20	Plastic bottle label	Plastic	
	V21	Cardboard carton	Paper and cardboard	
	V22	Paper	Paper and cardboard	
	V23	Transparent plastic bag	Plastic	
	V24	Cardboard carton	Paper and cardboard	
	V25	Plastic straw	Plastic	
	V26	Tetra Brik juice carton	Paper and cardboard	Laminated cardboard
	V27	Plastic plate	Plastic	
	V28	Laminated plastic potato chip bag	Plastic	Laminated plastic
	V29	Tetra Brik juice carton	Paper and cardboard	Laminated cardboard
	V30	Transparent plastic film	Plastic	

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PYROLYSIS OF SOLID WASTE AND ITS COMPONENTS IN A LAB SCALE INDUCTION-HEATING REACTOR

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ABSTRACT

The present study investigates the thermochemical conversion of Solid Recovered Fuel (SRF), represented by selected “model materials”. A laboratory-scale induction heated device was specifically developed to achieve fast pyrolysis conditions close to those encountered in a fluidized bed reactor. The novel device can handle up to 5 grams of solid, allowing fast heating rates (near 70°C/s) and a homogeneous distribution of temperature all along the reactor. Pyrolysis tests of a SRF sample and four model materials (Polyethylene, Polyethylene Terephthalate, beech wood, cardboard) were performed at 800°C. The yield and composition of the produced gas for each sample were determined. Experimental results will help to elucidate the relation between the initial components of waste derived fuels and the obtained reaction products.

1. INTRODUCTION

About 2.01 billion metric tons of municipal solid waste (MSW) are produced annually worldwide. By 2050, this quantity is expected to increase by up to 70% (Kaza et al., 2018). As of today, 66% of the produced waste is still dumped or landfilled, causing environmental and health risks. Current management policies try to reduce this share by focusing on prevention, reduction, and mechanical recycling. Other solid waste fractions that cannot be reused or recycled are increasingly used to produce waste derived fuels, offering an interesting alternative to conventional disposal methods. Refuse Derived Fuel (RDF) and Solid Recovered Fuel (SRF) are both produced from industrial and household wastes, the latter following specific criteria indicated in CEN/TS 15359 (Solid Recovered Fuels, 2006). Both fuels include combustible materials with high calorific values like plastics, textiles, wood, and elastomers (Garcés et al., 2016).

Among the available waste to energy technologies, gasification has received an increasing interest in the last years (Saghir et al., 2018). Gasification is a thermochemical process where carbonaceous feedstocks react above 700°C, in presence of an oxidizing agent that is fed below the stoichiometric amount needed for total oxidation. The produced syngas can be directly burnt to produce energy and heat or, after cleaning, used in a synthesis process to produce biofuels or chemicals. This process offers some environmental advantages compared to landfilling or incineration. However, upscaling still presents some technical barriers (Centi & Perathoner, 2020).

Pyrolysis is of great interest since it is considered here as the first stage of the gasification process. During this step, the feedstock decomposes in absence of oxygen, producing volatiles in the form of light permanent gases (such as H_2, CO, CO_2, CH_4, and H_2O), and condensable hydrocarbons (tar). The remaining solid residue is known as char. Those devolatilization products participate with the oxidizing agent in the gasification step, where cracking and reforming reactions produce syngas. Distribution and product yields are highly influenced by temperature, heating rate and residence time, features that are directly related to reactor configuration (Pasel & Wanzl, 2003). Slow heating rates induce high residence times and favor char formation, while high heating rates and high temperatures favor gas products (Efika et al., 2018).

Even in the same process conditions, final products are also affected by the raw composition of the feedstock (Esmaeili et al., 2020). In previous pyrolysis/gasification studies, researchers have attempted to evaluate the influence of the feedstock using real samples of waste fuels (Efika et al., 2015; Hwang et al., 2014). However, the heterogeneous composition of waste at many levels (source, season, components, etc.), as well as the different operations to which it is subjected during the production of derived fuels, makes it really challenging to apply the results to other fuels that

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may contain different feed compositions.

In this regard, several authors have conducted studies of the individual typical components found in waste (Chhabra et al., 2020; Meng et al., 2015; Zhou, Long, et al., 2015), most of them by thermogravimetric analysis (TGA). Mass loss curves and kinetic parameters are well known, yet few studies report the performance and composition of pyrolytic products. Moreover, the reaction conditions found in TGA and other laboratory-scale devices used in these works are not representative of full-scale gasifiers, where high heating rates and good gas-solid contact are achieved (Cortazar et al., 2020). Induction heating is an interesting alternative without the shortcomings of other conventional methods. Shorter heating times, better control, and better temperature distribution could be achieved in comparison with electrical resistance based furnaces as seen in the work of (Mishra et al., 2019). Nevertheless, the literature regarding pyrolysis or gasification applications at laboratory scale using this method is very scarce.

This work focuses on the distribution and characterization of products of pyrolysis, representing the first decomposition step of gasification of SRF in a lab scale reactor, especially developed to emulate fast pyrolysis conditions. The conversion of "model materials", selected to represent the main fractions found in solid waste, is of particular interest. Results will contribute to the development and validation of accurate prediction models, providing a better insight on the influence of feedstock on the reaction products.

1.1 Abbreviations

- MSW: Municipal Solid Waste
- RDF: Refuse Derived Fuel
- SRF: Solid Recovered Fuel
- TGA: Thermogravimetric Analysis
- LDPE: Low-Density Polyethylene
- PET: Polyethylene Terephthalate
- FID: Flame Ionization Detector
- NDIR: Non-Dispersive Infrared Detector

2. MATERIALS AND METHODS

2.1 Feedstock

A solid recovered fuel (SRF) sample produced from industrial and household waste was used for the tests. Typical discarded products from daily life were also collected and used as model materials.

Plastic samples consisted in low-density polyethylene (LDPE, referred here as PE), while polyethylene terephthalate (PET) was obtained from plastic water bottles. Wood sample consisted in pellets made from beech sawdust, and cardboard sample was obtained from packaging boxes. All materials were shredded up to particles between 1 and 3 mm, and then characterized by ultimate and proximate analyses. Moisture content was determined at 105°C while ash content was measured at 815°C according to the ISO 1171 standard. Elemental composition (C, H, N, S) was determined with an ELEMENTAR analyzer. Composition and characteristics for each sample are listed in Table 1.

2.2 Experimental setup and procedure

A laboratory scale unit was specifically designed to conduct pyrolysis and gasification tests under some reaction conditions that are representative of fast pyrolysis in a fluidized bed reactor. These characteristics include high heating rates, a temperature range between 700 and 900°C, and good gas/solid contact. Gas residence times inside the hot zone of the reactor are usually between 0.5 and 10 s for bubbling fluidized bed reactors, and between 0.5 and 1 s for circulating fluidized bed reactors (Marshall et al., 2014). For the present device, a residence time between 1 and 5 s was targeted, long enough to enable significant volatile secondary reactions. The experimental setup is shown in Figure 1.

A stainless-steel tube (560 mm in height, 30.15 mm in internal diameter) was externally heated by induction, with a water-cooled copper coil inductor (420 mm long) connected to a 12kW electrical generator (HFP 12, EFD induction Gmbh). Setpoint temperature was adjusted with a PID

TABLE 1: Characteristics of SRF and model materials.

[wt%]	SRF	PE	PET	Wood	Cardboard
Moisture (ar.)	5.17%	0.29%	0.5%	7.30%	6.69%
Ash (db.)	16.38%	0%	0%	2.53%	8.80%
C (db.)	48.00%	85.50%	63.00%	46.80%	43.60%
H (db.)	6.00%	13.90%	5.50%	6.70%	6.00%
O (db by diff.)	26.68%	0.50%	31.50%	43.77%	41.10%
N (db.)	1.33%	0.02%	0.02%	0.02%	0.03%
S (db.)	0.47%	0.03%	0.02%	0.1%	0.13%
Cl (db.)	1.14%	-	-	-	-

ar: as received, db: dry basis

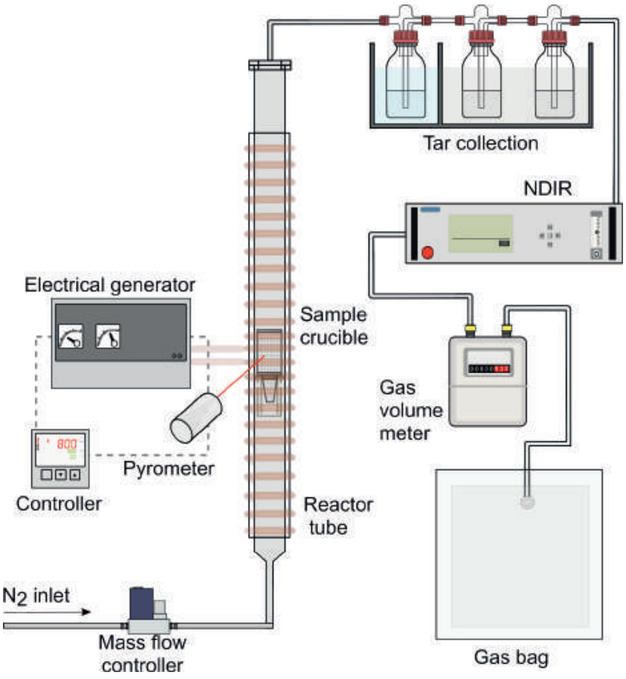

FIGURE 1: Schematic diagram of the lab scale device.

controller (GEFRAN 2500), connected to a two-color optical pyrometer (Impac IGAR 6). Its spot is pointing on the tube surface, at the height of the sample crucible.

The crucible was filled with 2 to 3g of dried feedstock (24h at 105°C) and placed in the isothermal zone of the reactor. N_2 (0.5 L/min) was used to flush the entire system and to carry the produced volatiles. The reactor was heated up to 800°C, with a heating rate of about 70°C/s, and then held at this temperature for about 20 minutes. Previous tests showed that the temperature was homogeneous in the sample, with a maximum temperature difference of 10°C.

Produced gases flowed upwards in the reactor top section and the outlet line, both traced and insulated to keep temperature above 250°C, and thus to prevent tar and water condensation before reaching the tar traps. Five gas washing bottles filled with 2-propanol were used to collect gas condensable species (water and tar) present in the stream. Glass wool and glass beads were used to improve the contact area between the gas and the solvent. The first two bottles were installed in an ice bath at 0°C, while the other three were immersed in a carbonic ice and 2-propanol bath at -70°C. Collected condensable hydrocarbons were sampled and subsequently analyzed by using a gas chromatograph system. Species were identified by the means of mass spectrometry coupled with a flame ionization detector for the quantification (GC-FID, Agilent 7890A).

Non-condensable gases were analyzed online using a Non-Dispersive Infrared Detector (NDIR Model 30, CAI), able to record CO, CO_2 and CH_4 concentrations every 1 s. The volume of the outlet gas was measured by a gas meter placed at the outlet of the NDIR analyzer. Permanent gases were then collected in a Tedlar bag to be further analyzed by micro gas chromatography (Agilent 3000A). Remaining solid products in the sample crucible were weighed and collected once the reactor had cooled down to ambient temperature.

3. RESULTS AND DISCUSSION

3.1 Carbon distribution

The carbon distribution among the pyrolysis products, namely gas, tar and char, is shown in Figure 2. These yields are calculated as the ratio of carbon in each product per mass of initial carbon in the dry ash free feedstock. Considered gas species consisted in H_2, CO, CO_2, CH_4, C2 hydrocarbons (C_2H_4, C_2H_6, C_2H_2), C3 hydrocarbons (C_3H_6, C_3H_8), benzene (C_6H_6) and toluene (C_7H_8). The tar fraction is defined here as all condensable organic compounds with a higher molecular weight than toluene.

For every feedstock, carbon was mostly converted into gas, as the high temperature and heating rate enhanced the thermal degradation of primary volatiles (E. C. Efika et al., 2015). The carbon fraction in solid was under 2 wt% for PE,and corresponds to carbon deposits in the walls of the sample crucible. Cellulosic materials (wood and cardboard) showed high C content in the solid fraction. Char yield was greater in the former case, since wood has a higher lignin content than cardboard, which is composed mostly of cellulose. Lignin presents a more complex molecular structure and high fixed carbon content, which makes it more thermally stable (Muley et al., 2016). The highest fractions of carbon in tar were found for SRF (12 wt%), followed by PE (10.7 wt%) and PET (7.4 wt%). Plastic content in waste fuels improves gas yield, but also increases tar content in syngas as observed here and also by other authors (Wilk & Hofbauer, 2013; Zaccariello & Mastellone, 2015). C-balance closure ranged between 75 and 90%. Deviations could be attributed to the volatilization of light compounds from the tar traps, and undetected species that were not quantified in the FID analysis.

3.2 Solid residue (char)

The chemical composition of the solid residue obtained after pyrolysis tests is listed in Table 2. The oxygen

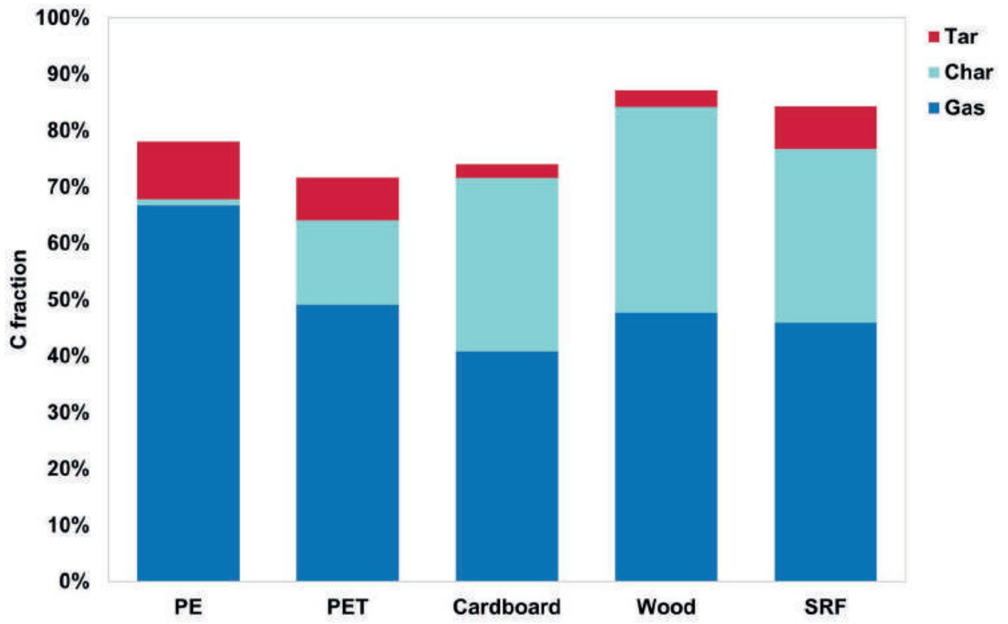

FIGURE 2: Carbon distribution from SRF and its components to pyrolysis products at 800°C.

content was calculated by difference, and the ash content was determined also at 815°C. No char was observed in the case of PE. The solid residue from SRF and cardboard pyrolysis showed the lowest amount of carbon, and high ash contents, which are related to the inorganics and inert materials from the original feedstock.

3.3 Gas composition

Yield and volumetric composition of the produced gases from SRF and model materials pyrolysis, are displayed in Table 3. Low density polyethylene showed the highest yield of gas. Its long-branched structure, follows a random chain scission mechanism during its devolatilization, giving high yields of hydrogen, light olefins (mostly C2 hydrocarbons) and no char as stated by other authors (Al-Salem et al., 2017; Block et al., 2019). Gas yields values were similar for SRF and the other studied materials.

CO and CO_2 were the main produced gas species in PET pyrolysis due to presence of oxygen groups from its monomer. Hydrocarbon production was much lower compared to PE, as observed also in the work of (Honus et al., 2018). Biogenic feedstock (wood, cardboard), which have an important O content, yielded the highest concentrations of oxygenated gases. CO and CO_2 contents were higher for cardboard case, while methane content was higher for wood. Raw materials used in the production of paper and cardboard are submitted to delignification processes, so their cellulose content can go up to 99% (Runchal et al., 2018). (Yang et al., 2007), suggested that cracking of carbonyl functional groups of cellulose gave high CO yields, while the degradation of aromatic rings and methoxyl groups in lignin enhanced CH_4 yields.

(Win et al., 2020) conducted flash pyrolysis experiments of wood pellets and polyethylene in an electric furnace at 900°C. In their results, yield of H_2 was higher for wood pellet (25% vol) than that for polyethylene (20% vol), which differs from the findings presented here. They attributed those results to water-gas reactions, enhanced from the moisture content of wood pellets. In our study, samples were dried prior to the experiments, and pyrolysis temperature was lower, hence these reactions were not favored. Yields of methane and ethylene were close to our results for both materials.

Regarding the composition of the gas produced in the pyrolysis of the SRF, CO was the major gas component (30%), followed by CH_4 (23%) and H_2 (22%). Gas distribution after SRF pyrolysis was intermediate between different materials, being plastics responsible for the high yields of CH_4 and C_2 hydrocarbons while biogenic materials for the majority of oxygenated compounds. In general, the results found in this study followed the same trends observed in RDF pyrolysis tests conducted by other researchers at 800°C (Blanco et al., 2012; Daouk et al., 2018). It is known that the sample used here presented small fractions of other fossil derived materials such as PS and rubber, known for generate high amounts of methane during their pyrolysis (Zaini et al., 2019).

TABLE 2: Chemical composition of the remaining solid residue after pyrolysis at 800°C.

wt %	%C	%H	%N	%S	%O	%Ash
SRF	35.97	1.32	0.96	1.05	11.60	49.1
PET	85.17	1.49	0.22	0.14	12.98	0.98
Wood	77.8	1.01	0.64	0.12	12.56	7.87
Cardboard	45.13	0.73	0.37	0.14	14.3	39.33
PE	No char					

3.4 Tar Composition and content

The condensed tars were sampled and then analyzed by GC – FID. Quantified tar components were categorizedaccording to the classification system proposed by the Energy research Center of the Netherlands (ECN) (Devi et al., 2005). This classification is based on the solubility and condensation properties of the different tar compounds, which define the downstream treatment conditions. Tars are grouped in five classes (Table 4) depending on the number of aromatic rings.

As seen in Figure 3, tar content was much higher for plastics compared to lignocellulosic materials. When comparing the relative distribution of tar products by group, wood ,cardboard and PET showed high amounts of hetorocyclics (class 2) , due to the presence of oxygenated compounds like phenol. Class 4 species content was higher for wood than for cardboard thanks to the presence of lignin, which is know to yield higher amounts of naphthalene (Zhou, Wu, et al., 2015). The primary products of PE pyrolysis consist mostly in alkenes, which react via Diels-Alder reactions producing single ring tar compounds like styrene and indene, and some 2- ring tars like naphthalene. PET monomer contains aromatic rings in its structure, which influence the formation of small amounts of heavy polyaromatic species (3 or more rings). SRF showed the highest tar yield, with high contents of heavy aromatics. This may be attributed to the presence of monomers with aromatic rings as evidenced with PET, but also others such as polystyrene.

4. CONCLUSIONS

Pyrolysis tests of SRF and selected modeled materials were carried out in a specifically developed induction heated reactor at 800°C. The most common polymers in waste (PE and PET) were the model materials for the plastic fraction, while beech wood and cardboard were

TABLE 3: Gas composition from pyrolysis at 800°C of SRF and model materials.

% vol	SRF	PE	PET	Wood	Cardboard
H2	22.1%	29.5%	24.8%	19.8%	20.5%
CO	30.1%	0.6%	31.6%	44.2%	48.7%
CO2	9.8%	0.4%	22.9%	12.0%	12.7%
CH4	22.4%	25.6%	8.6%	18.2%	13.2%
C2	9.5%	33.0%	1.8%	4.5%	4.2%
C3	0.1%	1.4%	0.0%	0.0%	0.1%
BTX	5.1%	4.0%	10.3%	0.9%	0.3%
Gas yield[L/g daf]	0.4215	0.7815	0.4463	0.4439	0.4203

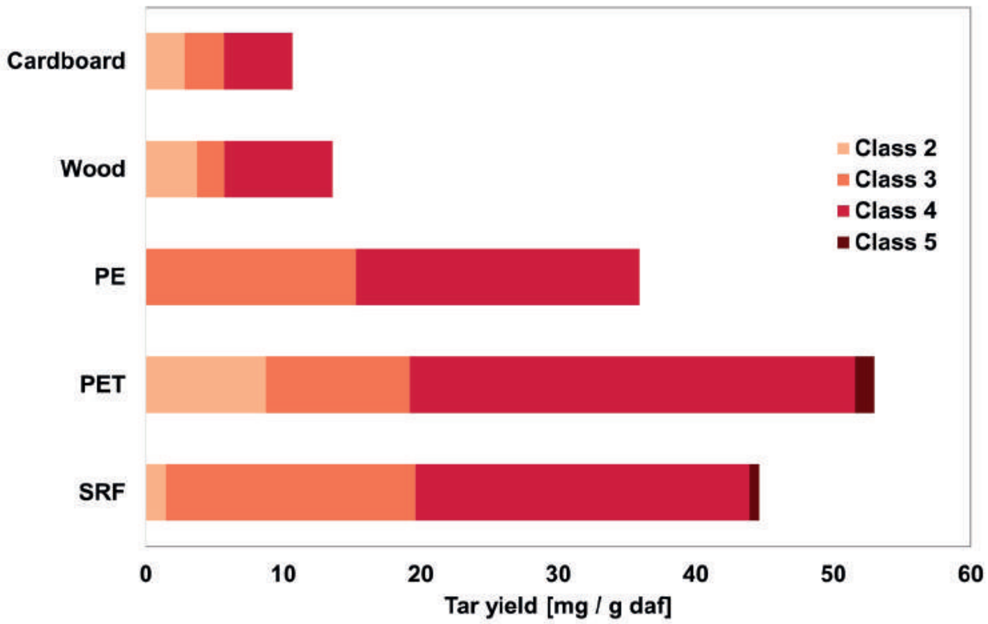

FIGURE 3: Composition of produced tar from the pyrolysis of SRF and model materials at 800°C.

representative of lignocellulosic biomass. The reaction products were quantified and analyzed, and the composition of the produced gas was reported for each material. For all cases, carbon conversion to gas was higher than 40%, as the high heating rates favored secondary reactions like the cracking of volatile products to lighter stable gases. Even if the elemental composition of some materials was very similar, differences were found in the gaseous and condensable products, showing that macromolecular structure can influence the final product distribution. These results will be helpful for the development of predictive models.

However, to have a better insight of the thermochemical conversion of solid waste and its derived fuels, additional studies must be conducted. Co-pyrolysis of waste is of particular interest, as synergetic effects may appear between the different materials modifying the final distribution of the products. Additional tests in partial oxidation conditions are also important to explore the impact of the gas phase reactions in the syngas and tar composition.

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TABLE 4: Tar classification according to ECN.

Tar class	Class Name	Properties	Compounds considered in this study
1	GC-undetectable	Very heavy tars, cannot be detected by GC	
2	Heretocyclic	Tars containing heteroatoms; highly water soluble compounds	Phenol, Benzofurane
3	Light aromatics	Usually light hydrocarbons with one ring; do not pose a problem regarding condensability and solubility	Ethylbenzene, Styrene a-Methylstyrene,
4	Light polyaromatics	Two and three ring compounds; condense at low temperature even at very low concentration	Indene, Naphthalene, methylnaphthalene, Biphenyl, Acénaphtylene,Acénaphtene, Dibenzofurane, Fluorene
5	Heavy polyaromatics	Larger than three-rings; these components condense at high temperatures at low concentrations	Fluoranthene, pyrene

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LANDFILLS VOLUME INCREASE WITH REINFORCED SOIL EMBANKMENTS: BASIC THEORY AND CASE STUDIES

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ABSTRACT

The construction of controlled landfills, either for municipal or industrial wastes, is currently acknowledged worldwide as a "social need". Indeed, today one of the main societal issues is the identification of an optimal means of disposing of the huge quantities of urban and industrial waste produced on a daily basis by individuals and factories. The task of environmental engineers is to design landfill systems capable to prevent any pollution to the water, the air, and the surrounding fauna and human life. In densely populated countries, the first problem to be solved is the location of the landfill. This social confrontation often leads to locate the landfill not in most geologically and geotechnically suited site, but in marginal areas which finally satisfy all the fighting communities. More and more geosynthetics are used to solve the problems associated with landfills located in marginal areas. The use of geogrids to construct steep reinforced embankments with the aim to increase the volume of wastes that can be disposed and at the same time to increase the stability of the wastes themselves, is getting more and more diffused. The paper describes the use of geosynthetics reinforced soil structures to increase the landfill volume with reinforced soil embankments. Some examples of structures already constructed, either for industrial or municipal wastes landfills, are described.

1. INTRODUCTION

The construction of controlled landfills, either for municipal wastes or for industrial wastes, is aknowoldeged as a "social need". Today, one of the main social issues problems is the identification of the optimal way of disposing of the huge quantity of urban and industrial waste produced daily by individuals and factories. Possible solutions vary among incineration, chemical and/or biological and/or mechanical treatments, burial in soil, or other special technologies. Whichever is the selected solution, at the end of all treatments there is always a fraction of the original waste that remains in the status of marginal or exhausted material.

The final destination of this material is the disposal in a technologically managed landfill. The task of environmental engineers is to design sustainable landfill systems able to prevent any pollution to the water, the air, and the surrounding fauna end human life, taking also into account of an adequate design of the capping (i.a. Cazzuffi & Recalcati, 2018, Grossule & Stegmann, 2020).

In densely populated countries, the first problem to be solved is the location of the landfill, since the NIMBY syndrome (Not In My Back Yard) is often the reason for harsh discussions between adjacent communities.

This social confrontation often leads to locate the landfill in a site that is not he most geologically and geotechnically suited; marginal areas which finally satisfy all the fighting communities are chosen. Environmental engineers are therefore challenged by new problems, which often require an interdisciplinary approach, where geologist, geotechnical engineers, chemists, landscape architects and other experts are involved.

Volume increase of the sites chosen for the landfill or in already existing landfill represents a good solution, provided that this choice is done without bringing any hazard to the stability of the amount of wastes. More and more geosynthetics are used to solve the problems associated with landfills located in marginal areas. The use of geogrids reinforced steep embankments to increase the volume of wastes and at the same time to increase the stability of the wastes is diffused. The first examples were mainly consisting in a single, high embankment containing the wastes; recently construction of superimposed embankments is becoming common, as this technique presents advantages from a logistic point of view, as shown later on.

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2. GEOGRID REINFORCED SLOPES AND EMBANKMENTS

2.1 Basic theory of reinforced soil

The reinforced soil technique is a construction method developed for thousand years, and has experienced different types of reinforcing materials, from bamboo to steel strips, from tree branches to geogrids. The latest types of reinforcement have the advantage of providing greater durability, strength, proven experience and finally a more theoretical design approach.

The first reinforcement application of geogrids in Europe dates about 1981, while the first use of geogrids in Italy dates back to 1986, when a reinforced embankment was built to contain wastes in a landfill in Modena (Northern Italy), as described by Cazzuffi et al. (1993).

Reinforced soil is a composite material which combines the typical resistance of two different materials in such a way to minimize the weakness of each one. Particularly, a relatively large quantity of the cheapest and compression resistant material, the soil, is improved in its engineering characteristics by the combination with a relatively small quantity of a more expensive and high tensile resistant material, the geogrids. A synergy between the tensile and compressive resistance of the two materials develops, improving the global characteristics of the composite material, as it happens when concrete is associate with steel in reinforced concrete. When geogrids are placed in the soil and the latter is compacted, the soil particles interlock with the grid apertures, which produce a very efficient confining action, thus limiting the relative movements and increasing the shear resistance of the soil-geogrid composite.

The main advantages of a reinforced soil structure are the following:

- lower global cost: steeper slopes reduce the quantity of fill material; moreover, less valuable and cheaper materials can be used;
- improved stability: the reinforcement guarantees an improvement in the factor of safety, even in seismic areas;
- possibility to build directly on low-bearing-capacity soils without a preliminary consolidation and great caution during construction.

2.2 Steep Slopes Analysis: Internal, External and Global Stability

For steep reinforced slopes, there are three failure modes (Figure 1):

- Internal, where the failure plane passes through the reinforcing elements
- External, (failure surface passes behind and underneath the reinforced mass)
- Compound-Global (failure surface passes behind and/or through the reinforced soil mass)

Design is based on modified versions of the classical limit equilibrium slope stability methods.

2.3 Use of geogrids reinforced steep slopes in landfills

The technology of geogrid reinforced slopes finds a natural and diffused application in the construction or in the enlargement of controlled landfills, as emphasized also by Cancelli & Cazzuffi (1994). Among several cases of embankment built to retain wastes in a landfill, it is worth mentioning a couple of projects.

The first known case of embankment of waste containment is the one already mentioned and dated 1986 (Modena landfill, Cazzuffi et al., 1993). It consisted in an embankment, 5.00 m high, with face inclined at 45° on the external side of the landfill and at 55° in the internal side; the embankment was constructed over the waste, and stability analysis was taking into account also settlements beneath the embankment.

More recently (Cowland, 2007) a 35m-high and 340 m long embankment bridging two hilltops was constructed to retain wastes of the North East New Territories (NENT) Landfill in Hong Kong. Both the faces were inclined at 68° on the horizontal; the external side was divided in steps 7.50 m high separated by berms 2.00 m wide, and was constructed with a fire resistance facing; the internal face was divided in steps 12.00 m high, with the same type of berms, and was covered with the lining system. Stability analysis on both sides of the reinforced embankment before filling were studied, as well as overall stability with waste deposit against the internal face.

As shown in the cases here described, the presence of the geogrid allows to construct embankments and em-

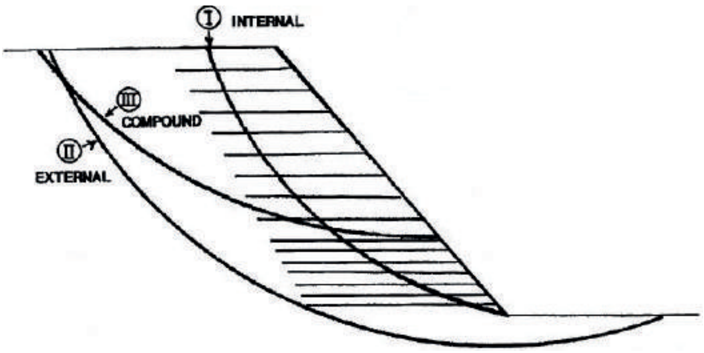


FIGURE 1: Failure modes for reinforced soil slopes.

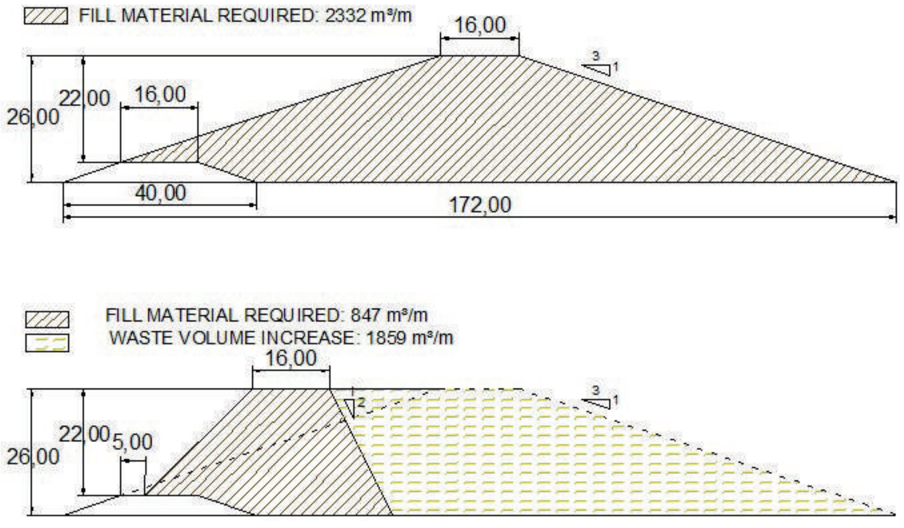


FIGURE 2: Comparison between a traditional shallow embankment and a geogrid reinforced steep slopes embankment.

bankments with an angle, both in the internal and the external face, much steeper respect to the unreinforced case, allowing for a reduction in the volume (and the cost) of fill material necessary and in the meantime an increase of the available volume. An example where a traditional embankment with very shallow slopes is compared to a geogrid reinforced embankment, is shown in Figure 2.

3. EXAMPLE OF GEOGRID REINFORCED EXAMPLES TO INCREASE THE AVAILABLE VOLUME

3.1 Teglio Landfill, Valtellina, Italy

The landfill was constructed in the late ’90 in Valtellina Valley (Northern Italy), as described by Montanelli et al., 1997. This valley is characterised by the presence of an important alluvial deposit, with different characteristics below the river (where it is mainly gravel) or at the side of the river (where the alluvial deposit are made of gravel, sand, silty sand with intercalation of silty peat); close to the side of the valley, there is a transition area with slope debris and alluvial fans made of gravel or sandy gravel alluvial deposits. Due to lack of better available areas, the landfill had to be constructed over an area where all the previously described situations were encountered.

Seven continuous dynamic penetration tests with 51 mm cone (DPT) and two static penetration tests (CPT) were performed; grain size distribution, unit weight and Atterbergh limits were measured. The subsoil was found to be mainly sand; peat was limited to thin intercalation layers, while there were almost no clay layers. The soil, according to U.S.C.S., could be defined mainly as SM (silty sand).

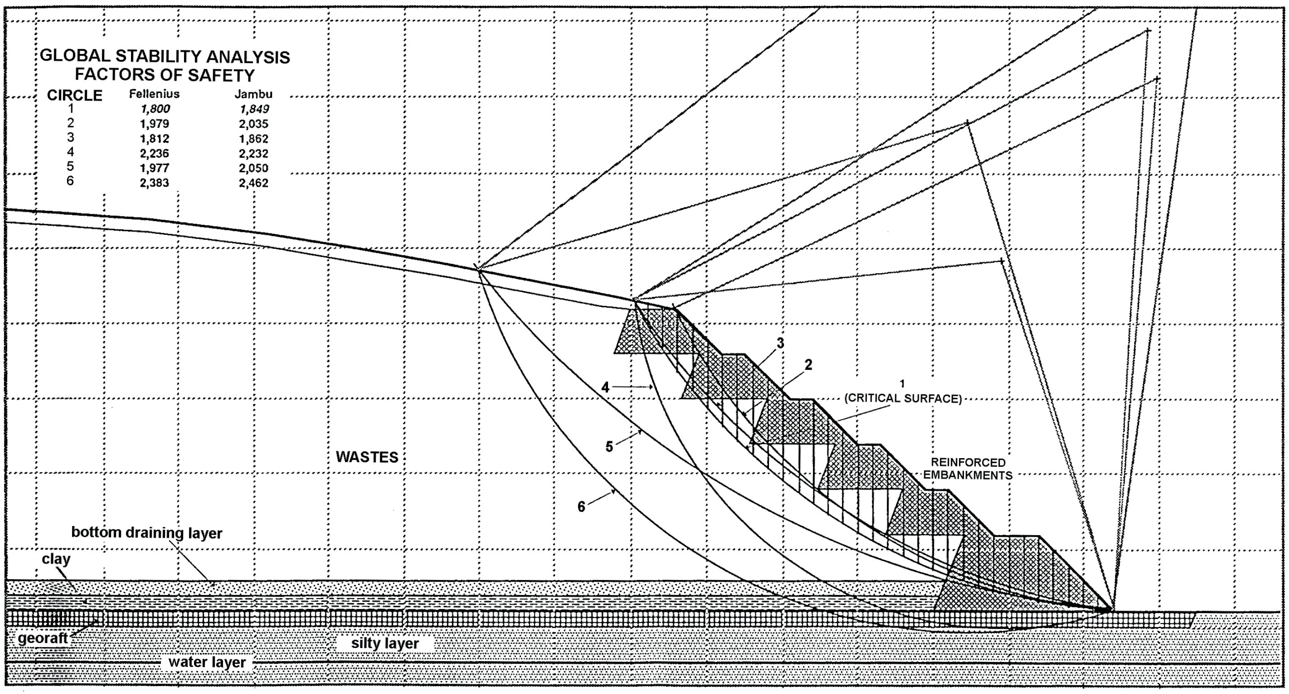
The relative density, determined after the penetration tests (with all the required correction factors), was found to be around 70-80% for the top 9.00 m, and 80-85% at a greater depth. Shear parameters were obtained from tests: a friction angle of 34° was found in the upper 9 m, while higher values (36°) were obtained at a greater depth.

Global stability analysis was performed on the whole landfill body. Both Fellenius and Janbu method were used; the check was extended to circular surfaces passing through the toe of the first block and interesting the whole landfill, but also to circles passing through horizontal surfaces beneath the first embankment, and finally to circular surfaces completely within the reinforced blocks. Results of global stability analysis are shown in Figure 3.

The front embankments necessary to provide the confinement to the wastes were built using monoriented HDPE geogrids. Movable formworks were used for the construction, in order to avoid any possible damage to the HDPE geomembrane used for waterproofing the side slopes. The formworks were made of scaffolding tubes with timber boards: the formworks were placed near the edge of the slope, then a mono-oriented geogrid was placed; a bi-oriented geogrid was placed at the edge of the slope, leaving an edge outside the movable formwork. After laying down and compacting the fill soil, the bi-oriented geogrid was wrapped around the face and then the formwork was extracted (with a back-hoe) and used for the following reinforcement layer.The layout and some details of this application are described in Figure 4, Figure 5 and Figure 6.

3.2 Genna Luas, Sardinia, Italy

Genna Luas was a mine for metal sulfurs (zinc, copper, lead, and nickel) operating till the 80’s. The need to collect residual hazardous wastes of an industrial site created in the area, were making it necessary to create a landfill in the dismissed mine. In order to have the maximum volume the use of a single embankment at the base was immediately disregarded, and it was substituted by a series of 10 embankments, with slopes inclined at 27° and 35°. This solution was superseded after the construction of the first 3 embankments, and the top 7 embankments were built with steep slopes. The increase in the volume available for wastes per running meter of superimposed embankments was over 1400 m³/m, and the saving in fill material was 150 m³/m. From 2006 to 2019 a total number of 7 superimposed reinforced embankments (total height of 36 m) have been constructed. Figure 7 shows a comparison between the two proposals, enhancing the volume increase.



GLOBAL STABILITY ANALYSIS
FACTORS OF SAFETY

CIRCLE	Fellenius	Jambu
1	*1,800*	*1,849*
2	1,979	2,035
3	1,812	1,862
4	2,236	2,232
5	1,977	2,050
6	2,383	2,462

FIGURE 3: Teglio landfill stability analysis.

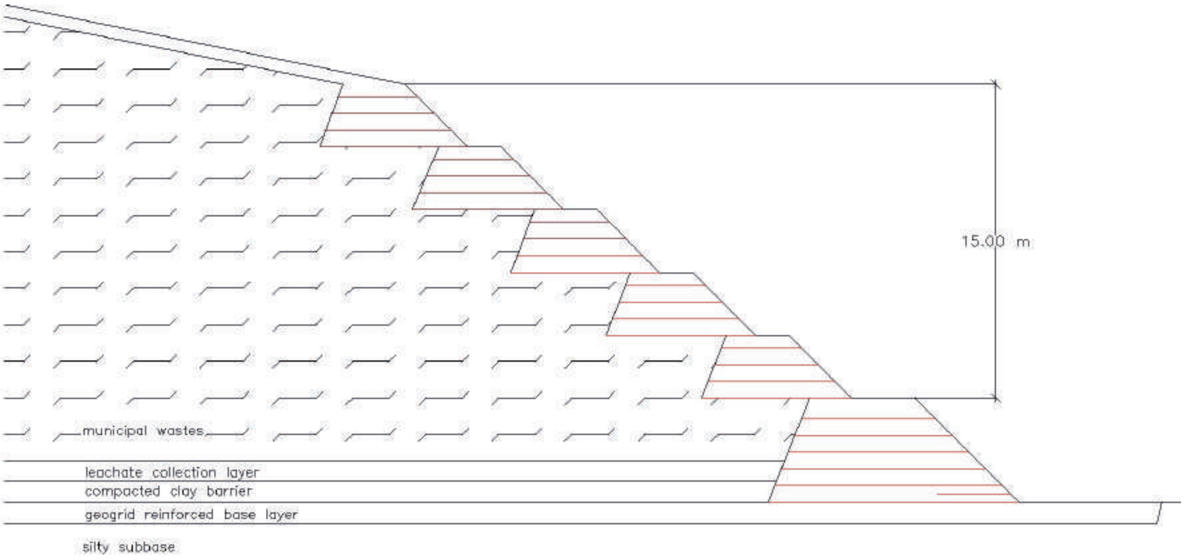


FIGURE 4: Teglio landfill layout.

The stability of the embankments has been studied before building every structure; the local stability of every embankment as well as the global stability of the whole landfill up to the level studied were evaluated. The different degree of compaction and consolidation of the wastes, that were showing a different behaviour in the short term and in the long term, was evaluated.

From geotechnical analysis, in the short term the material was purely frictional with Φ equal to 38°, while after consolidation it was turning into a cemented block, characterized by an undrained cohesion cu, measured from unconfined compressive test, equal to 2 MPa.

The fill material for the rienforced embankment was a sandy gravel coming from a nearby area characteirsed by a friction angle of 38° and a compacted unit weight of 19.5 kN/m³.

A limit equilibrium analyses (global stability as well as translational analyses on every geogrid layer) have been performed. The results both in the short and in the long term were well above the requirements of the regulations; Figure 8 shows the stability analysis of the landfill in the long term.

Details of this application are described in Figure 9, Figure 10 and Figure 11, while Figure 12 shows an aerial view during the construction of the top embankment with filling operation.

FIGURE 5: Teglio landfill; internal side of the landfill (compacted clay layer can be seen).

FIGURE 6: Teglio landfill; aerial view.

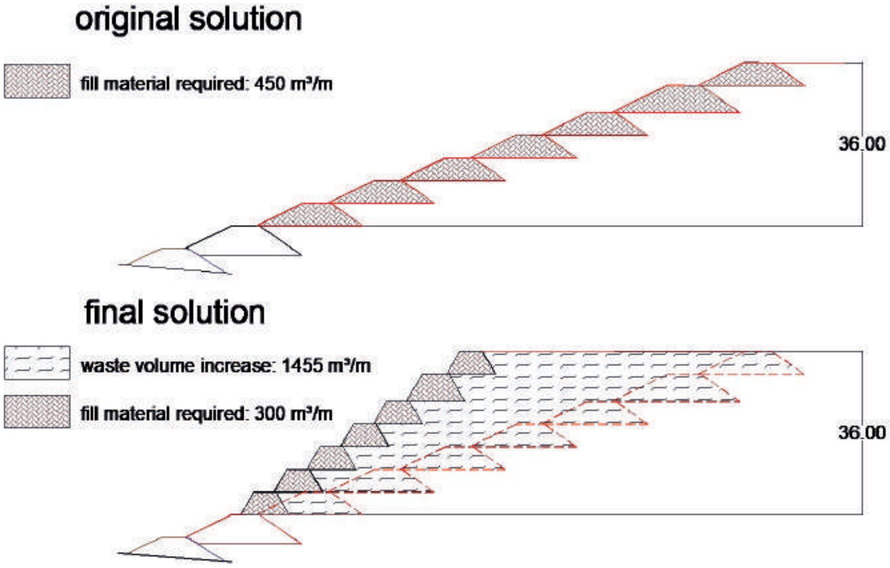


FIGURE 7: Comparison of traditional and reinforced embankments in Genna Luas landfill.

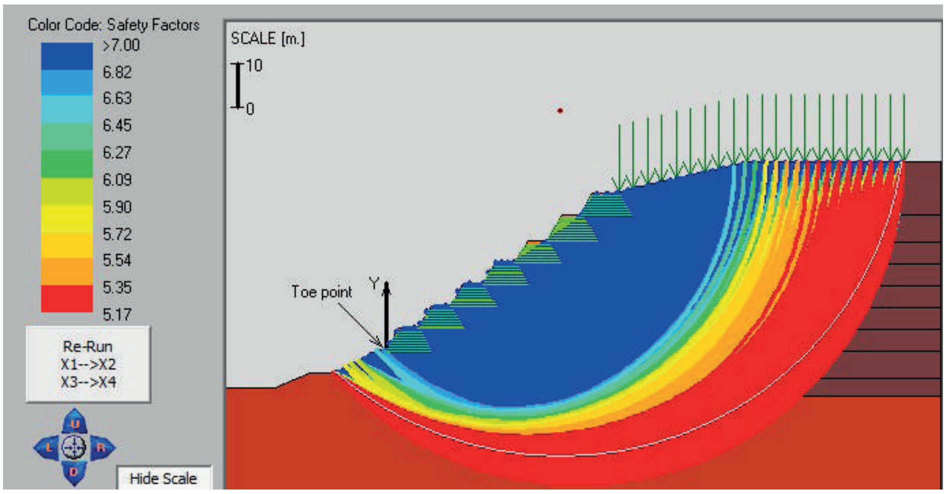


FIGURE 8: Long term global stability analysis results for the whole landfill.

FIGURE 9: Installation of geogrids in Genna Luas landfill.

FIGURE 10: Installation of waterproofing liner in the internal side.

FIGURE 11: Filling phase with ashes.

FIGURE 12: Aerial view during construction of the top embankment.

4. CONCLUSIONS

The use of steep slopes is getting more and more widespread in European, Asian and American countries for the construction of landfills, as it gives important advantages in terms of available volume, in terms of stability safety of the embankment and also in terms of costs. A proper selection of the type of geosynthetic should be done, considering the environment's boundary conditions typical of landfills. Successful examples of this technique, almost 25 years old, are present in Italy and in several other European, Asian and North American countries.

The solution of steep embankments allows to reduce the volume (and the cost) of fill material necessary and to increase the available volume for waste.

The use of geogrid reinforced steep slope guarantees important advantages as the composite material (fill + geogrids) has better compressive and shear behavior when compared to an unreinforced soil. These improvements give to a geogrid reinforced embankment the possibility to absorb quite large deformation without losing the integrity and functionality of the structure.

This aspect is particularly important when, rather than having a single embankment, a series of smaller size superimposed embankments are constructed.

This choice gives further advantages: the amount of fill material is reduced and the available landfill volume is further increased; it is possible to lay down the geosynthetics drainage and barrier systems only on a small portion of the landfill front face and not on the whole face, reducing the possible weathering of the geosynthetics and distributing according with the filling process of the landfill (and hence with the earnings by the owner) the cost of the purchase and installation.

The only possible minus of such solution could be the settlements under the portion of embankment directly laying over the wastes, particularly when the municipal wastes are present. The possibility to lay a large part of the bund directly upon the lower one, and the capability of the structure to absorb quite important strains and settlements without losing the functionality, allows somehow to overpass this problem.

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BIOGEOCHEMICAL AND MECHANICAL CHARACTERIZATION OF THE LANDFILL FRACTION GENERATED BY MECHANICAL WASTE SORTING

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ABSTRACT

When resource recovery from mixed waste streams is performed, new mixed waste streams are generated. Some of these waste streams does not fit well to existing waste management options, for example, they may hold a to low heating value to sustain combustion and they may have a too high carbon content to be accepted at non-hazardous landfills. Also various health and pollution risks may arise as well as practical handling issues due to the physical properties of such wastes. One such waste is the under sieve fraction generated when recovering metals and fuel from mixed waste streams using mechanical and magnetic separation tools. In this work, we examine the properties of one case of mechanical sorting of such under sieve fraction and analyse a broad spectrum of chemical, physical, and mechanical properties as well as some biological. Based on the data we develop recommendations for landfilling, what potential problems might arise and how to counteract them. The content of organic carbon and some trace elements is fairly high, but the biodegradability of the organic content is low and transports of water is also low in the material, especially after compression. We tested a range up to 700kPa. When building a mono fill with this material a special care should be given to dewatering the interior of it to avoid the buildup of water pressure. In summary we think that this material can be safely landfilled.

1. INTRODUCTION

A range of source separation and waste recovery systems are applied today to the majority of municipal solid waste (MSW) fractions in Sweden, whilst direct landfilling is limited to a small percentage of collected waste. Mixed waste streams such as bulky and construction wastes continue to be landfilled and are typically sorted on-site, mainly into a burnable fuel fraction, and a metallic fraction, which is recycled. The residual under-sieve fraction (see n.6 in Figure 1 and Figure 2 for a close-up) is mainly landfilled, at times being subjected to additional treatment. In line with current regulations, a total oxidizable carbon content (TOC) of up to 10% frequently constitutes the limit beyond which additional treatment should be applied prior to landfilling. In addition to TOC content, other constituents such as chloride leaching or metal content may also indicate the unsuitability of landfilling.

A case study of mechanical separation of a waste stream consisting of mixed bulky household waste and mixed construction wastes was investigated in the context of a master project carried out by Andersson & Jobs (2013). The study was performed in 2013 at the landfill of Högbytorp, 40 km northwest of Stockholm. The results obtained in this study are described here.

On-site procedures included shredding, separation of magnetic materials, and sorting of combustible materials by screening. The equipment used is shown in Figure 1. In this study, the landfilling of a non-magnetic fines fraction from the under-sieve fraction of mechanical sorting (USFMS) is investigated. The total flux of similar materials varies in line with landfill mining activities, construction and demolition activities and collection and handling routines. No standard statistics are available for this treatment residue, although based on case examples it is estimated to be in the order of one Mton/year in Sweden, or about 100kg per person per year, an amount comparable to levels reported for the food waste stream.

In this study we aim to cover a broader range of relevant aspects, for the management of this emerging type of waste, than we have seen in previously published literature. Mechanical properties, biodegradability and leaching properties of USFMS were characterized to assess the need for pre-treatment and type of treatment indicated, in addition

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to identifying a need for adaptation of the landfill design to mechanical properties of the waste. More specifically, the following questions were addressed:

1. How high is the pollution potential of USFMS in a landfill environment?
    - Degree of biological stability
    - Potential contributions to landfill emissions
    - Potentially beneficial pre-treatment methods
2. How should the properties of USFMS be considered in landfill design and operation?
    - Impact on stability (slope stability, settlement etc.)
    - Impact on transport of liquid phases
    - Influence on installations and treatment systems.
    - Implications for aftercare and post-operation usage

2. MATERIALS AND METHODS

2.1 Sampling

Over a 3-day sampling period, 360 m^3 (approx. 180 tons) of USFMS was collected (particle size < 18 mm). The USFMS collected over this period represented one month (December – January) of incoming, fresh waste received at the Högbytorp Landfill. An excavator randomly deposited three buckets out of every ten onto a separate pile and left the remaining buckets as spillage. The pile was developed in a linear row and two out of six segments were chosen (giving $180\cdot(3/10)\cdot2/6 = 18$ tons of USFMS in a separate pile). Inspired by Pierre Gy´s Theory of Sampling (Pitard, 1989), an attempt was made to reduce the degrees of freedom when subsampling. Thus, the pile was once again developed linearly and two out of four segments chosen randomly. Finally, one out of three segments was chosen randomly from the remaining row (giving $18\cdot(2/4)\cdot1/3 = 3$ tons for analysis). For the purpose of further subsampling in the laboratory, the 2D Japanese slab-cake method was applied together with the use of a large riffle splitter. This method emulates the field incremental subsampling process in a controlled laboratory setting: The entire sample is spread evenly onto a 2D surface at a depth that can be easily penetrated by a square scoop and a scoopful of USFMS representing a vertical column of the slabcake collected and the material placed in a container. This process is repeated at least 30 times at systematic random locations throughout the entire sample (ITRC 2017). Prior to the 3-day sampling period, a visual examination of the fresh, untreated waste was made. Observations made on the test rig are described in more detail in section 2.2.

2.2 Test rig observations

The majority of tests and analyses were performed in the test rig shown in Figure 3. Three cylindrical stainless-steel compression vessels with a depth of 0.7 m and a diameter of 0.5 m were used.

Compression vessels were filled with 0.07 m of water-saturated gravel (simulating a drainage layer in a landfill) and a 0.5 m layer of USFMS (approx. 65 kg) placed on top of the gravel, from which it was separated by means of a geotextile. During filling, total solid (TS) content was determined on random samples (100-150 g each) to establish

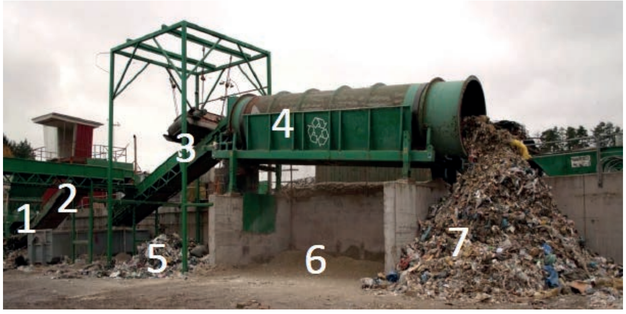

FIGURE 1: Pilot scale equipment for separation of magnetic materials, landfill fraction and fuel fraction. 1) mill, 2) conveyor belt, 3) magnetic separator, 4) drum sieve, 5) magnetic fraction, 6) under sieve fraction, and 7) fuel fraction.

FIGURE 2: The USFMS during sampling and measuring of the angle of repose.

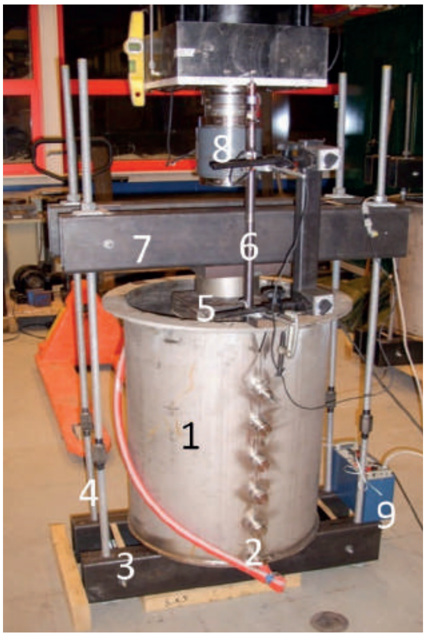

FIGURE 3: Test rig. (1) Compression vessel; (2) Outlet connected to hose; (3) Foundation; (4) Threaded rods; (5) Load cell and metal plate; (6) Linear variable differential transformer (LVDT); (7) Head beam; (8) Servo controlled cylinder, and (9); Driver for LVDT.

TS content of vessels. Subsequently, vertical compression of up to 700 kPa was applied to vessels to simulate normal stress at the bottom of an approx. 35m deep landfill. A supporting system ensured the maintenance of compression vessels under normal stress conditions of 700 kPa even after compression. However, the supporting system was not capable of maintaining normal stress at constant levels of 700 kPa due to compression in the USFMS. Accordingly, vessel compression was restored to 700 kPa at regular intervals. Six outlets facilitated water influx and leachate sample collection. Compression vessels were also used in determination of porosity, swelling pressure, field capacity, bulk density, dry density and compact density. The following observations were made on the test rig.

2.2.1 Compression

The compression test was conducted in the same way as an oedometer test due to lack of a suitable standard. Compression vessels used were larger than those of a standard test and during compression only upward-flow drainage was enabled. Compression was achieved in four steps due to a limited capacity of the servo-controlled cylinder. Compression was applied up to 136 kN, corresponding to a normal stress of 700 kPa. In the first step, the load was applied at 0.5 mm/s up to 13-25 kN, and in the second step at 0.5 mm/s up to 70 kN. The third step was intended to simulate a proctor compactor on a landfill, with load applied in 20 intervals at a range varying from 60 to- 80 kN. During the last step, the load was applied at 0.5 kN/s up to 136 kN. Ultimately, USFMS featured a constant load of 136 kN from supporting systems.

To compare compression in the three vessels with values reported in other studies, a compression ratio [Cr] was calculated. The compression ratio corresponded to the slope of the straight line in a diagram where compression [%] is related to normal stress [kPa] (see Figure 7). Logarithmic scale was not affected by differences in initial porosity.

2.2.2 Density

USFMS density was determined as follows: (1) with no load on sampling (2) before, during and after compression in the compression vessels (3) with a proctor compactor test and (4) in the shear boxes after packing.

Bulk density ρ is the ratio between total mass m and total volume V.

$$\rho = \frac{m}{V} \qquad [\mathrm{t/m^3}] \tag{1}$$

Dry density ρ_d is the ratio between dry mass m_s and total volume V.

$$\rho_d = \frac{m_s}{V} \qquad [\mathrm{t/m^3}] \tag{2}$$

Compact density ρ_s is the ratio between dry mass m_s and volume of dry mass V_s.

$$\rho_s = \frac{m_s}{V_s} \qquad [\mathrm{t/m^3}] \tag{3}$$

During sampling, both bulk density and dry density were determined on frozen USFMS. Containers (2*1 m^3) and beakers (1*0.001 m^3) were filled and weighed. Dry density was calculated based on observations made following randomized sampling during the process.

Bulk density and dry density were determined in compression vessels prior to start of compression (0 kPa), during the first compression step (65 kPa) and after the fourth compression step (700 kPa). During filling, USFMS mass was weighed and volume measured. Dry density was calculated on random samples. During the second compression step, water started to drain upwards onto the metal plate and water depth was measured following the last compression step; accordingly, density for the second and third steps was not determined. On reaching a compression of 700 kPa, calculation of compact density was facilitated due to the presence of water filled pores.

The proctor compaction test complied with SS 27109 standard. Two tests were conducted, one on a wet sample collected from the bottom of the container and one on a drained sample from the container surface.

After packing of the shear boxes, bulk and dry density were determined.

2.2.3 Unit weight

Unit weight γ is the ratio between total weight gm and volume V.

$$\gamma = \frac{g*m}{V} = g\rho \qquad [\mathrm{kN/m^3}] \tag{4}$$

Unit weight was calculated based on bulk densities. Good results were obtained for densities prior to compression, at the first and following the fourth compression steps (0, 65 and 700 kPa). As mentioned previously, during the second and third steps water had started to drain above the metal plate, therefore densities for the second and third compression steps were calculated based on the assumption that water drainage had not occurred. A trendline based on unit weight in the three vessels was calculated.

Unit weight was compared with the findings obtained by Zekkos et al. (2006) and Choudhury & Savoikar (2009). No swelling pressure was measured due to water drainage from USFMS during compression.

2.2.4 Permeability tests

Permeability tests were performed under constant head and upward flow at normal stress (700 kPa). An outlet hose (Ø 2 cm) was installed in the bottom outlet ((2) in Figure 3) of the vessel and connected to a water reservoir through a wall-mounted water hose. The head gradient was determined as the distance between the surface level in the water reservoir and the surface level in the compression vessel. In the upper outlet of the vessels a further hose was installed to collect spillage. After installation, vessels were filled with water. Measurements were undertaken once a clear equilibrium had been reached. The volume of spillage water was measured at intervals of 1 to 3 hours, with the extension of later intervals up to 16 hours. Permeability was determined by means of the equation:

$$K = \frac{Q}{A \times I} \tag{5}$$

where K = hydraulic conductivity [m/s]; Q = water flow [m³/s]; A = cross-sectional area [m²]; I = dH/dL = hydraulic gradient; dH = pressure head [m] and dL = water column [m].

A mean daily value was calculated for each vessel. The test lasted for a total of 20 days and a mean value of permeability was subsequently estimated from the mean value of each compression vessel.

2.2.5 Field capacity

Field capacity FC was determined by measuring the amount of water discharged above the metal plate ((5) in Figure 3) at maximum normal stress (700 kPa). The amount of free water (i.e. field capacity) retained by the USFMS at 700 kPA was estimated by subtracting the amount of water above the metal plate from the amount of water contained in the USFMS prior to compression. A mean value for field capacity was estimated from the mean value for each compression vessel.

$$FC = \frac{m_w}{m_d} \tag{6}$$

where FC = mass of free water per kg of dry weight [kg water/kg TS]; m_w =mass of free water [kg]; m_d = dry mass [kg].

2.2.6 Porosity

Porosity was measured at 700 kPa at known input values of TS (%). All pores were assumed as being water saturated. Water volume in the vessel at 700 kPa was calculated by subtracting the volume of water discharged above the metal plate from the total volume of water prior to compression. Porosity was then estimated by

$$n = \frac{V_p}{V} \tag{7}$$

where n=porosity [-], v_p = pore volume [m³] and v = total volume of USFMS [m³].

Three months after compression, a comparative value of porosity was calculated.

2.3 Other tests and analyses

2.3.1 Standard tests used

Table 1 shows the material properties of USFMS that were examined using standardized methods. Other testing proceedures are briefly described under 2.3.2-2.3.7.

2.3.2 Shear strength

Shear strength was determined by means of a direct shear test. The shear boxes had a size of 0.24*0.26*0.14 m and were packed with approximately 14-15 kg of USFMS. Three direct shear tests were carried out at three different normal stresses; 200, 300 and 400 kPa and the shear applied perpendicular to packed layers. To calculate shear strength, shear failure was assumed to occur at a deformation of 25 mm (although the test was carried out up to a deformation of 30 mm). The shear strength envelope with cohesion intercept (c) and friction angle (φ) was calculated according to the Mohr-Coulomb failure criterion.

The Mohr-Coulomb failure criterion may be expressed as an equation for the line representing the failure envelope. The general equation is:

$$\tau_f = c + \sigma_f . \tan\phi \tag{8}$$

where τ_f = shear stress on the failure plane
c = apparent cohesion
σ_f = normal stress on the failure plane
φ = angle of internal friction

2.3.3 Angle of repose

During subsampling the angle of repose was determined trigonometrically on three heaps of 360 m³ frozen UFSFM.

2.3.4 Soil gradation

Based on results obtained from the dry and wet sieve the uninform coefficient (Cu) was calculated by:

$$Cu = \frac{d_{60}}{d_{10}} \tag{9}$$

where d_{60} is the grain diameter at 60% passing and d_{10} is the grain diameter at 10% passing.

2.3.5 Column test

On reaching a 700 kPa load, water was added at the bottom of the models and leachate collected from each vessel through the third to bottom outlet. L/S ratio was calculated to be approx. 0.6.

2.3.6 Agitation batch tests (standard leaching test)

In addition to permeability measurements in the com-

TABLE 1: Standard tests used.

Property tested	Method id
Mechanical characterization	
Grain size distribution	Dry sieving (SS-ISO 11277)
	Wet sieving (SS-EN 933-1)
Density	Proctor compactor (SS 027109)
Biogeochemical characterization	
Total content	Total solids,TS (SS-EN 14346)
	Volatile solids, VS (SS-EN 15169) Total organic carbon, TOC; Total carbon, TC; Total incinerated carbon, TIC (SS-EN 13137), Total content of elements Modified EPA methods 200.7 (US EPA, 1994a) and 200.8 (US EPA,1994b) were used for the total element composition analysis with ICP-AES/SFM
Leaching properties	Agitation batch test, (L/S 10) (SS-EN 12457-4)

pression vessels, agitation batch tests were also performed at a liquid/solid ratio of L/S 10. Tests complied with Swedish standards, but were modified for use under anaerobic conditions by adding 0.7 g of 1%-methanol prior to agitation. The amount of methanol required to consume oxygen in 500 ml of water was calculated by: $2CH_3\,OH+O_2 \leftrightarrow 2HCHO+2H_2O$.

The bottles were sealed with butyl rubber stoppers, air was expelled and replaced by nitrogen gas. The bottles were stored in a dark place for two days before agitation. Leachate was collected using an injection needle and directly filtered before analysis.

2.3.7 Gas potential

Gas potential was determined indirectly by promoting complete mineralization, assessing chemical oxidation, and measurement of direct biochemical methane potential (BMP). Complete mineralization of USFMS was achieved in a bomb calorimeter, whilst chemical oxidation was evaluated through determination of COD. COD content was measured after blending USFMS with water to an L/S ratio of 100, which was then mixed and homogenized. COD content in 2 ml of water after 24 hours agitation was analyzed in a COD Cell Test (MERCK).

Biological degradability was assessed by means of biochemical methane production assays (BMP) using 18 glass bottles (120 ml) prepared with inoculum and substrate (USFMS) at a ratio of 1:1, 1:2 and 2:1, respectively (based on g VS). Total volume of inoculum and substrate in each bottle was approx. 50 g. To optimize degradation, the mixture was crushed and homogenized before bottles were filled. Anaerobic conditions were achieved by sealing the bottles with butyl rubber stoppers. BMP test was carried out at a temperature of 30°C and 55°C (9 bottles at each temperature). At both temperatures, 3 blank tests were performed with bottles containing inoculum alone. The volume of produced gas was measured until gas production was negligible.

All observations for organic content were compared using corresponding amounts of methane. For this purpose, the calorific value of methane was used as comparison with the measured heating value of the material; a 40% assumed carbon content and 0 carbon oxidation state were adopted as comparison with VS, using the same assumed oxidation in conversion of TOC values to carbon; COD was merely taken as four-fold the CH_4 mass. The CH_4 content of formed gas was assumed to be 50%.

3. RESULTS AND DISCUSSION – POLLUTION POTENTIAL

The biogeochemical properties of USFMS, i.e. degree of biological stability and potential landfill emissons, are of importance when discussing the materials pollution potential. These aspects should also be taken into account when considering beneficial pre-treatment methods.

3.1 Is the waste biologically stable?

The observations made regarding organic content are illustrated in Table 2 together with an estimation of corresponding methane potential based on related properties.

This is for comparison and for illustrating the character of the material. The methane amounts have been determined from the other observations as; $COD/4=CH_4$ (g/g) and from the heating value by deviding by that of CH_4. VS_{1000} and TOC values can be compared to establish carbon content of the material. Assuming that a mean oxidation of the carbon corresponds to zero $VS*0.3*16/12=VS*0.4=CH_4$. The corresponding amount of methane estimated from the TOC value will be $TOC/2*16/12$.

The data obtained however raises a few issues, namely, COD is lower than TOC and COD/ BMP ratio corresponds to approx. 25% of expected values. In spite of the highly variable material and small analytical samples, the majority of observations fall into a fairly narrow range when expressed as corresponding methane amounts, with the exception of COD and BMP. In both cases, these values reflect the fate of solubilized organics, which hint at a limited rate of solubilization of the material. When assessing data quality, observations made for VS are those most likely to reliably reflect the amount of organic material available, particularly as the largest samples with the lowest degree of inter-sample variation were used for VS measurement.

The marked variation of VS in line with temperature highlights the scarce volatilization of the material. Moreover, heating value is low in comparison to TOC and VS, possibly due to the presence in the material of flame retarding substances such as PVC. Thermal stability is also an indicator of low biodegradability.

A COD/TOC ratio of approx. 0.6 was detected, low-

TABLE 2: Average observations related to organic material and corresponding calculated amount of methane. N=3.

Component	Unit	Average	Standard dev	CH_4-calc g/kg TS
COD	g/kg TS	90.59	22.2	23
TOC	g/kg TS	151.4	43.8	101
VS 550°C	g/kg TS	146.5	5.06	60
VS 750°C	g/kg TS	178.4	5.98	73
VS 925°C	g/kg TS	180	6.01	74
VS 1000°C	g/kg TS	246	2.58	101
Heating value	MJ/kg TS	2.98	1.06	60
BMP	l/kg TS	31.5	1.4	23

er therefore than the ratio obtained for any organic substance, with a typical range exceeding 2 up to a maximum for methane of 5.33. This is a further indication that only a fraction of the organic material was actually oxidized in the test environment. USFMS therefore may be classified as an almost stable waste, although featuring a high carbon content.

3.2 To what extent does the waste contribute to landfill emissions?

Bearing in mind the variations in sampling and analytical procedures, the findings obtained indicated that a prevalent share of the material was comprised of scarcely degradable polymers with a high carbon content. Thus, in an anaerobic landfill environment the material degrades very slowly, with the resulting methane emissions approx. one order of magnitude lower than those yielded by mixed household waste. Thus, passive treatment methods such as design of the landfill cover to act as a suitable habitat for methane oxidizing biota, may be sufficient to reduce methane emissions.

The compressed material however features low permeability, thus implying that even slight gas generation may result in substantial pressure build-up. Accordingly, it would be prudent to add a series of vertically-connected drainage layers throughout the landfill body to allow for the release of pressure and channeling of the gas generated to methane oxidation areas.

The total content of several hazardous trace elements (As, Ba, Cr, Cu, Ni, V, Zn) was higher than that reported in previous studies performed on excavated and screened waste (Table 3). The lower concentrations found in other studies could be due to the presence of sand and gravel from daily cover in landfilled and excavated waste. Due to a lack of relevant studies in literature, no comparison of the metal content in fresh USFMS was possible. Several elements (As, Ba, Cr, Cu, Zn) also exceeded Swedish EPA guideline values relating to the less sensitive use of land. A large inter-sample variation was observed, and in some cases (Cr, Cu and S), a heavily skewed distribution.

TABLE 3: Total content of elements (mg/kg TS) in USFMS (mean value based on 3 samples). Values marked by red and red bold type indicates the exceeding of Swedish EPA guideline values for sensitive and less sensitive land use, respectively. Concentrations obtained in previous studies are listed in right-hand columns.

Element	USFMS	±	(IWCS, 2009)1)
TS (%)	51.3	2.55	
As	**31.7**	19.1	2.0 – 7.8
Ba	**1167**	425.7	98 - 265
Be	0.97	0.053	0.10 – 0.21
Cd	1.76	0.305	1.4 – 8.0
Co	14.6	5.41	0.8 – 2.0
Cr	**151**	47.4	12.8 – 35.9
Cu	**1116**	1299	19.0 – 96.5
Hg	0.279	0.135	0.03 – 0.35
Mo	4.47	2.88	
Nb	6.31	0.842	
Ni	88.2	32.8	7.7 – 43.4
Pb	274	107	9.7 – 900
S	15 758	23296	
V	48.2	5.64	9.4 - 17
Zn	**1490**	260	130 - 680

1) Analysis performed on 8 samples after screening (<25.4 mm) of excavated waste from a depth of approx. 4.6m in Perdido Landfill, Florida.

The results of leaching tests are shown in Tables 4 and 5 and Figures 4 and 5. The elements exceeding threshold values for disposal of waste in a Swedish class 2 landfill (non-hazardous landfill) were Sb and Zn. Calculations revealed that the various concentrations obtained at L/S ratio 10 corresponded to 0.1-5.3% of the total content, excluding Mo, Sb and S (92.5; 21.7 and 39.5% respectively). In addition to Sb and Zn, the only element exceeding threshold values for disposal of waste is DOC – a result which, combined with a TOC of 15.1% probits landfilling of the material. The organic material content of USFMS, both total content and dissolved in leachate, tends to indicate another source of emission. Runoff water collected during thawing featured similar concentrations of elements to leachate at L/S 0.6. Drainage water will need to be collected and treated to prevent emissions from impervious surfaces.

The difference in concentration of Cl at L/S ratio 0.6

TABLE 4: Concentration of elements in leachate (mean value based on 3 samples) from column test (L/S ratio 0.6) in compression vessels (Ø 0.5 m) at normal stress 700 kPa. Values marked in red exceeds threshold limits for disposal of waste in Swedish class 2 landfill (non-hazardous landfill).

Element	Unit	USFMS L/S 0.6	[±]	c_0 (L/S 0,1)1)
Al	µg/l	246	57.9	
As	µg/l	27.9	10.2	300
Ba	µg/l	368	47.2	20000
Cd	µg/l	1.55	0.100	300
Co	µg/l	103	14.6	
Cr	µg/l	40.8	5.12	2500
Cu	µg/l	104	28.9	30000
Hg	µg/l	0.355	0.03	30
Mn	µg/l	9377	1301	
Mo	µg/l	156	55.1	3500
Ni	µg/l	479	119	3000
Pb	µg/l	24.0	1.60	3000
Sb	µg/l	77.2	7.47	150
Se	µg/l	6.05	1.79	200
Zn	µg/l	9960	1760	15 000
Cl	mg/l	1720	255	8500
F	mg/l	< 0.500	-	40
SO_4	mg/l	2067	176	7000
DOC	mg/l	4150	771	250
P-tot	mg/l	0.463	0.249	

1) 30 § NFS 2004:10 (Swedish EPA. 2004)

TABLE 5: Concentration of elements (mg/kg TS) in USFMS leachate from agitation batch tests (L/S ratio 10) under anaerobic conditions (mean value based on 3 samples). Anaerobic conditions were obtained by the addition of methanol (0.007 ml/500 ml of water). Values marked in red indicates the exceeding of threshold limits for disposal of waste in a Swedish class 2 landfill (non-hazardous landfill).

Element	USFMS L/S 10	±	c_0 (L/S 10)1)
Al	3.18	0.392	
As	0.657	0.0591	2
Ba	9.57	0.224	100
Cd	0.0152	0.00119	1
Co	0.350	0.0291	
Cr	0.289	0.00934	10
Cu	1.14	0.114	50
Hg	0.00536	0.000116	0.2
Mn	56.7	4.57	
Mo	4.13	0.0911	10
Ni	1.70	0.0497	10
Pb	0.642	0.0297	10
Sb	3.44	0.0623	0.7
Se	0.232	0.00469	0.5
Zn	79.7	9.99	50
SO4	17 984	44.2	20 000
Cl	1358	3.79	15 000
F	< 5.25	0.0207	150
DOC	1963	63.6	800
P-tot	6.46	0.155	

and 10, respectively, corresponds to the difference in dilution between the two tests. Likewise, redox-sensitive elements such as Cr, Co and Fe, also display differences in concentration corresponding to difference in dilution. Thus, redox-sensitive elements are reduced to a similar degree in both tests, with methanol addition being assumed to have acted as a reduction agent for oxygen in the agitation batch test.

Emission potential is present in both gas and liquid phases; however, given the slow degradation rate and low permeability of the material, only low intensity emissions are expected, which may promote the use of passive treatment methods. The exception would be run-off from surfaces on which shredding and intermediate material storage is carried out.

3.3 Potentially beneficial pre-treatment methods

The separation of degradable organics could be improved by additional processing, although costs may rise beyond the impact produced. Additional treatments such as washing or hydrothermal carbonation could be considered.

Taking into account the biogeochemical properties of USFMS, there may be a small potential for bioconversion, although the limited permeability would render composting largely inefficient. Application therefore of a simple form of biological treatment would not appear to be a feasible option.

Other options may include chemical oxidation, electro techniques or mixing with stabilizers, e.g. washing dust from gravel production. These options would all be possible, although the implicated costs may be prohibitive.

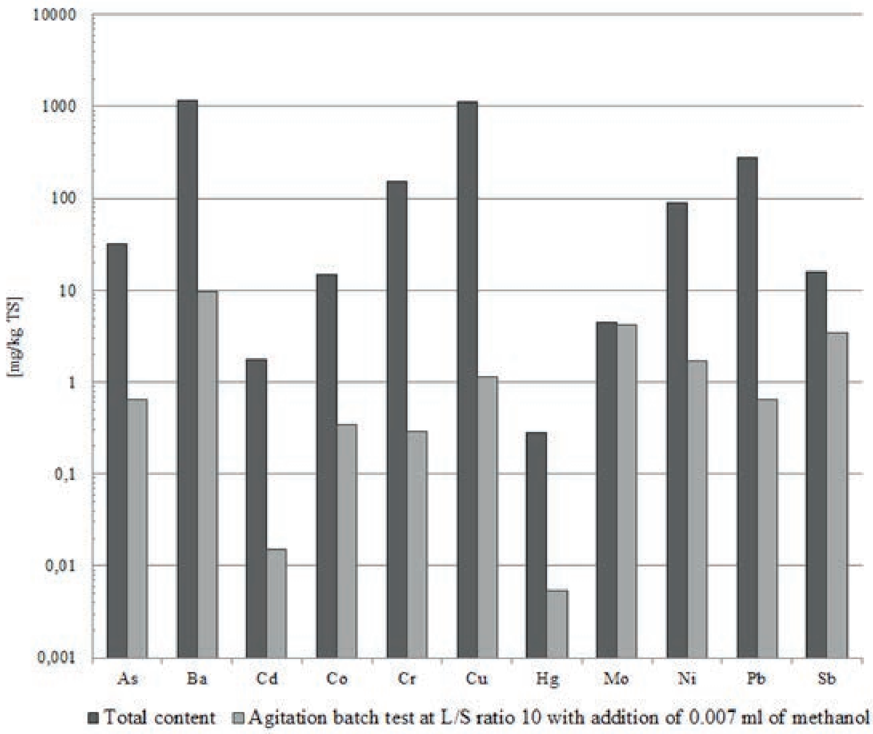

FIGURE 4: Concentration of elements in USFMS leachate at L/S ratio 10 compared to the total content in USFMS. Methanol was added to achieve anaerobic conditions in sample bottles.

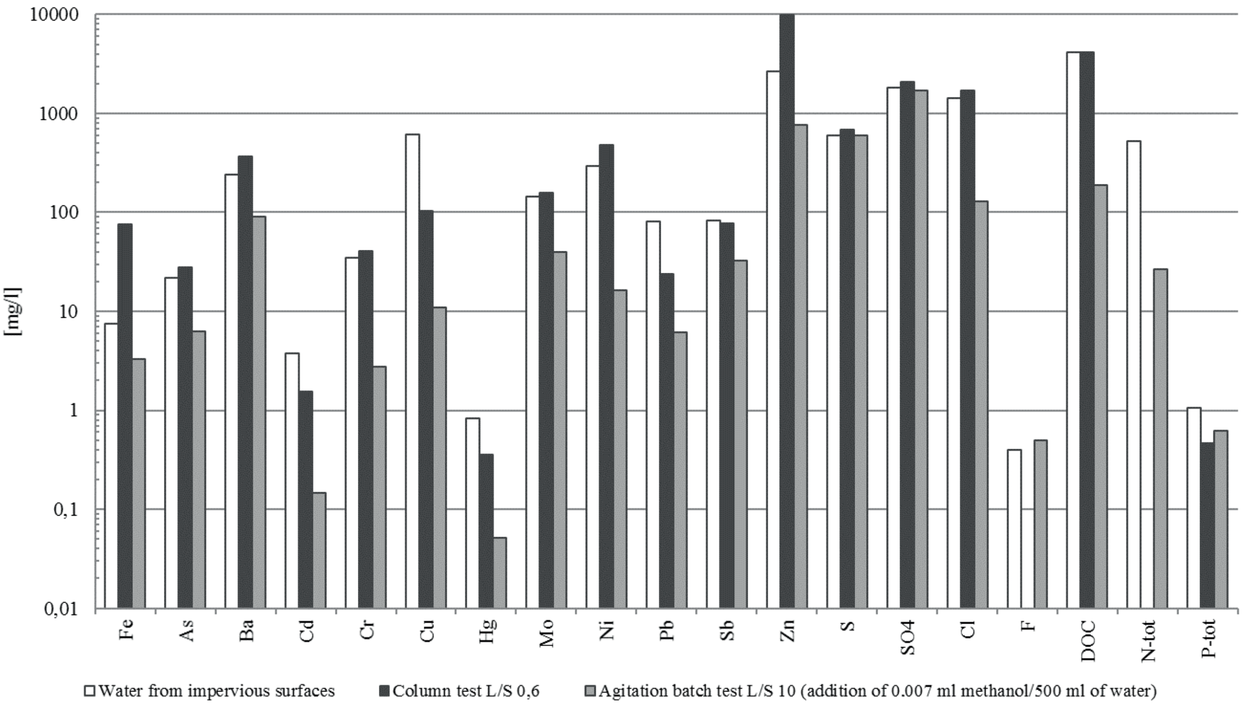

FIGURE 5: Concentration of elements in leachate from USFMS; water from impervious surfaces (1 sample, filtered), leachate from column test at L/S ratio 0.6 (3 samples, non-filtered) and leachate from agitation batch test at L/S ratio 10 (3 samples, filtered).

4. RESULTS AND DISCUSSION – LANDFILL DESIGN AND OPERATION

The mechanical properties of USFMS, capable of impacting on slope stability, settlement, water-holding capacity and permeability, should be taken into account when designing and operating a landfill. Some of the more critical aspects are discussed below.

4.1 Mechanical properties

A summary of the results of mechanical tests are shown in Table 6.

4.2 Impact on stability

4.2.1 Slope stability

If shear stress is higher than shear strength, slope failure may occur. Shear strength is largely dependent on three factors: (1) friction in the contact surface between individual particles, (2) wedging of particles and (3) adhesion between particles (SGI, 2007). Accordingly, in an MSW landfill shear strength is influenced by waste composition, degree of compaction, daily cover, moisture conditions, age, leachate management and overburden pressure.

Test results reveal how the shear strength envelope of USFMS is governed by a cohesion intercept c = 26 kPa and friction angle φ = 47°, see Figure 6. Shear tests were performed up to a horizontal shear displacement of 30 mm, without producing any shear failure. According to Stark et al. (2009), shear failure was assumed to occur at 25 mm, as is routine for underlying soils or sealing layers. In this study, the shear test was performed perpendicular to packing layers, thus preventing measurement of critical shear strength. In previous studies, (Zekkos, 2010; Reddy et al., 2009; Stark et al., 2009) the cohesion intercept c generally ranged from 6 -30 kPa and the friction angle φ ranged from 30-35°. Therefore, in the present study the cohesion intercept is at the high end of a normal range, whilst the friction angle is higher. Figure 6 show shear strength of the USFMS compared to models of shear strength reported in previous studies. Hossain et al. (2009) demonstrated a reduction in the friction angle at a lower R-value, where R-value is represented by particle size/equipment size ratio. A study by Dewaele et al. (2011) showed a reduction in cohesion intercept after landfill mining and out-sorting of larger items. A lower shear strength would be expected for USFMS, subjected to milling, compared to untreated MSW.

The high friction angle in the shear strength envelope may be explained by grain size distribution and waste composition. Result obtained for grain size distribution by wet sieve highlighted a small grain size distribution (45% gravel, 35% sand and 20% silt) with a high uniform coefficient (Cu40), see Figure 11 and 12. A material with a high uniform coefficient is evenly distributed between different grain sizes, thus allowing smaller grains to fill the voids between larger grains. This contributes to a more stable structure which produces a positive effect on shear strength. Shredding may also result in edgy particles which contribute to increased friction between the particles. The material itself contains fibrous material with a high specific surface, such as mineral heat insulation products. Fibrous and edgy particles add to the shear strength. Wood splinters, wires, etc. protruding from the failure planes were observed. In conclusion therefore, the risk of slope failure appears to be lower for this type of material than for ordinary MSW.

Dry density is used as a measure of the degree of com-

TABLE 6: Mechanical properties (Standard deviations are given in brackets).

Mechanical property	Result
Angle of repose	31.7° (±2.29)
Grain size distribution	Dry sieve 45% gravel. 50% sand. 5% silt. Cu20 Wet sieve 45% gravel. 35% sand. 20% silt. Cu40
Shear strength	c = 26 kPa. φ = 47°
Compression ratio	C_r = 0.19 (±0.006)
Density	1. During sampling ρ = 0.62 t/m^3 (±0.07). ρ_d = 0.29 ton/m^3 (±0.03) TS 47.1% 2. Compression test ρ = 0.68 t/m^3 (±0.05). ρ_d = 0.42 ton/m^3 (±0.03) at 0 kPa ρ = 0.92 t/m^3 (±0.1). ρ_d = 0.59 t/m^3 (±0.06) at 65 kPa ρ = 1.55 t/m^3 (±0.06). ρ_d = 1.02 ton/m^3 (±0.04) ρ_s = 2.20 ton/m^3 (±0.2) at 700 kPa TS 62.2% ρ = 1.50 t/m^3 (±0.0). ρ_d = 1.01 ton/m^3 (±0.03) ρ_s = 2.01 ton/m^3 (±0.05) at 700 kPa for three months TS 66% 3. Proctor compactor test ρ = 1.37 t/m^3. ρ_d = 0.67 t/m^3 . ρ_s = 2.14 at 500 kJ/m^3. TS 48.73% ρ = 0.7 t/m^3. ρ_d = 0.51 t/m^3 at 500 kJ/m^3. TS 64.57% 4. Shear test ρ = 1.56 (± 0.06) t/m^3. ρ_d = 1.01 (±0.05) t/m^3 at after shear box packing at TS 64.9%
Porosity	n = 53.98% (±2.09) at 700 kPa n = 49.82% (±3.04) at 700 kPa for three months
Field capacity	54 (±0.2)%
Permeability	6.7×10^{-9} (±2.5×10^{-9}) m/s

paction in shear boxes. There was no significant difference between dry densities in the three shear boxes, with density before shearing being almost as high as dry density measured after compaction to 700 kPa. The material in hear boxes may have been over consolidated (I e the material has earlier been exposed to higher normal stresses) before shearing and the resulting measured shear strength overestimated; maximum shear strength was not however reached during the shear test.

During compression, USFMS was found to be water saturated during the second compression round (from 67- 135 kPa to 360 kPa), thus implying release of water from USFMS under pressure. No swelling pressure was observed.

TS (47 ± 6%) was lower in USFMS as compared to previous studies on excavated and screened landfill fractions (Dewaele, 2011). It should however be noted that TS was determined for USFMS samples collected at temperatures below 0°C, and therefore considered as "frozen" material, likely to hold more water than a thawed drained sample. For drained USFMS, TS of 64 ± 1.7% was determined, therefore in the same range as TS for fresh MSW. Visual examination revealed a high content of absorbing materials (madrasses, isolation etc.) in USFMS, which likely explains the low TS obtained when frozen. It is reasonable to assume that in a future landfill TS will be subject to seasonal variations due to rain storms, dry winters etc. The results

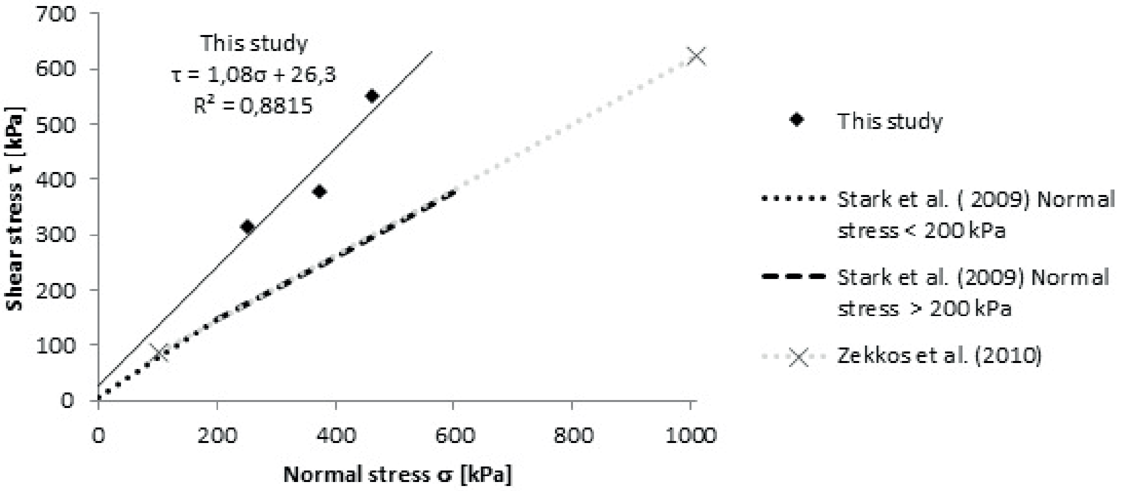

FIGURE 6: Shear strength envelope for USFMS and results from previous studies. R2 denotes the coefficient of determination in the linear regression formula.

demonstrate the ability of USFMS to absorb a high quantity of water - a good draining system is therefore mandatory to avoid the risk of pore water pressure increase in the landfill.

Turer and Turer (2011) adopted a simplified slope stability approach for the purpose of developing slope stability charts for use in analysis of uncontrolled waste dumps. These can be assumed to yield the worst-case scenario for USFMS. Analysis is performed using the shear strength envelope according to Stark et al. (2009) and unit weight equation of Choudhury and Savoikar (2009), based on the assumption that the failure plane passes through the toe of the slope. To assess USFMS input data comprised a slope height of 35 m, safety factor of 1.5, bulk density measured at 700 kPa corresponding to unit weight of USFMS at 35 m depth, and a pore water pressure ratio of 0.3. This pore water pressure corresponds to half the fill height being water saturated. As pore water pressure rises the effective stress decreases. A decrease in effective stresses will result in poorer stability. With regard to USFMS, simplified slope analysis yields a critical slope angle around 18°. Swedish EPA (2004) recommends a slope angle between 1:2 (~ 26.6°) and 1:3 (~ 18.4°) in the final cover. USFMS features a higher shear strength and a higher unit weight, which contributes to a better stability than that recommended by simplified slope analysis. Moreover, the results underline the importance of ensuring an effective draining system.

4.2.2 Settlements

Total settlements are divided into primary- and secondary settlements. Primary settlement is affected by load, which in turn depends prevalently on fill height and waste density. Secondary settlements are produced by mechanical creep and biodegradation. Creep is largely caused through particle stiffness, with biodegradations being linked to chemical composition of the materials, moisture, and temperature.

Compression ratio C_r is deemed to correspond to primary settlements. The measurements obtained are provided in Figure 7.

Compression ratio detected for USFMS was in the normal range, although lower than that observed in previous studies, see Table 7.

According to Hossain et al (2009), a waste with a high R-value has a lower compression ratio due to the presence of large items that produce a reinforcement effect. R-value is the ratio of particle to equipment size. R-value of the waste studied here was low (0.036) following the shredding process. Shredding may contribute to the presence of more angular particles capable of increasing inter-particle friction. A lower organic material content may likewise contribute to the finding of slightly lower compression ratio than that detected for MSW.

USFMS was in a position to drain upward during compression, therefore, the high water content of USFMS during compaction (TS of 62 (± 1.1) in the compression vessels) likely did not affect the compression ratio.

Unaffected by load, the bulk density for USFMS, 0.65 t/m^3 represents the average density of both frozen and unfrozen USFMS. The results of this study reveal how bulk density is affected by transport handling (shaking), thawing and drainage potential. Bulk density and dry density for USFMS under high pressure were found to be higher than those observed in previous studies conducted by Beaven and Powrie (1995) and Hudson et al. (2004) . This finding may be explained by waste composition, i.e. a high content of heavy materials such as concrete and tiles, but also due to a higher compression of USFMS. As reported by Beaven and Powrie (1995) and Hudson et al. (2004), waste was compressed to 600 kPa and 463 kPa, respectively. According to Stoltz et al. (2010) a normal compact density for American MSW is 1.6-1.65 ton/m^3. USFMS features a lower organic material content due to mechanical separation and is thus heavier.

In the 3rd compression round 20 compression intervals of 60-80 kN ~ 305-410 kPa were used, aiming to simulate a compactor working at a landfill. There was no visible difference in compression after the 20 compression intervals.

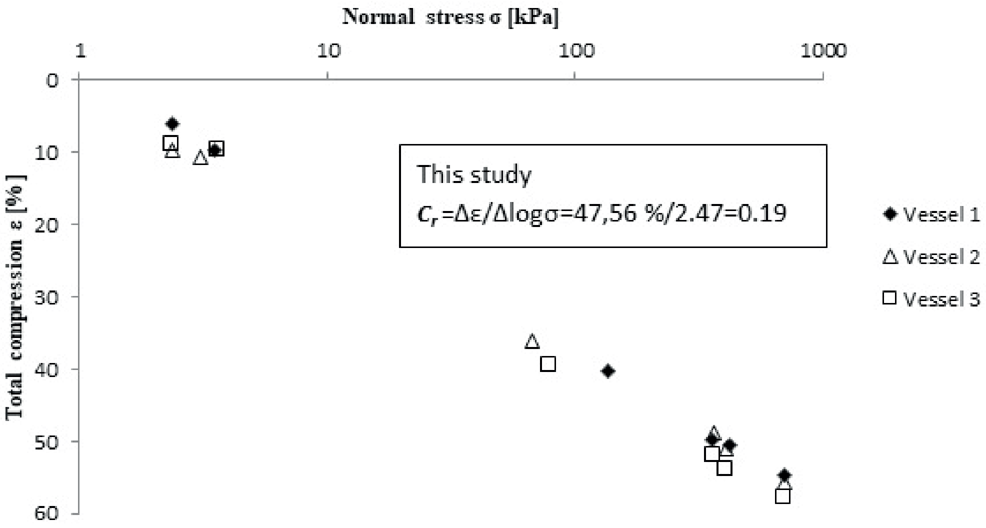

FIGURE 7: Total compression ε [%] as function of normal stress σ [kPa] in log scale for the three used cylinders. The compression index Cr is defined as the slope of the curve.

TABLE 7: Compressibility of USFMS and different MSW.

Source	σ [kPa]	C_r [-]	Method	Comments
USFMS	700	0.19	See Chapter 2.	
Reddy et al. (2009)	750	0.24-0.33	Oedmeter test	Fresh MSW. Estimated maximal normal stress from a graph. Different water content.
Stoltz et al. (2010)	140-300	0.31	Oedmeter test	Fresh MSW. Shredded before testing. maximal waste size 0.07 m.
Hossain et al. (2009)	1050	25.5-34.5	Oedmeter test	Fresh MSW. Different R-values*.
Reddy et al. (2009) quoted by Hossain (2005)	-	0.16-0.25	Oedmeter test	Fresh MSW. Water saturated.

** where the R-value is the ratio of particle size to equipment size.*

Compared to previous studies investigating MSW (Machado et al., 2012) USFMS is characterized by a smaller grain size. According to Machado et al. (2012), grain size distribution increases in line with landfill age due to degradation of organic material. USFMS contains less organic material than a traditional MSW and grain size distribution will not be affected to the same extent by degradation; the risk of secondary settlements produced by organic degradation is likewise lower than for traditional MSW.

4.2.3 Deformations over time

The initial settlements of a UFSFM landfill were estimated from measured unit weights and deformations in the compression vessels. The following assumptions were made:

- A proctor compactor vibrated all waste layers successively applying a normal stress of 40 kPa;
- The stress from the final cover is comparatively small and can thus be included in normal stress from the compactor;
- The landfill is divided into one-meter layers and unit weight is assumed to be constant within each layer.

Unit weight is taken as the mean value of bulk density in the three compression vessels. The measured unit weight for normal stress of circa 400 kPa remains unclear as the amount of water that drained from compression vessels was not measured continuously, but was first measured on completion of compression at 700 kPa.

Figure 7 shows an increase in unit weight from 7 to 15 kN/m^3 on a par with an increase in normal stress from 0 to 700 kPa. Unit weights of all landfill layers were affected by normal stress from a compactor of 40 kPa. The trend line from 40 kPa and upwards in Figure 8 was used to calculate unit weight as a function of landfill depth. Unit weight of the top layer was only subjected to 40 kPa and was calculated as 8.432 kN/m^3. The weight of the USFMS added to overall unit weight for every subsequent landfill layer as shown in Figure 8, increasing for a 36m landfill from 8.4 kN/m^3 at the top to approx. 14 kN/m^3 at the bottom.

To calculate deformations throughout the different layers, the curve in Figure 9 was used. This curve describes the compression (%) measured during compaction of the vessels. Normal stress in each layer is based on unit weight at the corresponding depth. As each layer had pre-

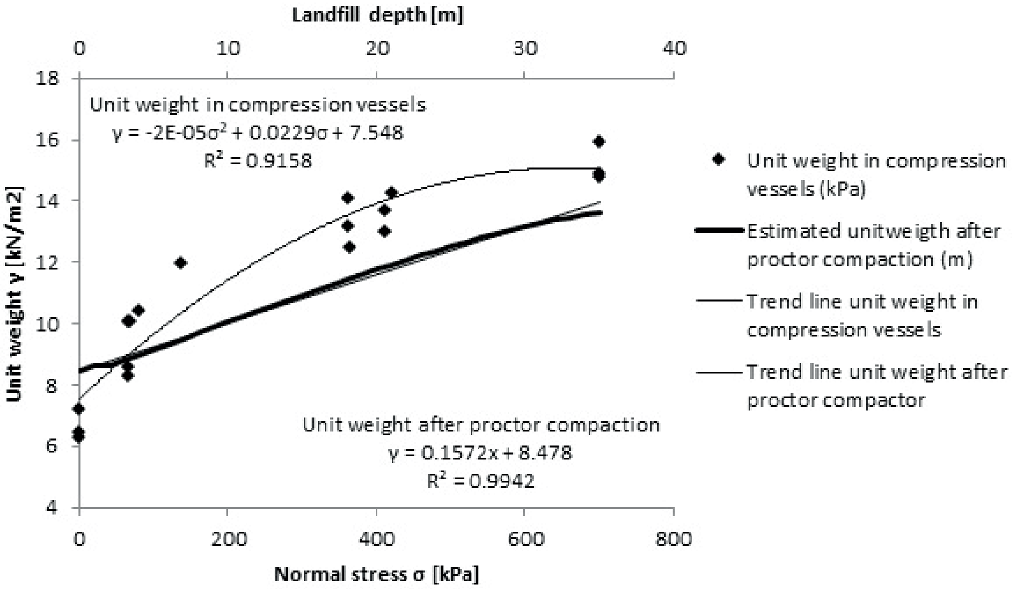

FIGURE 8: Unit weight in the three compression vessels as function of compaction stress and a trend line indicating mean value for the three vessels. Calculated unit weight as function of landfill depth after compaction with 40 kPa.

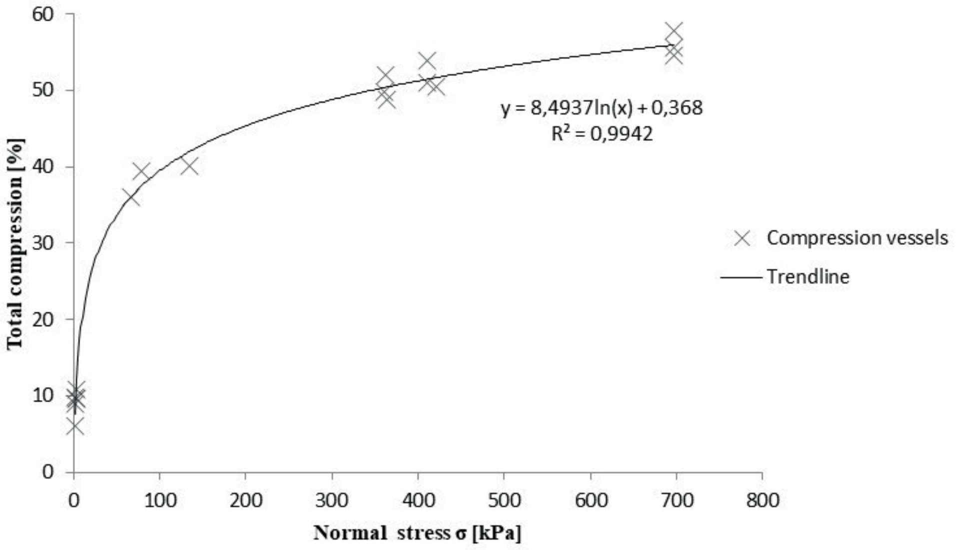

FIGURE 9: Total compression in% and trend line.

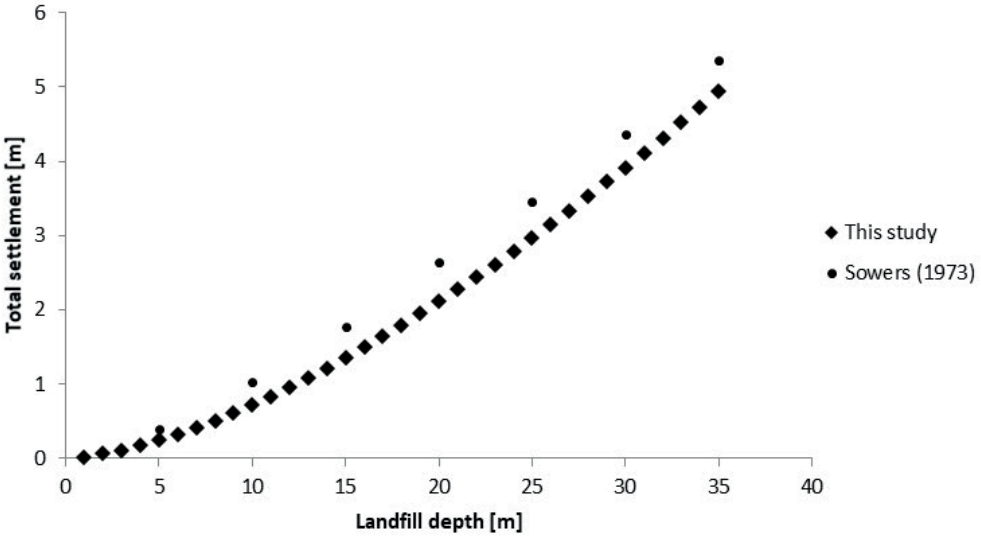

FIGURE 10: Calculated total settlements for a USFMS landfill.

viously been compressed up to 40 kPa, only deformations for higher values were used.

The initial settlement calculated using this approach was 4.94 m, corresponding to 14% - see Figure 10. For the sake of comparison, the initial settlement was also calculated using the method described by Sowers (1973). Using the same unit weight as for UFSFM and 5m layers, total settlements amounted to 5.64 m, corresponding to 16.1%.

Maraques et al. (2003) developed models for use in calculating final settlements based on primary compression, mechanical creep and biodegradation. Babu et al. (2011) calculated total settlements of MSW using three models for a landfill height of 30 m. The initial settlement ranged from 18-19.2%, the secondary settlement for creep was 2.3-2.8%, and biodegradation was 7.9-10.1%. USFMS has a lower organic content than traditional MSW and therefore settlement due to biodegradation may be assumed to be smaller. For USFMS, secondary settlement can be assumed as being inferior to 10%, corresponding to 3.5 m in a 35m landfill m.

Estimated total settlement, including initial and secondary deformations, therefore corresponds to approx. 8.5 meters, indicating a reduced tendency to deformation in USFMS compared to traditional MSW.

For wastewater pipes in general, a minimum slope of 1% is recommended to assist gravity flow. EPA (2000) recommends a 4% slope in capping systems, and Lagerkvist (2003) recommends 2%. To ensure effective drainage and reduce the risk of build-up of pore water pressure in the landfill due to settlement, drainage layers should be installed at intervals of 12 m of waste with a 4% slope.

A possible landfill outline is illustrated in Figure 13.

4.3 Impact on transportation of liquid phases

Impact of USFMS on transportation of liquid phases will depend largely on permeability, in turn affected by porosity, homogeneity, water holding capacity and other factors.

As shown in Figure 11, particle size distribution of USFMS was fairly even in the range between 0.063 mm and 16 mm. A comparison of results obtained with the wet sieve with those of the dry sieve highlighted a significantly higher amount of fines, implying that smaller particles adhere to the surface of larger ones in dried material. These fine particles may be mobilized and redistributed in a landfill and cause a further decrease in permeability by clogging of pores.

It is therefore to be expected that voids between larger particles would be filled prevalently by smaller particles having a very small average pore size. Accordingly, a low permeability of 6.7 x 10^{-9} ± 2.5 x 10^{-9} m/s at 700 kPa, bordering on the permeability of liner materials, was observed. This is comparable to findings reported in previous studies for compressed food wastes (Lagerkvist & Cossu, 2005). A higher permeability would be obtained on application of lower normal pressure; to this regard, Beaven et al (2008) observed a range of 10^{-8}-10^{-6} m/s at 600 kPa for compressed MSW. It should also be kept in mind that the hydraulic conductivity observed here was vertical and counter-directed to the applied pressure. Flow resistance in the horizontal direction was not observed in our test, but it is typically an order of magnitude lower that observed vertically. Hudson et al (2009) reported a hydraulic conductivity 5-7-fold higher in a horizontal compared to vertical direction at an applied stress of 40 kPa, and almost 10-fold higher when the applied stress was increased to 300 – 500 kPa.

Thus, it is likely that to a considerable extent both gas and leachate move laterally, and the inclusion of vertical drains may be required.

The high proportion of fines in USFMS also affects the water-holding capacity of the material (field capacity). The large surface area attracts a boundary layer of stagnant water that does not drain during compression. A porosity of 52% was observed after three months at 700 kPa. Both the latter value and field capacity of 54+0.2% measured several days after compaction, were fairly high compared to values reported in the majority of landfill studies.

FIGURE 12: Photos from dry sieving. Above: to the left waste > 8 mm, to the right waste > 2mm. Below: to the left waste > 0.5 mm, to the right waste > 0.063 mm.

No statistical differences were detected in mean values of permeability in the various compression vessels. USFMS therefore may be considered relatively homogenous on a macro-scale as a result of pre-treatment – as also highlighted by the sieving tests. Thus, future local variations in permeability may vary to a lesser degree than waste which has not been mechanically treated and screened prior to landfilling. Based on the low permeability and high field capacity, a water and gas pressure build up may occur at the site of USFMS deposition. The issue of how to avoid stability problems caused by this phenomenon is further discussed in the following section.

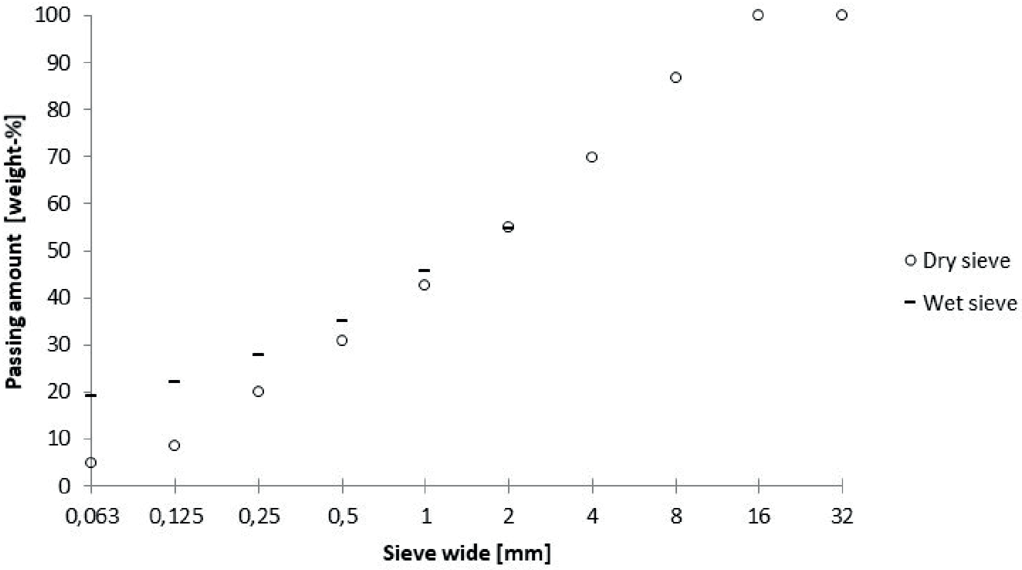

FIGURE 11: Particle size distribution based on mean values from both dry- and wet sieving.

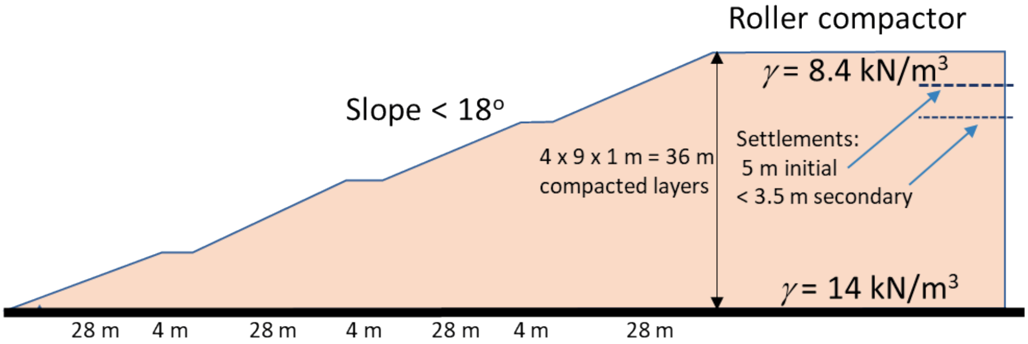

FIGURE 13: Proposed principle outline of a landfill with 36 compacted layers of 1 m with approximate initial and secondary settlements. Slopes should feature horizontal terraces every 5 to 10 meters to increase stability.

4.4 Influence on operation, maintenance, installations and treatment systems

4.4.1 Gas pressure

Despite the low gas formation potential, during landfill design the ability of high gas pressure yielded by low permeability and high field capacity of USFMS should be accounted for. The full extent of this scenario is not covered by this study, but stability issues will likely be manifested if adequate precautions (e.g. gas wells, combined drainage layer for gas and water etc.) are not taken. More than one drainage layer should be applied during landfill construction to contrast the build-up of high gas and water pressures.

4.4.2 Treatment of leachate

Treatment systems for landfill leachate are based upon different techniques for separation, stabilization and/or degradation of contaminants. This can be achieved through biological, chemical or mechanical methods, depending on types of contaminants and time needed for treatment.

Due to the low permeability of USFMS, an L/S ratio of 10 will not be reached within a time limit of, in order of magnitude, 2500 years (assuming maximum infiltration rate 50 l/m^2/year according to Swedish legislation for non-hazardous landfills). It is therefore of fundamental importance to identify appropriate treatment based on the mobility of contaminants detected in the leachate at L/S ratio 0.6 (reflecting natural water content in USFMS and actual L/S ratio at a hypothetical discharge today).

Permeability thus obtained corresponds to 200 mm/year. This implies that in order to meet legislation requirements of 50mm/year, approx. 75% (150 mm/year) of water infiltrating the landfill should be managed directly by the bottom liner, and that the resulting USFMS leachate should undergo treatment due to the presence of organic material, nitrogen and zinc (based on permeability and density at 700 kPa). Calculations reveal an amount of organic content to be reduced corresponding to 0.4 kg/ton USFMS per year. With regard to nitrogen and zinc, the corresponding reduction demand is 0.1 kg/ton USFMS per year, and 1 g/ton USFMS per year, respectively. It should be noted that these values represent the reduction demands at minimum load of discharged contaminants. An estimation of time needed for treatment can be achieved when comparing results from the column tests with results from agitation batch tests. For a pH-dependent parameter such as zinc, it will take up to about 160 years before the total leachable content has leached out. In reality though, pH will probably be close to neutral when degradation starts and the redox potential is reduced. Therefore, the discharge of zinc may be lower than shown.

For factors dependent on biology, such as DOC and nitrogen, time needed for treatment should be estimated on the basis of additional assumptions (time period for agitation batch tests to prevent onset of total degradation). Indeed, values obtained for potential gas formation (60 m^3 CH_4/t VS, corresponding to 2.7 kg CH_4/t) indicate that degradation will occur in the landfill. Based on maximum gas formation rate, organic material and nitrogen may need to undergo treatment over several decades.

As shown above, low permeability will result in leaching of contaminants from USFMS over a very lengthy time frame. To avoid the need to ensure operations and maintenance over such a long period, the use of natural passive treatment, such as wetlands, is recommended by the authors. A constructed wetland is often designed to treat leachate and promote the degradation of organic material or transformation of nitrogen. Nitrogen removal may be achieved by a series of methods aimed at promoting nitrification and denitrification. Mobility of any metals present (in this case zinc) will decrease through oxidization and precipitation. A wetland is frequently represented by aerobic pretreatment (e.g. aerated lagoon), that contributes towards reducing the organic load (in terms of BOD). In the case of an aerated lagoon, sludge treatment will also be required. Equalization ponds are also used to manage seasonal fluctuations of nitrogen reduction.

4.4.3 Compaction Properties

Bearing capacity is affected by water content, as shown during the proctor compaction test at TS 48%. Compaction issues have been experienced using MBP-waste (Mechanical and biological pretreated waste) in Germany (Ohlsson et al. (2002), that was solved through the use of a vibrating sheepfoot roller (Ohlsson et al. (2002). Proctor compaction test at TS 64% showed good compaction properties.

During subsampling, the angle of repose was measured. USFMS was frozen and behaved as a granular material with an angle of repose of 31.7°, similar to a moraine. In the laboratory, the angle of repose was also studied on a material compressed to 700 kPa. When digging in the material, even a vertical angle remained stable.

4.4.4 Indication for design

A simplified sketch of a landfill is shown in Figure 13. To meet stability requirements, slopes should also feature horizontal terraces every 5 to 10 m. Terraces may also be used during road construction. The important issue of drainage and associated risk of clogging should also be addressed.

Infiltration during heavy rainfall may result in a risk of stability issues:

4.5 Aftercare and post-operational implications

In comparison to a landfill containing mixed residual wastes, a USFMS monofill will provide more uniform properties. Thus, the risk of differential settlements and subsequent damage to the final cover will likely be mitigated.

Low permeability of the materials and slow bioconversion processes will imply a need to manage leachate contamination for several decades following covering and completion.

Accordingly, post-treatments such as aeration may also be hampered for the same reason, whilst low gas generation should enhance the feasibility of passive treatments.

5. CONCLUSIONS

The following answers are given to the questions in the introduction.

How high is the pollution potential of the under sieve fraction of mechanical sorting, USFMS, in a landfill environment?

- Is it biologically stable? – Yes, it is almost stable, although featuring a high carbon content (total oxidable carbon, TOC ≈ 150 g/kg total solids)
- How might it contribute to landfill emissions? - Emission potential is present in both gas and liquid phases; however, given the slow degradation rate and low permeability of the material, only low intensity emissions are expected, which may promote the use of passive treatment methods. The exception would be run-off from surfaces on which shredding and intermediate material storage is carried out
- What kind of pre-treatment could be applied? – Additional separation of degradable organics, washing or hydrothermal carbonation could be considered. However, additional costs may rise beyond the impact.

How should the properties of USFMS be considered in landfill design and operation?

- Impact on stability (slope stability, settlement etc.) - High shear strength combined with a lower compression ratio than standard MSW and a well-graded material will contribute towards ensuring good stability and decreasing risk of slides and settlements. A low organic content implies a limited degree of degradation, thus reducing the risk of differential settlements. However, the most critical mechanical property is represented by low permeability, which may result in stability issues in the presence of a build-up of pore water pressure due to ineffective drainage.
- Impact on transport of liquid phases - Gas and leachate may move laterally, which may require for the inclusion of vertical drains.
- Influence on installations and treatment systems. – Drainage layers for gas and water should be provided.
- Implications for aftercare and post-operation usage – There is little risk of differential settlements and damage to the final cover but there may be a need to manage leachate contamination for several decades following covering and completion.

In addition to the answers to the questions, the following conclusions could be reached:

- The total content of several metals exceeded guideline values issued by the Swedish Environmental Protection Agency for less sensitive land use. Thus, in the present study, mechanically treated wastes were not indicated for use as construction material for off-site applications. Due to its high water retaining capacity, it could be used in a protective layer above a liner system for supply of water to plants and demotivating deeper root penetration.

It is likely that the amount of similar waste will increase as more processing and material recovery is undertaken. It does not fit well into existing waste management systems, it does not burn well and it contains more organic carbon than what present limit values prescribe for landfilling. Landfilling is still the realistic option, but it must be shown that it can be done with rather limited environmental impacts, which our study indicates.

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EMPLOYMENT EFFECTS OF DIFFERENT MUNICIPAL WASTE TREATMENT SYSTEMS BASED ON DATA FROM AUSTRIA

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ABSTRACT

Although waste management has long been an important economic sector, its employment effects have so far been neglected in economic calculations. Recently, employment effects have been addressed in the European Green Deal or the Climate Neutrality Strategy. A scientific calculation is, however, still missing.
To close this gap, our paper develops a set of alternative models as to residual waste treatment and investigates their potential to produce direct employment effects. The models include employment required for treatment plants plus employees needed in collection and transports. The models range from basic to sophisticated, from unregulated landfilling via 100 % waste incineration to mechanical-biological treatment to improved separate collection and advanced recycling. Data for devising these models were derived from a secondary analysis (e.g. residual waste analyses of the Austrian provinces) and from expert interviews. Although the models are simplified and build on generic waste management systems, they allow for a rough estimation of employment effects: The more sophisticated the waste management system, the more employees are necessary. In all models, but especially in improved separate collection, collection accounts for a significant part of the additional employment. The models show an increase in employment many times over with the improved separate collection model compared to the other models. Although our results can only be scaled up to European Union level in a limited way, we can state that shifting from less sophisticated waste management models to more desirable ones can involve an increase in employment of at least some 40.000 jobs.

1. INTRODUCTION

Waste management is an important sector of the economy and has over the last 50 years changed significantly in Europe. Many countries have moved from a rudimentary stage of “disregarding waste” to a sophisticated stage of “industrial material cycles” (Klampfl-Pernold & Gelbmann 2006). The last stage involves comprehensive recycling and reuse of the material contained in waste and is reinforced by the objectives of the Circular Economy (EC 2015), as well as the European Green Deal (EC 2019) and the Climate Neutrality Strategy 2050 (EC 2018). Recycling, however, requires a more elaborate collection system and a more intensive treatment of waste - and this increases the number of employees involved in the activities. Although waste enterprises might not appreciate this fact, it could be a contribution to job creation within the European Union (EU).

The employment effects in waste management have so far hardly been considered. Studies commissioned by the European Commission (EC) provide values on the increase in direct jobs scenarios of increasing recycling (Hogg, Vergunst, Elliott, Elliott, & Corbin 2016; Gibbs et al. 2014b; Hogg et al. 2015). However, Gibbs et al. (2014a) warn of the inaccuracy of the data they use, as these originate from old, intransparent literature. Moreover, these studies do not describe the method of calculation, and it was not possible to gain more information here.

Anyway, by meeting the circular economy targets the European Commission aims at an increase of over 170,000 direct jobs in the waste management sector by 2030 (EC, 2015). These calculations and figures are criticized as fuzzy by Tamma & Hervey (2018).

For Austria, our country of focus, employment figures in waste management are also sparse and imprecise: To date, employment figures in the waste sector have been compiled on the basis of various statistics and rough estimates. Weingärtler (2009) assumed an approximate number of 27,000 employees in waste management to be

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appropriate for the year 2005. The current Federal Waste Management Plan 2017 estimates the number of employees in the waste industry around 40,000 (BMNT, 2017). In both cases, there is no information available on how these estimates were retrieved.

To fill this gap at least partially, our paper takes a closer look at the Austrian mixed municipal ("residual") waste sub-sector. The primary aim is to show the personnel potential in the area of residual waste treatment in Austria. Through an orientation towards the circular economy - by means of improvements already at the collection stage - it is shown that this also increases the number of jobs. Thus, jobs can be generated in addition to the conservation of resources.

The primary focus is on how the various recovery or disposal methods affect direct employment. Hence, we investigate the following hypothesis:

The more differentiated the collection and treatment system of municipal waste, the higher the required number of employees and thus of jobs provided.

On this hypothesis we build our central research question:

- What is the direct employment potential in the area of residual waste treatment?
- split into several sub-questions:
- What employment level do the individual models require?
- What influence does the collection of residual waste or separately collected waste materials have on the employment effect?
- By what factor does the employment level of the individual models change compared to the basic model?

To test this assumption and answer the research questions, we model and compare different waste management models. To this end, the individual treatment steps or plants are assigned employment coefficients that we derive from data generated by means of data triangulation. By the aid of the coefficients and based on a normalized system input of 100,000 t per year, we calculate the respective accumulated employment effect of each model and compare the results. The rather idealized and simplified models give an estimate of the extent of the number of jobs created by a shift from one model to the other.

Within the framework of this research, we consider the following models for which we derive distinct changes in employment for collection and treatment:

- model 1: Unregulated landfilling [UL]
- model 2: 100 % waste incineration [WI]
- model 3: Mechanical-biological treatment (MBT)
  - model 3.1: MBT + incineration [MI]
  - model 3.2: MBT + incineration + cement plant [MIC]
  - model 3.3: MBT + cement plant [MC]
  - model 3.4: MBT with dry stabilisation + incineration + cement plant [MDIC]
- model 4: Improved separate collection
  - model 4.a: Improved separate collection according to the Benchmark-Study (Brunner et al. 2015) [ISCa]
  - model 4.b: Improved separate collection according to Best-Practice Vorarlberg [ISCb]

The current Austrian situation is reflected in model 2 with 100 percent incineration, which we use as a reference for comparing the other models. In improved separate collection (models 4.a and 4.b), a part of the residual waste stream is shifted to the recyclables collection. The goal here is to increase the quality and quantity of the material going into recycling (Kranzinger, Schopf, Pomberger, & Punesch, 2017). We supposed from the outset that the model of improved separate collection can help to multiply the number of direct jobs by far in comparison to the other models. We had, however, only vague ideas on the effect on indirect employment effects and the effects induced by a shift between our models. Thus, we invited the Austrian Institute of Economic Research (WIFO, Meyer & Sommer 2019) to calculate indirect and induced employment effects and will present their results in short.

The structure of the paper is as follows: After the introductory section we present the methods of data collection including a short excursus to reuse. Next, we describe deriving and compiling the individual research models. The next section comprises the comparison of our models and their employment effects, amended by the results of the research on indirect and induced employment effects by the WIFO. The discussion of the results follows en suite and finally, we examine the results critically, determine limitations and the need for further research.

2. METHODS OF DATA COLLECTION

We started our research by collecting and calculating employment figures and transfer coefficients. This requires an extensive survey of waste collection and treatment in Austria as provided by the Austrian Federal Waste Management Plan (BMNT, 2017). Here, the average generation and composition of residual waste are calculated from the residual waste analyses of the individual federal provinces. However, data on employment, residual waste analyses or transfer coefficients of facilities are hardly available.

To generate meaningful transfer coefficients and employment related data for the models, we chose an empirical three-step method (Hug, 2015). We applied a mixed methods approach equally making use of qualitative and quantitative data collection methods and used data triangulation to increase the validity of the output (Döring and Bortz, 2016; Flick, 2011).

We started gathering the data by secondary research on the current state of research and discovered that very few data are available in literature. Accordingly, we investigated a large number of plant types by means of meticulously conducted interviews.

After establishing an initial contact by telephone, most of the interviews were conducted by telephone or by email. The survey guide consisted of unstructured open questions adapted to the respective locations and plants. The aim was to obtain figures for calculating the employment effect for an input of 100,000 t of waste per year. Quite often we only received approximate figures due to confidentiality - a reduction in quality that we accepted in order to obtain data at all.

In total, we got data from 105 plant operators with sites from all provinces of Austria as well as three foreign sites were surveyed on the following treatment steps:

- waste collection and waste transport,
- landfill,
- waste incineration,
- mechanical-biological treatment (MBT),
- substitute fuel production (SFP),
- use of substitute fuels in cement plants,
- sorting of light weight packaging,
- plastics recycling,
- composting,
- wastepaper sorting,
- waste glass processing,
- scrap metal processing.

In the following chapters we will present how the data were compiled. As there is a huge employment effect of reuse not included in our models (due to the small share of reuse) we at least mention some of the potential contributions.

2.1 Residual Waste Data

The focus is on mixed municipal waste from Austria, for which there are clear differences in the per capita generation across Austria, ranging from 83 kg in Vorarlberg to 289 kg in Vienna (BMNT, 2017), as listed in Table 1.

The data from the Federal Waste Management Plan 2017, which contains the inventory for the year 2015, was used as the starting point for the waste volume. In addition, each federal province has records on the volume of residual waste and carried out residual waste analyses. Table 2 shows the analyses of the federal states used for the calculation.

We freed the residual waste analyses from any inconsistencies in accordance with the guidelines for the preparation of residual waste analyses (BMLFUW, 2017) and could then divide the residual into its main fractions (Figure 1).

Gaining data on improved separate collection (for models 4.a and 4.b) has to start as early as in households with the separation into the individual collection fractions. To this end, we chose two different approaches: On the one hand and in accordance with the "Benchmarking für die österreichische Abfallwirtschaft" (Brunner et al., 2015) we applied the assumption of a 50% reduction from the baseline; on the other hand, we used the average residual waste volume of Vorarlberg as a best practice example (ÖÖI and TBH, 2012). Table 3 shows the comparison between them and the Austrian average.

TABLE 1: Mixed municipal waste – generation by federal states in Austria

Federal states	**Generation** [t]	**Generation** [kg/capita]
Burgenland	35,000	121
Carinthia	97,300	174
Lower Austria	230,900	141
Upper Austria	167,300	116
Salzburg	93,300	172
Styria	155,900	127
Tirol	96,600	132
Vorarlberg	31,800	83
Vienna	523,500	289
Austria	1,431,600	166

Table based on BMNT, 2017, p. 51

TABLE 2: Selected residual waste analyses from the federal states of Austria with study year and source.

Federal states	**Year**	**Source**
Burgenland	2010	TB Hauer, 2010
Carinthia	2011	Amt der Kärntner Landesregierung, 2012
Lower Austria	2011	Amt der Niederösterreichischen Landesregierung, 2011
Upper Austria	2013	BMLFUW, 2015
Salzburg	2007	Amt der Niederösterreichischen Landesregierung, 2011
Styria	2014	BMNT, 2017
Tirol	2009	FHAanalytik und TB Hauer, 2010
Vorarlberg	2012	ÖÖI und TBH, 2012
Vienna	2016	Stadt Wien, 2016

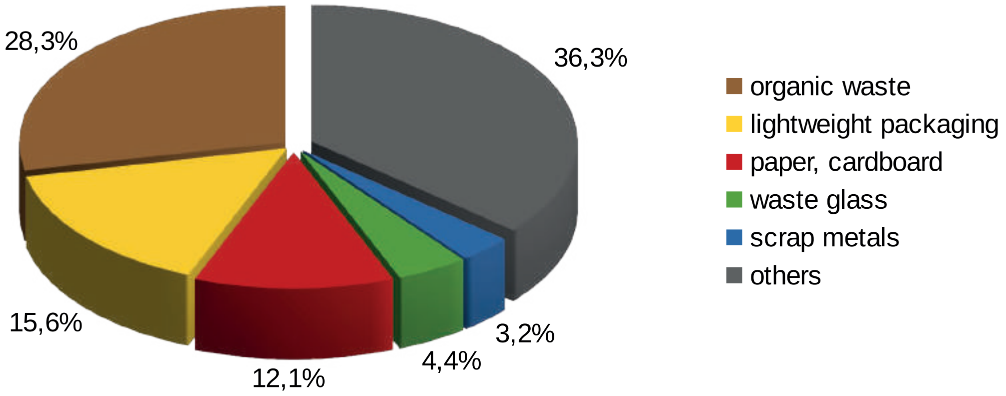

FIGURE 1: Splitting of fractions into main groups and percent per mass.

TABLE 3: Comparison of the collection fractions - Austrian average with benchmark study and best practice Vorarlberg

Collection system	Austrian average	Benchmark Study	Best Practice Vorarlberg
collection of residual waste	100.0%	66.7%	54.7%
collection of lightweight packaging	0.0%	8.1%	10.4%
collection of organic waste	0.0%	14.6%	20.0%
collection of wastepaper	0.0%	7.0%	9.9%
collection of waste glass	0.0%	2.3%	3.2%
collection of scrap metals	0.0%	1.3%	1.8%
total	**100.0%**	**100.0%**	**100.0%**

2.2 Waste Collection and Transport

The first step to be considered is waste collection, on which data proved to be difficult to obtain. This was due to a lack of literature and the lacking willingness and ability of companies to provide data. Furthermore, a plethora of influencing factors like container size, the ratio of drivers and loaders, collection intervals or the collection logic made a standardization impossible, as they can differ even at the levels of individual municipalities.

Accordingly, we assumed that the collection of residual waste is carried out in a pick-up (collection) system. A pick-up system is furthermore also assumed for the models of improved separate collection, the collection of lightweight packaging, biogenic waste and wastepaper. Only for waste glass and scrap metals we adopted bring-it-yourself systems, with households bringing the pre-separated material to waste collection points. The respective employment effect calculated for each fraction is shown in Table 4.

As for transport between the individual plants, we could not determine secondary references and information from the interviews on neither the related employment nor the transport routes, only some isolated information is available as to environmental impacts (Frischenschlager et al. 2010). We discussed the values with an expert and had to accept a set of simplifying assumptions.

We assumed that all transports are carried out by truck, although in reality, the distances are also covered by rail. We reduced the load weight to three sizes and divided the estimated transport routes into 50 km intervals. Finally, we reduced the loading and unloading times as well as the regional and supraregional travel times to half-hourly intervals (Altendorfer, 2018). Table A.1 in the appendix contains the values for the loading weight, the transport routes and the employment per 100,000 t for the transport routes used.

TABLE 4: Selected employment effects for the collection in the models.

Collection system	Employees
per 100,000 t	100.0%
collection of residual waste	90.0
collection of lightweight packaging	200.0
collection of organic waste	90.0
collection of wastepaper	70.0
collection of waste glass	24.0
collection of scrap metals	200.0

2.3 Waste Treatment Plant Data

Gaining data on the employment of waste treatment plants also proved to be difficult. Literature only yielded information on sub-areas ranging from the negative impacts of unregulated landfilling to alternatives like waste incineration, co-incineration, MBT or substitute fuel production, but not on the employment required for these treatment methods. Reprocessing plants nearly go unnoticed in literature, except for the economic efficiency of recycling or the effects on the environment. An overview of waste treatment facilities in Austria is provided in the Federal Waste Management Plan, and also the homepage of the Association of Austrian Waste Management Companies (VOEB, 2017) offers an extensive list of members, which is useful for identifying relevant facilities. Our own survey also proved difficult as most of the interviewees could not reveal confidential data or lacked the time to compile the data. Again, we consulted with experts in the field and finally, with the information content still limited, we were able to calculate plausible values for the impact of different waste management models on employment. Into our models, we again integrated only direct jobs. These are summarized in Table 5. A detailed elaboration can be found in Altendorfer (2018).

2.4 The Employment Approach

In this paper we relate to the direct employment effects as in additional direct jobs of different waste management models. This is to mean that we look at full-time equivalents of people working in the waste residual waste related system, in transportation, sorting, preprocessing and treatment itself, but not in the downstream recycling process itself. The respective system boundaries will be explained in detail where necessary. Not part of our study, but of a subsequent study by the WIFO (Meyer & Sommer 2019) were the indirect and induced effects on employment.

“Indirect effects” or upstream effects include the production of all intermediate inputs necessary for the direct activities. The corresponding data are abstracted in a so-called input-output matrix as intermediate inputs. “Induced effects” relate to the reactions of private households concerning consumption that are to be expected due to changed incomes and are calculated on the basis of an average propensity to consume (Meyer & Sommer, 2019).

2.5 Implications of Reuse

Although the reuse sector is not included in the mod-

TABLE 5: Selected employment effects for the treatment facilities in the models

Abbreviation	**Plant designation**	**Employees** per 100,000 t
Cem	waste co-incineration plant with rotary kiln in the cement plant	4.0
Comp	composting plant	35.0
Gl-S	waste glass sorting plant	10.0
Gl-T	waste glass transfer station	3.0
L-Shr	large shredder plant	40.0
LF-D	domestic waste landfill	5.0
LF-M	mass waste landfill	10.0
LF-R	residual waste landfill	10.0
LWP-S	lightweight packaging sorting plant	200.0
MBT	MBT plant	18.0
MBT-dry	MBT plant with dry stabilisation	18.0
Met-T	scrap metal transfer station	3.0
P-Shr	post shredder plant	150.0
PAP-M	wastepaper sorting plant	25.0
Pla-R	plastic recycling plant	200.0
SFP-P	substitute fuel production plant	16.0
WI-F	waste incineration plant with fluidised bed combustion	22.0
WI-G	waste incineration plant with grate firing	24.0

els as it for a bigger part occurs outside the waste regime, reuse offers an enormous potential for jobs in the area of waste avoidance. What used to be a common practice of repairing things and putting them back into operation has almost completely disappeared in recent times. However, to some extent there has been a paradigm shift, and in recent years the quantities of goods collected for preparation for reuse have increased and are now at about 0.1% of the total Austrian volume of municipal waste from households and similar establishments. An increase to 1% is a realistic assumption, as the example in Flanders shows (Neitsch & Wagner, 2017). Even though the amount of waste diverted from the waste stream is very small, it has a large impact on employment effects. Processing, sorting, cleaning and refurbishing are very labor-intensive processes requiring a high amount of manpower which not necessarily has to be well-skilled. This leads to another special feature of the reuse sector: Reuse companies feature a unique personnel structure by providing transit training jobs for people furthest from the labor market, permanent jobs for people with special needs and opportunities for voluntary work (Neitsch & Wagner, 2017). According to them, the potential number of jobs in the reuse sector ranges from 4,000 to 7,500 per 100,000 t of reuse material.

3. DERIVING THE MODELS

From the data compiled we derived our waste management models that depict treatment paths in a simplified way. These do not claim to represent reality exactly but show a sequence of the waste stream concentrated on the fundamentals. Due to the complexity and the large regional differences, a real representation of waste management is hardly possible. But although in reality mostly combinations of several of the ideal-typical models will be found and small regional deviations are possible, we are convinced that the material flows and personnel coefficients meet the actual situation in Austria very well.

We devised four different models for the comparison of waste management systems. The analysis starts with collection and ends either with landfilling, in facilities where the end of waste is reached or before the recycling process takes place. In each model we trace an input of 100,000 t through each step. In models 1 to 3, this amount is directly distributed to the various treatment plants. Only in model 4, a part of the residual waste already attains the recyclables collection at the waste producer, from where it is sent to the different recycling processes. Collection is considered a subsystem that is required throughout the whole process but is excluded as a basic collection in the comparison of the models, as this procedure is indispensable for each model. Furthermore, the employment effects are broken down into people working in the facilities and transport. On this basis, we calculate the respective required employment for each treatment step including collection and transport and subsequently sum up the number of employees for the entire model.

3.1 Model 1: Unregulated Landfilling

In this model, the entire volume is brought untreated to a nearby unregulated domestic waste landfill (see Figure 2), a method that has been prohibited in Austria since the Landfill Ordinance of 2004 where a treatment prior to landfilling is required in order to reduce the amount of total organic carbon contained in the waste. We still chose to include this model, as it is still common practice in European countries such as the Czech Republic, Bulgaria or Romania.

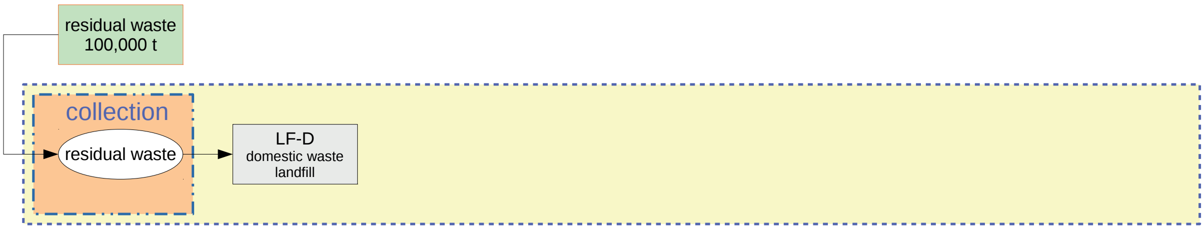

FIGURE 2: Model 1 - Unregulated landfilling.

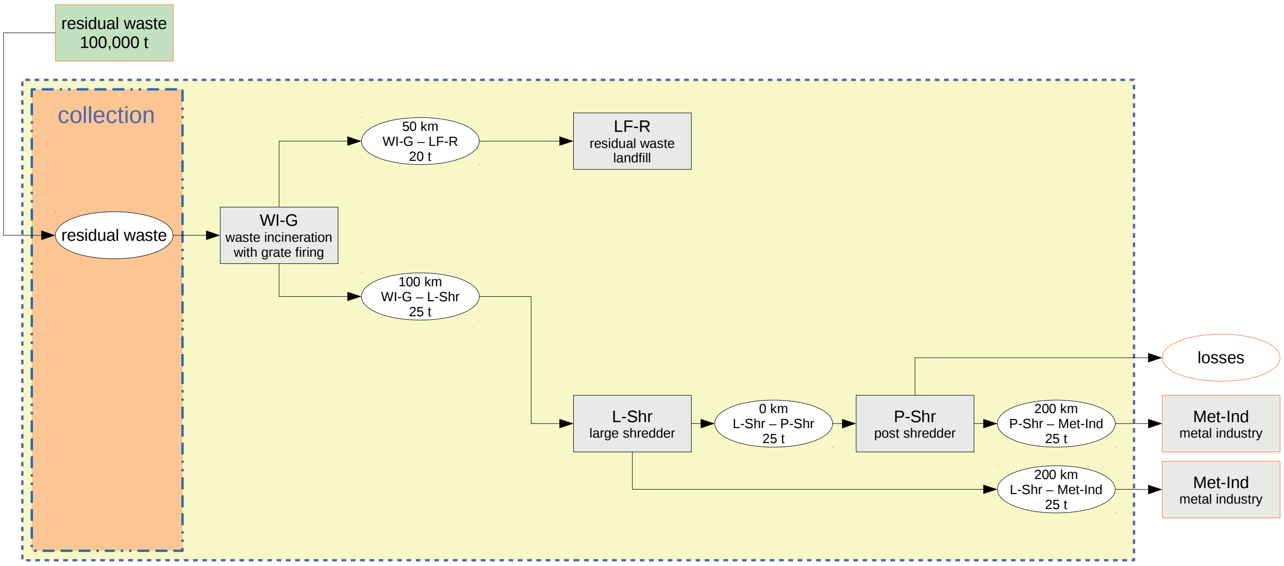

FIGURE 3: Model 2 - 100 % waste incineration.

3.2 Model 2: 100% Waste Incineration

Contrary to model 1, in model 2 all residual waste is directly brought to a waste incineration plant with grate firing. These plants are capable of directly incinerating the untreated waste. Residues of incineration go to a residual waste landfill. These residues (ash, dust, ...) still contain a small amount of metals, which are separated from the landfill fraction and fed into the metal recycling process (Figure 3).

3.3 Model 3: MBT

In model 3, the collected residual waste is first brought to a MBT plant (MBT). Depending on the type of plant, it can produce different qualities and quantities of high-calorific value fractions and landfill fractions. Accordingly, we divided model 3 into four sub-models:

- model 3.1 MBT + incineration
- model 3.2 MBT + incineration + cement plant
- model 3.3 MBT + cement plant
- model 3.4 MBT with dry stabilization + incineration + cement plant

Models 3.1 to 3.3 use the identical type of MBT. However, the output is utilized in different ways. MBT with dry stabilization attempts to produce a maximum proportion of the high-calorific fraction and is included in model 3.4. The next sections describe models 3 in more detail.

3.3.1 Model 3.1: MBT plus Incineration

In model 3.1 the entire medium-caloric fraction and high-calorific fraction (about 60%) from the MBT goes to a waste incineration plant with fluidized bed firing. The residues remaining there are brought to a residual waste landfill. Material which features a calorific value too low for thermal recovery is sent to a mass waste landfill for final disposal. During processing in the MBT, iron scrap can still be separated as a secondary material. To this end, the fraction is sent to a large shredder plant and a subsequent post-shredder plant, where the processing and sorting into metals and non-ferrous metals takes place. Transporting the scrap metal to a metal processing plant to the metal industry is still inside our system boundaries and hence included in our calculation. The employment requirements and the related expenditure in the metal industry, however, are outside of our system boundaries (Figure 4).

3.3.2 Model 3.2: MBT plus Incineration plus Cement Plant

Model 3.2 features one main difference to model 3.1, that is the destination of the high-calorific fraction in the cement industry and the use of the medium-calorific fraction for waste incineration in a fluidized bed incinerator. The residues from the fluidized bed combustion are again sent to a residual waste landfill. For use in the cement industry, the high-calorific fraction is processed in substitute fuel production plants in such a way that it can be added

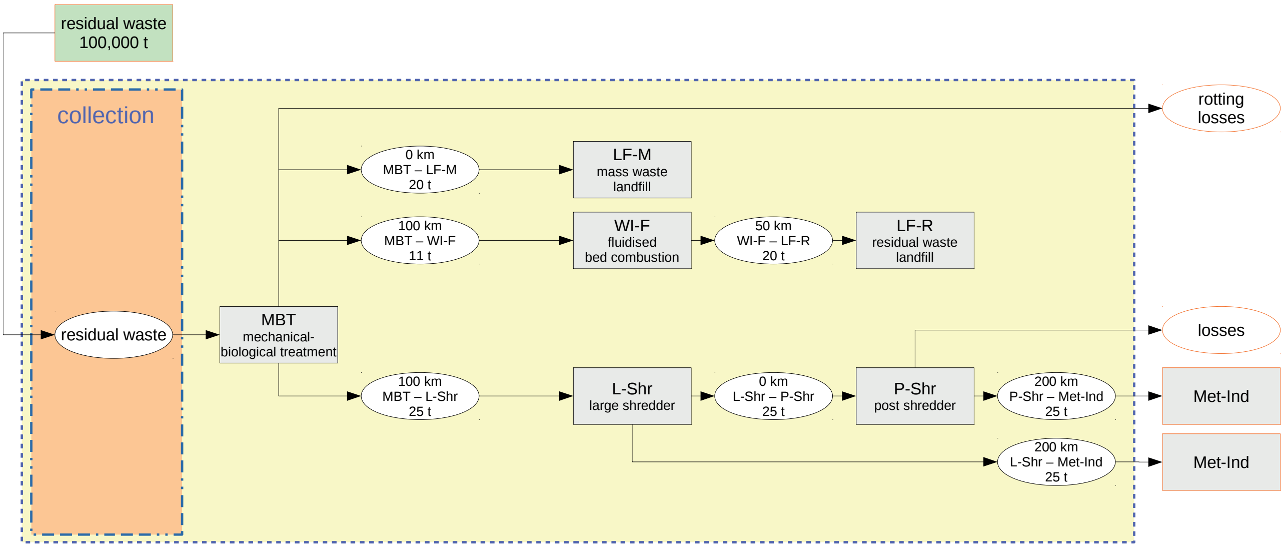

FIGURE 4: Model 3.1 - MBT + incineration.

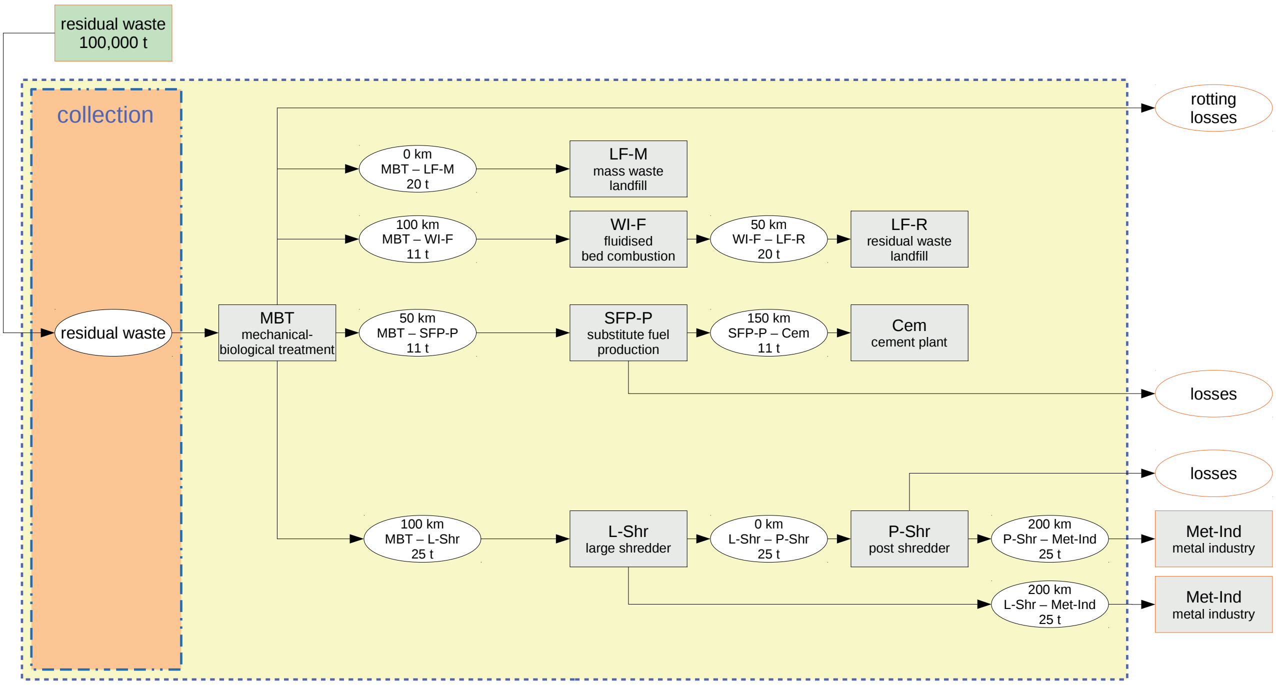

FIGURE 5: Model 3.2 - MBT + incineration + cement plant.

to the rotary kiln as a substitute for fossil fuels. Since no residues remain when waste is incinerated in the cement plants, our calculation ends here. Material which features a calorific value too low for thermal recovery is again sent to a mass waste landfill. As in the previous model, scrap metal is discharged from the MBT and processed in shredder plants for the recycling process in the metal industry (Figure 5).

3.3.3 Model 3.3: MBT plus Cement Plant

In model 3.3 both medium-calorific and high-calorific fractions are fed into the cement production process. The processing of the high-calorific material takes place as described in model 3.2 in a plant for substitute fuel production. The main difference is that only a small amount remains to be landfilled, as the material is caught up in the cement kiln (Figure 6). The landfill fractions from the MBT go to a mass waste landfill as described above and the scrap metals are also treated as in the previous models.

3.3.4 Model 3.4: MBT with Dry Stabilization plus Incineration plus Cement Plant

Model 3.4 involves MBT with dry stabilization. Here, the percentage of rotting losses and the residues for depositing in mass waste landfills can be reduced and a

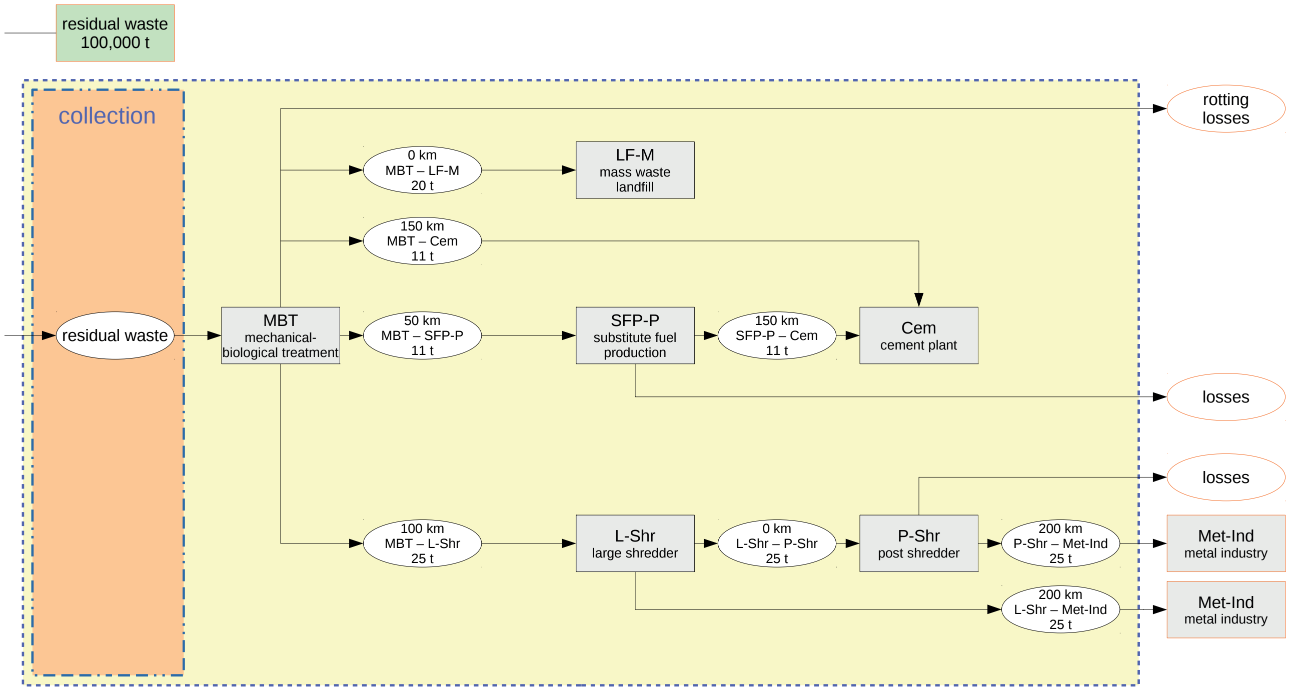

FIGURE 6: Model 3.3 - MBT + cement plant

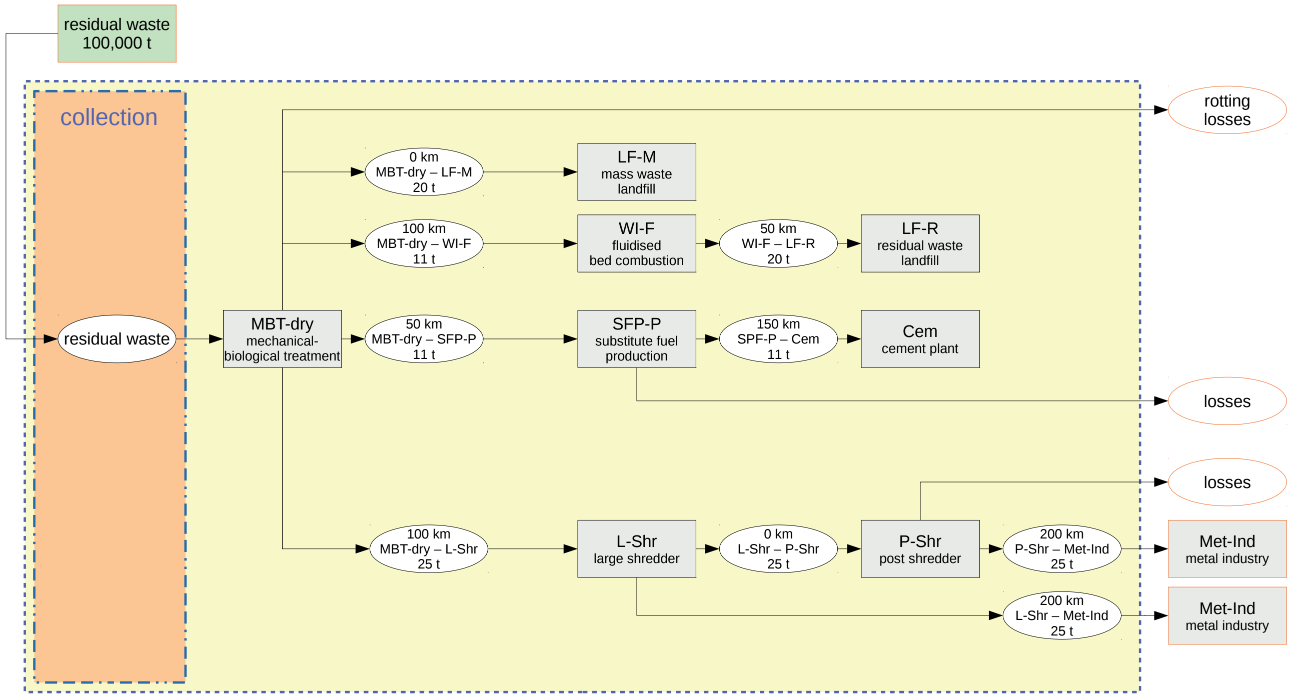

FIGURE 7: Model 3.4 - MBT with dry stabilization + incineration + cement plant.

considerably larger part of a thermally recoverable fraction is produced. Almost 60% of the high-calorific material is transported to plants with fluidized bed combustion (Figure 7), as in model 3.2.

The other part is sent to a substitute fuel production plant for conditioning and then to the cement plants. Again, scrap metal is discharged as in the other MBT models and processed for the metal industry.

3.4 Model 4: Improved Separate Collection

This model stipulates improved separate collection already at the household and hence the collection levels, as in the previous model for an input of 100,000 t. The main difference is the division of the input into further waste streams in addition to the residual waste fraction. Two different sub-models are presented for this purpose.

- Model 4.a: Improved separate collection according to benchmark study (Brunner et al. 2015)
- Model 4.b: Improved separate collection according to Best Practice Vorarlberg (ÖÖI & TBH 2012)

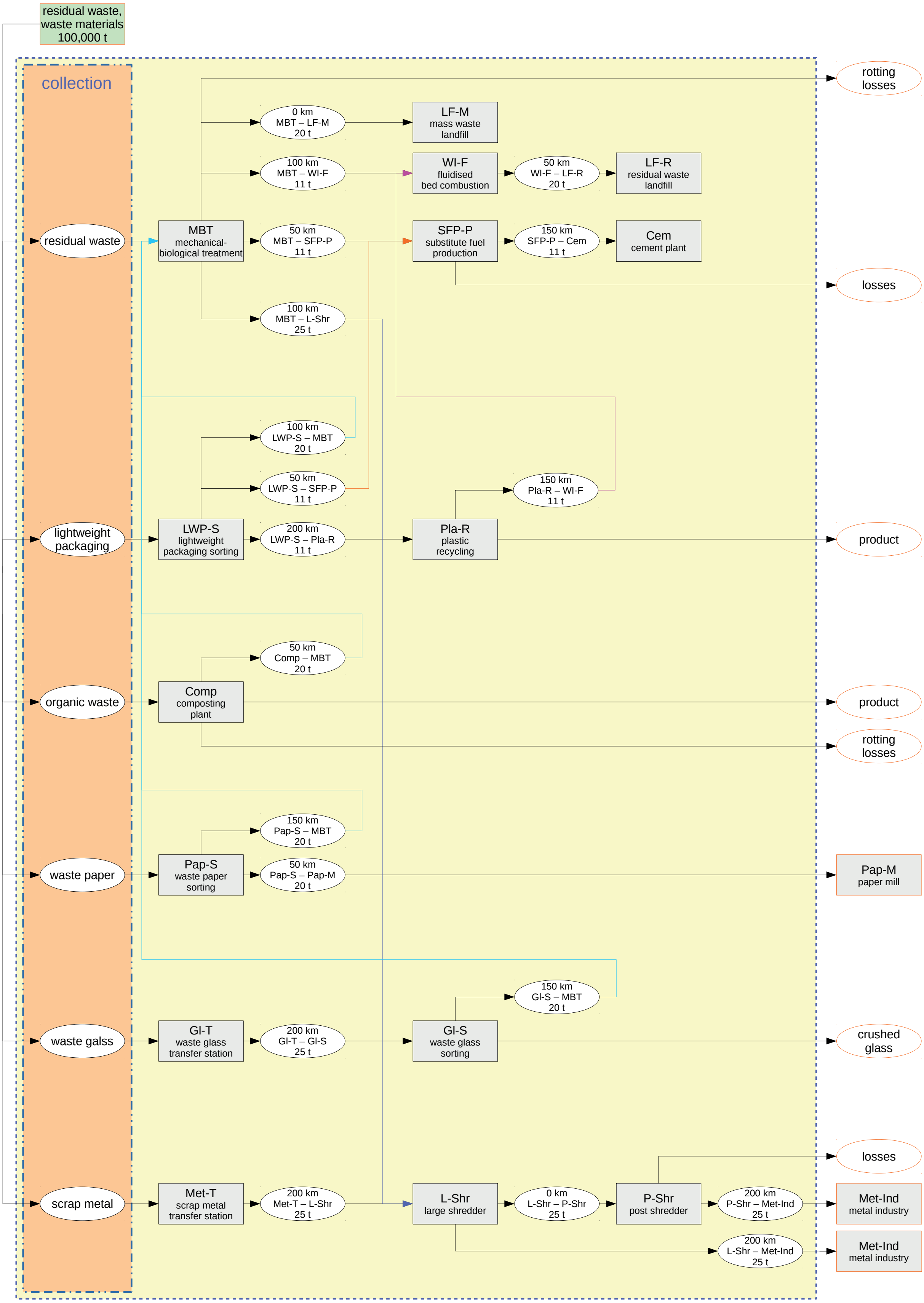

FIGURE 8: Model 4 - Improved separate collection.

Collection is basically provided in a pick-up system, only the fractions "wastepaper" and "waste metals" are collected in a bring-it-yourself system at waste collection points. By now considering six different material flows, this model features the greatest complexity (Figure 8). Similar to the collection and the different treatment stages for residual waste in the other models, the models require even more simplifications for the other waste fractions. We will describe these in the following. It has to be mentioned that labor in households (e.g. for separating) remains uncompensated for and is thus not included in our models.

- Residual waste: The processing of the residual waste equals model 3.2. After MBT, the low-calorific part goes directly to landfill, the medium-calorific part to fluidized bed incineration and the high-calorific part, after processing in a substitute fuel production plant to the cement industry.
- Lightweight packaging (LWP): The fraction of lightweight packaging separated at household level is collected there in a pick-up system and brought to a sorting plant where recyclable plastic (e.g. PET) is sorted out and taken to plastic recycling plants. Our system boundaries end with the input into the production of plastic granulate or plastic flakes, as the granulate is no longer a waste and most production facilities for recycling plastic are not located in Austria. However, the much bigger part of lightweight packaging cannot be reintroduced into the cycle and thus goes to SFP production. Due to the high-calorific value of plastic, the cement industry is a major customer for this waste product.
- Organic waste: Residual waste contains a significant amount of biogenic waste falsely inserted there. Reducing the amount of misdirected organic waste and providing organic waste bins nationwide can create a significant reduction of residual waste and a large organic waste material flow. This material is processed in composting plants, which produce compost soil in various rotting stages. Since compost constitutes an end of waste, it is thus outside the considered system.
- Wastepaper: Wastepaper collection is one of the oldest and best-known collection services in Austria. After collection, the wastepaper is sent to a sorting plant where large contaminants are sorted out. Pressed into bales, the wastepaper comes to the paper mill where it is fed into the recycling process. The consideration of the wastepaper stream ends with the delivery to the paper mill. Processes such as de-inking are no longer considered within the system boundaries.
- Waste glass: Like wastepaper waste glass has been inserted into recycling processes in Austria for several decades. Collection is mainly provided in a bring-it-yourself system at waste collection points. The collected waste glass is taken to a transfer station and from there transported to the waste glass sorting plants. These sorting plans are usually located directly at or in the vicinity of glass producers. Therefore, the system boundary is drawn directly after the waste glass sorting plant.
- Scrap metals: The collection of scrap metals at waste collection points is very similar to that of waste glass. From the transfer station the material is transported to the metal sorting plants. The process is analogous to the previous models. In the first step, processing takes place in large shredder plants. Ferrous metals are sold to the metal industry as recyclable material. Non-ferrous metals are further processed at the same site to subsequently be sold to the metal industry.
  We will next describe the two models in this respect.

3.4.1 Model 4.a: Improved Separate Collection According to the Benchmark Study

To compile model 4.a, we referred to scenario R2 (resource conservation) in the "Benchmarking für die österreichische Abfallwirtschaft" (Brunner et al. 2015). Table 3 shows a comparison of the individual collection fractions in percent. For model 4.a, the remains in the residual waste collection add up to about 67%. The remaining quantities of waste are dispersed among the collection of light packaging, organic waste, wastepaper, waste glass and waste metals.

3.4.2 Model 4.b: improved separate collection according to Best Practice Vorarlberg

Model 4.b is based on the results of the Vorarlberg residual waste analysis (ÖÖI & TBH 2012). Table 3 compares the distribution of the individual collection streams to the baseline situation and the benchmark model. Due to a good separation of the recoverable waste, only about 55% of the residual waste remains.

4. RESULTS – COMPARISON OF THE MODELS

4.1 Direct Employment Effects

In this section we present the results of our research by comparing and contrasting the employment effects calculated according to the development of our models. Table 6 lists the resulting number of employees required for each model.

Collection is assumed to be comprehensive and thus considered separately by determining and calculating the number of staff required for collection for each individual model. On this basis, we compare the models to each other in order to obtain the ratio of employment between the individual models.

An increase in the number of staff required at the plants can be noticed already in model 2. The additional employment is largely contributed by the waste incineration plant, and a small share of additional employment is due to transport. All models 3.1 to 3.4 involving a MBT plant as the first treatment step require roughly the same number of employees. Only in model 3.3, the number of employees in the plants is significantly lower than in the others, which can be explained by taking into account that running a cement plant on secondary instead of primary fuel does not require many additional staff and there is no landfill fraction remaining. In terms of transport, model 3.4 stands out with a value of 26.9 persons in comparison to models 3.1

to 3.3. In model 3.4, there are lower quantities of residues going to mass waste landfill and lower rotting losses. Also, the share of high-calorific fractions and their processing increases, which subsequently also explains the increase in transports.

The improved separate waste collection in models 4.a and 4.b are obviously more labor-intensive. Interestingly enough, in model 4.b the effort in collection increases only marginally, although almost half of the residual waste is here collected among the recoverables. Neither is there an increase of the employment related to transport in comparison to models 3.1 to 3.4. Visibly more employees, however, are needed in the plants.

At any rate, in all models, collection accounts for a significant part of the additional employment regardless of whether the residual waste goes to landfill or is treated in another facility. The number of staff necessary for residual collection in models 1 to 3 amounts to 90 jobs per 100,000 t of input. Only in the models 4 with improved separate collection the employment intensity increases slightly by 7.4 persons in model 4.a and by 9.3 persons in model 4.b.

Thus, we use the number of 90 persons as a basic feature for the amount of staff required for collection. We therefore detract this number from the total number to scale the results. Only if the collection exceeds 90 persons, the surplus is included in our calculations and has an influence in the comparisons (Table 7 and Figure 9).

This scaling has the greatest influence on model 1

TABLE 6: Employment effects of all models per 100,000 t of input.

	abbreviation	collection	plants	transport	total
model 1	UL	90.0	5.0	-	95.0
model 2	WI	90.0	27.9	3.0	120.9
model 3.1	MI	90.0	35.6	18.3	143.9
model 3.2	MIC	90.0	34.7	21.5	146.2
model 3.3	MC	90.0	26.6	22.8	139.4
model 3.4	MDIC	90.0	38.5	26.9	155.4
model 4.a	ISCa	97.4	56.4	20.2	174.0
model 4.b	ISCb	99.3	62.5	19.5	181.3

TABLE 7: Employment effects - model comparison per 100,000 t of input (without basic collection).

	abbreviation	collection	plants	transport	total
model 1	UL	-	5.0	-	5.0
model 2	WI	-	27.9	3.0	30.9
model 3.1	MI	-	35.6	18.3	53.9
model 3.2	MIC	-	34.7	21.5	56.2
model 3.3	MC	-	26.6	22.8	49.4
model 3.4	MDIC	-	38.5	26.9	65.4
model 4.a	ISCa	7.4	56.4	20.2	84.0
model 4.b	ISCb	9.3	62.5	19.5	91.3

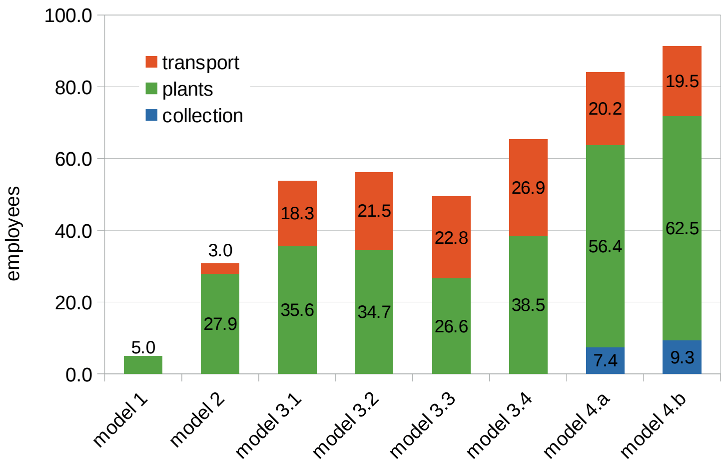

FIGURE 9: Comparison of employment of the models (without basic collection).

TABLE 8: Factor comparison of the models (without basic collection).

		Ratio	
	Abbreviation	Base model 1	Base model 2
model 1	UL	**1.0**	0.2
model 2	WI	6.2	**1.0**
model 3.1	MI	10.8	1.7
model 3.2	MIC	11.2	1.8
model 3.3	MC	9.9	1.6
model 3.4	MDIC	13.1	2.1
model 4.a	ISCa	16.8	2.7
model 4.b	ISCb	18.3	3.0

since here the employment is almost exclusively bound in collection.

To allow for a better comparability, we define two base models on the basis of employment figures calculated above:

- base model 1: Unregulated landfilling
- base model 2: 100 % waste incineration

and normalize each base model to the baseline of 1. According to this frame of reference we calculate to which extent the employment effect for the other models differs (Table 8).

Base Model 1: Unregulated Landfilling

Base model 1 is built on the (for more sophisticated waste management systems) unrealistic assumption that all residual waste goes to landfill without any other treatment. It shows that even with model 2 the employment increases by a factor of more than six. The processing of the residual waste in an MBT (models 3) increases the employment effect compared to base model 1. from 9.9 (model 3.3) to 13.1 (model 3.4), with a significant contribution arising from transport.

With improved separate collection (models 4), the employment effect is about 17 times higher than with unregulated landfilling. There is only a slight increase in the effort required for collection. Compared to models 3.1 to 3.4, the employment figures for transport remain approximately the same or even decrease. The main reasons for the staff changes are up to in the facilities. Figure 10 shows the graphical representation of the individual models compared with model 1 as the baseline.

Base Model 2: 100 % Incineration

Base model 2 is built on the assumption that all residual waste goes directly into incineration, which represents a very popular process for dealing with residual waste in Austria. Of course, the ratios of the individual models among each other remain the same, only the actual numerical values change due to the shift of the base to model 2. When compared with the models applying MBT followed by incineration or co-incineration, employment increases by a factor of 1.6 to 2.1 (Figure 11). This is mainly due to transporting in between the plants. A switch from 100 % waste incineration to improved separate collection and thus also a significant increase in the recycling rates causes an increase in the employment of about three times.

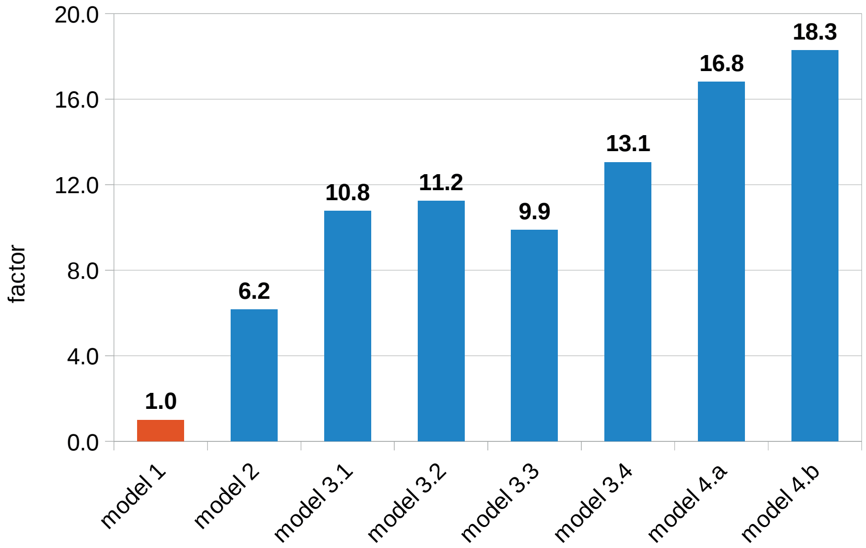

FIGURE 10: Factor comparison of the models (without basic collection): base model 1.

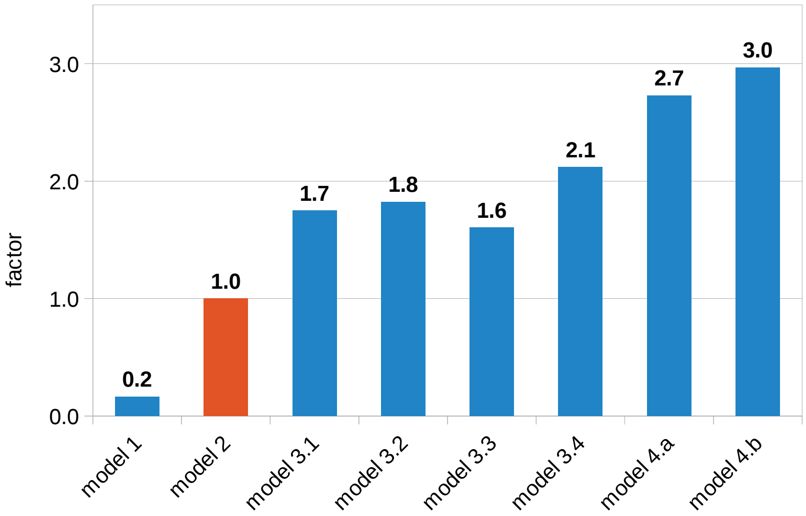

FIGURE 11: Factor comparison of the models (without basic collection): base model 2.

4.2 Indirect and Induced Employment Effects

Paradigm shifts in waste management do not only cause direct employment effects, but also induce indirect ones. The WIFO used our previous calculations and results to include the indirect and induced employment effects (Meyer & Sommer 2019). For their calculation, they adapted the macroeconomic model WIFO.DYNC, a dynamic single-region and multi-sector model based on the revenue and expenditure tables of Statistics Austria. The results are differentiated into their direct, indirect and induced effects and include changes in added value, employment and energy demand (Meyer & Sommer 2019).

As with the direct employment effects a more complex and costly residual waste treatment entails a higher demand for labor (Figure 12). Unlike our comparison of models, the WIFO comparison did not exclude employment required for collection. Thus, the highest result from their calculations is for model 4.b at 317 employees. model 2 features only 197 employees, the lowest number of all models (Meyer & Sommer 2019).

5. DISCUSSION

In order to fulfil the aim of comparing potential employment effects in the field of residual waste treatment we developed a set of four different models, representing four basic stages of evolution in waste management and with the plants involved as the distinctive elements: Unregulated landfilling, 100 % incineration, several combinations of MBT and additional methods and an improved waste sepa-

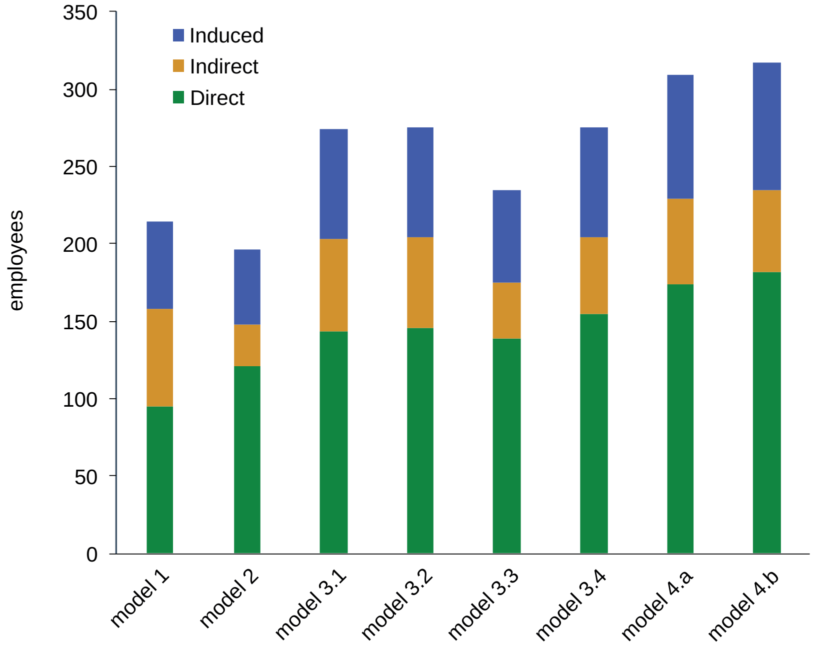

FIGURE 12: Employment effects including direct, indirect and induced effects (Meyer & Sommer 2019).

ration already at consumer level. In this respect, a variety of factors have a considerable influence on employment figures. These include the differences in waste composition, but also the sizes and operational structure of the plants as well as the further use of the output.

In order to compare the models properly, we designed them in a way that each of them ranged from collection and transport to processing in individual plants and was based on a standardized input quantity of 100,000 t of residual waste. The average input was calculated from the residual waste analyses of the various provinces in Austria. In the two models of improved separate collection (models 4.a and 4.b), this input was also divided into different waste- and their respective recovery streams. The selected data and the models are of course simplified and the numerical results should be used with caution. However, they do show the enormous employment potential of sophisticated waste management approaches.

In the first step, the labor for an input of 100,000 t was calculated for each individual model (Table 6). It also shows the employment figures for the collection. From this it can be seen that improved separate collection does not have a significant impact on the employment effect. The significant changes between the models are in the areas of facilities and transport. Since the collection causes approximately the same employment in all models, the fixed value of 90 persons for the “basic collection” is deducted for the factor comparison.

From an Austrian perspective, model 2 serves as a suitable base model. However, model 4.b can be considered a realistic target. It is based on the residual waste volume of Vorarlberg and thus represents an already established situation. An improved waste management system provides an enormous potential for new jobs - compared to 100 % waste incineration (model 2) even a tripling of the direct employment effect (Table 9). Additionally, this approach helps conserve finite resources as significantly more materials are recycled through better processing of the waste materials.

We detect the biggest difference of 86 jobs more between base model 1 and model 4.b, comparison of model 4.b with base model 2 still yields 60 more jobs. The results in detail are shown in Table 10.

Transferred to the real situation in Austria and thus to a basis of more than 1.2 million t of residual and bulky waste directly sent to incineration in 2015 (BMNT 2017), a shift from model 2 to model 4.b would create 730 direct jobs (Table 11), a significantly higher recycling rate and further indirect employment effects. Projecting our results to EU level is of course sketchy, as systems and approaches throughout the EU differ as much as the level of automation and other external drivers. Conceding this restriction, based on 66 million t of residual waste incinerated (Eurostat 2018) up to 40,000 additional jobs could be created. Assuming the 59 million t of waste landfilled in the EU in 2016 (Eurostat 2018) going to unregulated landfilling this would create a potential of up to 91,000 new jobs at EU level.

TABLE 10: Combined factor comparison of the models (without basic collection).

		compare models	
target model	abbreviation	model 1 [UL]	model 2 [WI]
model 4.b	ISCb	86.4	60.6

growth in jobs per 100,000 t of input

TABLE 11: Potential additional jobs in full-time equivalents for Austria and the EU.

	[UL] → [ISCb]	[WI] → [ISCb]	
Austria	-	730	jobs
EU	51,000	40,000	jobs

values rounded

A glance at re-use, which due to a lack of data, is not included in this work, shows that in this area of waste management the greatest employment effect could be skimmed. Hardly noticed, since the goods are only in the waste regime for a short time, waste prevention represents an enormous employment potential and value added, although only a small volume of waste is involved. With 100,000 t, diverting only 1% of goods from the waste stream would mean an increase of about 40-75 jobs.

Creating a realistic representation of all of Austria is impossible, as there are considerable differences at municipal levels and, even more importantly, there is a lack of employment-related figures in waste management. Models suitable for the individual consideration of a region or a municipality, including specifics like seasonal (e.g. tourism) or area-related aspects (e.g. urban or rural), will consist of a combination of the models created here and can be amended by adjusting the values or by taking into account diverse influence factors of residual waste separation, collection, and treatment.

Furthermore, the results and figures on employment effects obtained in this work constitute only one component of a large and very complex system and cannot be used as an exclusive basis for waste management decisions. To gain a holistic view of waste management, other system components must be taken into account in addition to employment effects. Thus, the models can serve as a starting point and be supplemented by additional factors such as:

- environmental impacts,

TABLE 9: Combined factor comparison of the models (without basic collection).

	model 1		model 2		model 3		model 4
	[UL]		[WI]		[MBT]*		[ISC]**
factor	1	:	6	:	11	:	18
factor			1	:	2	:	3

*values rounded / * Mean values from [MI] & [MIC] & [MC] & [MDIC] / ** Mean values from [ISCa] & [ISCb]*

- energy balances,
- CO_2 balances,
- recycling rates or
- economic efficiency.

On this basis it will be possible to compare the sustainability effects, including effects pertaining to employment, of goods made from primary raw materials and those made from secondary raw materials resulting from separate collection and the reprocessing process.

6. CONCLUSIONS AND OUTLOOK

The result of this work shows that there is a great employment potential for the treatment of residual waste. A shift from 100 % waste incineration to improved separate collection allows for a tripling of the number of direct jobs, and thus hundreds of jobs for Austria and tens of thousands of jobs for the EU. Against this background, the EU assumption of creating some 170,000 jobs by 2030 by shifting to a circular economy (EC, 2015) seems very conservative.

This estimate holds good, even as it is one limitation of our work that we had to simplify our models: Especially as to collection modes and the means of transport we had to take simplifying assumptions, which might influence the impact on employment significantly. Here, an adaptation to real-world situations might be useful.

Another limitation is the absence of unified statistical data, as a guideline for standardized residual waste analysis has only been in place since 2017 (BMLFUW, 2017). Finally, we have to take into account as a limitation that an increase in recycling over simpler ways of waste treatment could involve sinking marginal labor intensity in the recycling sector. This diminishes the chance of linearly scaling the model up to a significant rise in recycling rates.

Further room for research would involve a more intense consideration of reuse which can yield a hundredfold employment effect, but also considering bulky waste or different types of collection points for recoverables provide additional starting points. Further research could also extend to energy or CO_2 balances.

It will be important to consider indirect and induced interdependencies as researched by WIFO: More sophisticated waste management systems contribute to job creation and economic performance, but they also cause higher (domestic) energy consumption due to the different forms of treatment. In models 3 and 4, however, this can be partly made up for by generating heat and thus substituting fossil energy. Furthermore, models 4 add to the substitution of primary raw materials and thus to a lower (global) energy consumption and CO_2 emission.

This results in more sophisticated waste management systems contributing to the EU waste hierarchy in a more satisfactory way (including a substantial contribution to a circular economy with lower demand for primary resources). Additional positive environmental effects pertain to avoiding exhaust air and wastewater emissions from landfilling and incineration (Meyer & Sommer 2019).

The key to success especially as to implementing an improved collection is education and awareness-raising among the general public (Gelbmann & Zimek 2018). The aim should be for all actors to cooperate in a systemic way. Thus, a shift in waste management also requires the adaptation of legal regulations, more transparency, product responsibility already at the generation of products stage or mergers in regions for waste disposal. This will pave the way from linear growth to a circular economy.

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APPENDIX

TABLE A1: Transport: loading weight, transport routes and employment per 100,000 tons per year for the individual transports

abbreviation	transport route designation	**loading weight** [t]	**transport route** [km]	**employees** per 100,000 t
WI-G – LF-R	waste incineration plant with grate firing >> residual waste landfill	20	50	9.0
WI-G – L-Shr	waste incineration plant with grate firing >> large shredder plant	25	100	12.0
WI-F – LF-R	waste incineration plant with fluidised bed combustion >> residual waste landfill	20	50	9.0
MBT – LF-M	MBT plant >> mass waste landfill	20	0	0.0
MBT – WI-F	MBT plant >> waste incineration plant with fluidised bed combustion	11	100	27.3
MBT – SFP-P	MBT plant >> substitute fuel production plant	11	50	16.4
MBT – Cem	MBT plant >> waste co-incineration plant with rotary kiln in the cement plant	11	150	32.8
MBT – L-Shr	MBT plant >> large shredder plant	25	100	12.0
SFP-P – Cem	substitute fuel production plant >> waste co-incineration plant with rotary kiln in the cement plant	11	150	32.8
L-Shr – P-Shr	large shredder plant >> post shredder plant	25	0	0.0
L-Shr – Met-Ind	large shredder plant >> metal industry	25	200	19.2
Po-Sh – Met-Ind	post shredder plant >> metal industry	25	200	19.2
LWP-S – MBT	lightweight packaging sorting plant >> MBT plant	20	100	15.0
LWP-S – SFP-P	lightweight packaging sorting plant >> substitute fuel production plant	11	50	16.4
LWP-S – Pla-R	lightweight packaging sorting plant >> plastic recycling plant	11	200	43.7
Pla-R – WI-F	plastic recycling plant >> waste incineration plant with fluidised bed combustion	11	150	32.8
Comp – MBT	composting plant >> MBT plant	20	50	9.0
Pap-S – Pap-M	wastepaper sorting plant >> paper mill	20	50	9.0
Pap-S – MBT	wastepaper sorting plant >> MBT plant	20	150	18.0
Gl-T – Gl-S	waste glass transfer station >> waste glass sorting plant	25	200	19.2
Gl-S – MBT	waste glass sorting plant >> MBT plant	20	150	18.0
Met-T – L-Shr	scrap metal transfer station >> large shredder plant	25	200	19.2

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EVOLUTION OF THE URBAN WASTE MANAGEMENT SYSTEM IN THE EMILIA-ROMAGNA REGION

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ABSTRACT

This multidisciplinary study aims to analyse how the urban waste management system has changed in the Italian region of Emilia-Romagna, during the decade in which a single regional regulatory unit, the Emilia-Romagna Territorial Agency for Water and Waste Services (ATERSIR), was established and became operational, and the waste management planning was centralized at regional level. Particularly, the following changes have been analysed: i) the methods of municipal waste management (WM), considering waste generation, separate waste collection and waste treatment; ii) the costs of WM service, with a focus on cost of treatment and disposal of unsorted waste; and iii) the urban solid WM policies, in terms of levels of governance, territorial planning and implementation of policies on the regional territory. The period within which the analysis was carried out covers the years from 2008 to 2018, comparing two time frames, before and after ATERSIR establishment. Data at municipal level were gathered and analysed. The results of the technical, economical and institutional assessment show that relevant benefits occurred, such as a constant improvement of environmental performances, the optimisation of the waste flows to plants, a higher level of uniformity of WM cost among Municipalities and a better quality of data collected from waste providers for the technical and economic regulation of the sector. Potential improvements are identified, whilst the institutional reform is positively evaluated in all the analysed aspects.

1. INTRODUCTION

Waste services are considered of public and general economic interest since they are essential to human comfort, public health and environmental quality, and since they are key elements for economy's competitiveness and society's overall well-being (Hoornweg and Bhada-Tata, 2012; Marques et al., 2018).

As argued by Campitelli and Schebek (2020), who reviewed 366 studies on waste management systems (WMSs) of cities or countries and focused on municipal solid waste (MSW), the assessment of WMSs is a crucial and still relevant topic, according to the increasing number of publications in the last 40 years. In recent years, the public interest associated with municipal waste management (MWM) has broadened, as it is concerned with much more far-reaching sustainability issues: materials consumption, availability of disposal sites, pollution from treatment cycles (Antonioli and Massarutto, 2011). According to Campitelli and Schebek (2020), besides environmental, organizational, and technical aspects, many studies have considered other aspects, in compliance with the increasing demand for broader sustainability assessment, where the environment, society and the economy are integrated (Hellweg and Canals, 2014). Furthermore, governance aspects have gained attention. Indeed, the governance is crucial for building up a WMS (Filho et al., 2016) and the institutional framework should be well designed in order to ensure the quality of the service and its environmental and economic sustainability.

In addition to technical and quality of the service regulation, most literature agree on the need of economic regulation in the waste sector in many different countries, as the sector can be subject to inefficient conditions due to market failures and lack of incentives (Di Foggia and Beccarello, 2020a; Sarra et al., 2020; Marques et al.,

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2018; Simões and Marques, 2012a; Simões and Marques, 2012b). Besides promoting the implementation of the principle of economic and financial sustainability and ensuring the suitability of fee structures at national, regional, or local levels (Marques et al., 2018), the role of economic regulation in enhancing the quality of the service (Marques and Simões, 2009; Simões and Marques, 2012a) and promoting the implementation of circular economy (CE) principles (Di Foggia and Beccarello, 2020a) is also recognised.

An extensive literature on the optimal operational size of (private or public) waste service providers is available: the impact of territorial scale on the efficiency and on the achievement of economies of scale and scope, both for waste collection providers and for owners of waste treatment or recycling plants, was debated (Stevens, 1978; Antonioli and Filippini, 2002; Simões et al., 2013; Abrate et al., 2014; Carvalho and Marques, 2014; Carvalho et al., 2015; Tanguy et al., 2017). Other studies discussed the optimal extension of the area to be served by single operators and the variables which should be considered to select Municipalities thereinto by adopting the perspective of regulatory authorities (Sarra et al., 2017; Sarra et al., 2020).

To the best of our knowledge, no studies have discussed the optimal territorial scope under the responsibility of a single economic regulation authority (i.e., in terms of number of service providers, or geographical extension), or the impacts on WMSs caused by a reform of the institutional structure. This study has the ambition to contribute to such discussion, through an analysis of the evolution of a MWM system in the Italian region of Emilia-Romagna, over the period 2008-2018.

In 2011, thanks to an internal reform, the Emilia-Romagna Region started to strengthen its role in the planning and regulation of urban WM services. The objective of this research paper is to study what impacts this transition had on the management of solid urban waste (UW). The analysis of variables directly affected by the economic regulation is complemented by an assessment of environmental and institutional aspects, to study the joint effects and the synergies between WM planning and economic regulation itself. This is a novelty, since the effects of economic regulation are usually evaluated in terms of effectiveness, efficiency, or productivity of providers, while neglecting an in-deep analysis of the locally pursued WM strategies. This partial approach might deliver misleading messages for decision makers, since achieving CE objectives might hinder efficiency of waste service operators, as highlighted by Di Foggia and Beccarello (2020b). Moreover, the territorial dimension suitable for the implementation of CE might be bigger than the area served by single operators.

Thus, since in the Emilia-Romagna region the CE policies are at the core of regional waste management planning, this study attempts to analyse how an institutional reform has affected these aspects. Particular attention is given to the analysis of the homogeneity level of performances among Municipalities in the region, as its increase is an objective of public policies and it might result from a centralised regulation.

2. MUNICIPAL WASTE MANAGEMENT IN THE EMILIA-ROMAGNA REGION

Emilia-Romagna is a region in Northern Italy that extends inland westward from the Adriatic coast, with a population equal to 4.459.477 inhabitants (2019). Emilia-Romagna is one of the wealthiest and most developed regions in Europe; it was the third wealthiest region in Italy in 2017, considering the indicator “primary income of private households”. The region is divided in 9 Provinces (Figure S1) and it includes 328 Municipalities (2019). In the same Figure S1, the MSW providers are also shown. The service providers in the Region remained almost the same over the analysed years (2008-2018).

2.1 Regulation of municipal waste management

In Italy, the public service regime applies to municipal waste, including household waste, orphan waste and a fraction of industrial and commercial waste, while the market regime applies to the remaining of commercial and business waste (special waste).

As a local public service, the municipal solid waste (MSW) management is regulated by a multilevel regulatory asset, organized in a central, national authority (ARERA, Italian Regulatory Authority for Energy, Networks and Environment), a secondary level, organized in areas defined by each Region, often corresponding to Italian provinces, and a municipal level, still important in the institutional framework and in the different institutional roles.

Focusing on the secondary level, since the beginning of the 1990s’, the management of local public services, and particularly water and wastewater services and MWM, was delegated to a new over-municipal structure, called Optimal Territorial Area (OTA), a territorial partition specifically designed to exploit fully economies of scale/scope/density (Massarutto, 2010). A single authority for each OTA (on the board of which all the municipalities that make it up were represented) was responsible for the choices made to organize/coordinate the entire waste cycle (public management, private management or public–private partnership, concession to operate the services, tariffs, etc.) (Sarra et al., 2017).The aim was the improvement in efficiency and effectiveness of the previous municipal-based providers, by applying a more industrial approach to the service and promoting managerial integration among the different activities involved in the waste cycle. As far as the Emilia-Romagna Region is concerned, the OTAs were set in 1999, with a provincial dimension. Therefore, nine different regulatory agencies were defined in the region.

Nowadays, the economic regulation of the Italian MSW is scattered in various regional scenarios. The regulatory agencies, defined by each Region, can have the same size of the region (for example, Emilia-Romagna), a multi-province dimension (as it happens in Toscana Region) or even do not exist at all. In the last case, single Municipalities, or small associations of few Municipalities, are in charge of the MSW collection and treatment service (Biagini, 2016).

According to Italian law, the identification of the optimal size for the organization of the waste service must be based on the application of the following criteria (Legislative Decree 152/2006, art. 200):

- overcoming the fragmentation through an integrated WM service;
- achievement of adequate management dimensions, defined on the basis of physical, demographic, and technical parameters, and considering the political-administrative subdivisions;
- appropriate evaluation of the transportation systems, in order to optimise the transport within the OTA;
- exploitation of common needs and similarities in waste production and management;
- reconnaissance of WM plants already built and in operation;
- consideration of previous delimitations, so that new OTAs deviate from previous ones only on the basis of justified requirements of effectiveness, efficiency, and affordability.

Furthermore, the principle of proximity of the places of waste production to recovery and disposal plants should be considered. Additionally, the achievement of self-sufficiency in disposal, within the region, is recommended, in order to reduce movements of waste, taking into account the geographical and social context or the need for specialized plants for certain types of waste (Legislative Decree 152/2006 art. 182-bis).

It is worth noting that “optimal organizational size” and “optimal managerial size” do not always coincide. Thus, according to Article 34 of Law 221/2012, the OTA authority is not obliged to entrust the management of the collection service to a single operator. Indeed, an OTA itself can be divided into multiple sub-areas (service-specific optimal territorial areas, or SOTAs) if specific services (collection, treatment, etc.) can achieve economic efficiency at smaller territorial scales, and several operators can be entrusted with the service, one for each SOTA, under the supervision of the OTA authority, which guarantees the coherence of the whole system (Sarra et al., 2017).

In 2011, with the regional Law 23/2011, the Emilia-Romagna region reformed the regulatory architecture of the OTAs: a new unique regional OTA was defined, and at the same time, a new institution, the “Emilia-Romagna territorial agency for the water, sanitation and waste services”, (or ATERSIR, in the Italian acronym) was established, dismissing the provincial ones (Emilia-Romagna Region, 2011). From an administrative point of view, the new regional agency was set up at an intermediate level between the strictly local level and the national one.

So, since 2012 ATERSIR has been responsible for the economic regulation of the MWM. Its main tasks are those expected by the national legislative framework: economic regulation of the service; financial and economic planning; entrusting MWM service; verifying that the services are fully compliant with the European framework directives on competition; planning and scheduling projects finalized to comply with the European and Italian environmental performances and quality standards.

The governance was structured at two administrative levels: a regional board assuming the main functions and responsibilities and nine local councils of provincial (sub-regional) dimension (Biagini, 2016).

Following Massarutto (2010), in the value chain of MWM three main phases can be identified, which give rise to three distinct markets. The first one concerns collection service, whose counterparts are waste producers and operators. The second is the market for the handling and disposal of waste, whose counterparts are operators of collection services and owners of disposal sites. The third is recovery/recycling, whose counterparts are again collection operators and final users of waste-derived materials. In this framework, ATERSIR regulates the collection services and the unsorted waste treatment and final destination (waste-to-energy (WTE) plants and landfills), while for the third phase mentioned above, the Agency ensures and supervises the application of the best treatment prices. In particular, since 2014, accordingly to a specific regional regulation, ATERSIR has performed the regulation of “gate fees” for the treatment plants for unsorted UW, i.e., WTE plants, mechanical-biological treatment plants and landfills.

In Emilia-Romagna, UW service is managed by 11 different providers (2019). ATERSIR, managing 18 contracts, approves each year the financial plans for each single municipality for a total amount of approximately € 750.000.000.

2.2 Regional legislation and planning of waste management system

In Italy, responsibility for environmental policies is held by the national government, but the regional government is the responsible authority for the Waste Management Plan (Legislative Decree 152/2006, art. 199). In the General Direction for Environment and for Soil and Coast protection of Emilia-Romagna Region, a specific team works on the implementation of objectives for a sustainable WM.

In 2011, the transition from 9 OTAs to one regional OTA implied the transition from 9 Provincial WM Plans to a single Regional WM Plan, given that the planning function had been delegated to the Provinces in previous years.

In the “WM plan of Emilia-Romagna Region”, definitively approved in May 2016, the Region defined the strategic objectives for sustainable WM, in line with the European hierarchy which puts prevention and recycling first. The plan includes a prevention programme, with a time frame of 7 years (2013-2020); a lot of typologies of waste prevention measures are set, with impact on different product life stages (Magrini et al., 2021). Before 2012, indeed, the implementation of waste prevention programmes was on voluntary basis, and only few provinces had implemented waste prevention policies in their territory.

Furthermore, as promoted by European policies, Emilia-Romagna was the first Italian region to assume the CE principles in its waste policy by law, with the aim of reducing waste production and recovering as much material as possible for recycling.

The Regional Law 16/2015 explicitly introduces a new concept of "sustainable development", assuming that the winning approach is the one which tends towards a holistic, comprehensive, and not sectorial vision. The transition to a more CE offers great opportunities for development with resulting advantages in economic terms (i.e., employment, competitiveness), as well as energy savings and environmental benefits. Moreover, the Regional Law 16/2015 defines targets for waste prevention and management, to be achieved by 2020:

- reduction in per capita waste generation by 15%-20%, compared to the 2011 values, mainly through the implementation of Pay-as-you-throw (PAYT) systems;
- annual per capita amount of UW not sent for recycling not exceeding 150 kilograms per inhabitant;
- 73% of separate waste collection rate at regional level, differentiated for homogeneous area;
- recycling of paper, metals, plastic, wood, glass and organic waste by at least 70%;
- self-sufficiency in the disposal of non-hazardous municipal and special waste at regional level, through the optimal use of existing facilities;
- minimisation of disposal, in particular landfill.

Emilia-Romagna presents a rather uniform socioeconomic situation, but it has a variety of geographic, urban and WM characteristics (Passarini et al., 2011). The creation of a regional governance made possible the grouping of similar municipalities (Figure 1): the entire regional territory was divided into three "homogeneous areas", identified by crossing physical-geographical factors (geomorphological elements, altimetry) with human-related factors and WM characteristics (population density, percentage of separate collection already achieved).

At last, it is important to underline that the regional WM plan has been monitored yearly, in order to be reactive to any need that might arise from the municipalities.

3. MATERIALS AND METHODS

3.1 Methodology of the assessment

The methodology used in this study included the selection and calculation of some quantitative indicators, as well as a qualitative assessment, to assess technical, environmental, economic, and institutional aspects of the regional WMS. Since specific aim is the analysis of the evolution of the homogeneity level of performances among the Municipalities in the region, over the time frame 2008-2018, the average and the standard deviation of some indicators calculated at municipal level were also assessed.

As far as technical and environmental aspects are concerned, three stages of integrated WMS were analysed: waste generation, separate waste collection and waste treatment. The main targets set by policies were evaluated.

With growing concerns about the economic sustainability of waste services, a number of studies focused on their cost structure. The authors chose to analyse the total cost of WM and provide a focus on cost of treatment and disposal of unsorted waste (CTS, in the Italian acronym, according to the national law D.P.R. 158/1999).

The analyses were carried out, if possible and relevant, by comparing two different time frames in order to illustrate the level of some indicators before (2008-2012) and after (2013-2018) the governance reform process and the unique OTA establishment. As an exception to this rule, the analyses on the costs of treatment and disposal of unsorted waste considered two different time frames (2008-2013 and 2014-2018) since in 2014 the regional legislation reg-

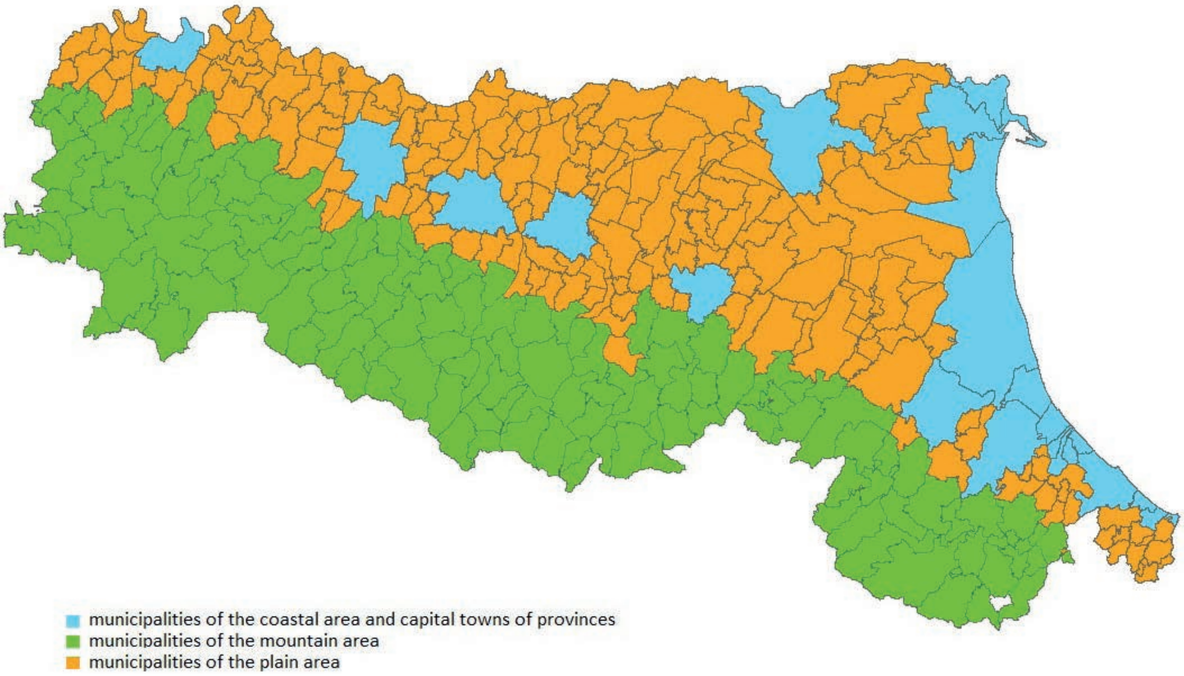

FIGURE 1: Municipalities of Emilia-Romagna region, clustered by “homogeneous areas”. Source: Emilia-Romagna Region, 2016.

ulating the disposal fee was applied for the first time. The economic performances of the Emilia-Romagna region were compared to the ones of other Italian Regions, selected according to some relevant criteria:

- similar geographical position and economic situation;
- presence of an operating regulatory agency, according to Invitalia (2019). Further details are provided by Table S1.

The selected Regions are: Lombardia, Veneto, Toscana, Umbria, Friuli-Venezia Giulia, Liguria, Marche and Province of Trento. It should be noticed that Lombardia was selected, despite not having an operating regulatory agency, as it represents an alternative model, compared to Emilia-Romagna. Nevertheless, it should be noticed that Emilia-Romagna is the only Region which regulates waste treatment fees.

Finally, some relevant aspects of the institutional assessment were selected and some consequences of a regionally unified regulatory agency for the MSW management, together with a unique regional planning were discussed.

3.2 Data sources

Data for each Municipality in the Emilia-Romagna region were gathered, referring to the time frame 2008-2018. In particular, the following data sets were systematised:

- generation of sorted and unsorted waste;
- resident population;
- economic data (i.e., total cost of the service and CTS).

On the other hand, data on WM (waste sent to landfill and incineration, recycling rates) were gathered and analysed on a regional scale per single year. The data sources were the Emilia-Romagna region, the Regional Agency for Prevention, Environment and Energy (ARPAE), and ATERSIR.

As far as economic data are concerned, a demanding historical research and data systematisation activity were necessary. Economic data indeed were collected from the databases of the provincial OTAs or from ATERSIR database, if referred to the period from 2008 to 2011 or from 2012 to 2018 respectively. Data from the municipal economic-financial plans were mainly used: these plans aim to estimate the costs of the service which contribute to establish the fees for the users. If this value was not available, data were gathered from a regional database, which usually reports real costs, instead of the planned ones. Details on the number of Municipalities considered in the economic assessment are available in Table S2, which also details the number and percentage of Municipalities whose data do not come from financial-economic plans, but from the regional database.

To compare the regional system with either the national one or the one of other Regions, data made publicly available by the Italian National Institute for Environmental Protection and Research (ISPRA) were used (ISPRA, 2019; ISPRA, 2020a; ISPRA, 2020b).

4. RESULTS AND DISCUSSION

4.1 Technical and environmental assessment

4.1.1 Waste generation

Table 1 shows the total UW and unsorted UW generation, waste generation per capita, at regional level, in the analysed period from 2008 to 2018. The regional legislative approach and regulation seem to have a limited impact on the decrease of the total amount of waste production, over the time frame analysed. On the other hand, the regional policies and the institutional framework led to a considerable and constant reduction of unsorted waste production.

As shown in Figure S2, per capita UW generation in Emilia-Romagna was significantly higher than the average Italian value in all the eleven years of analysis. The critical factors are structural and related to:

- the economic situation: the Region is one of the most economically developed and flourishing areas in Italy. The direct correlation between waste generation and the main economic indicators, such as gross domes-

TABLE 1: Evolution of total urban waste production and per capita production in the Emilia-Romagna region. Source: authors' elaboration based on Emilia-Romagna Region data.

Year	Urban waste generation [tonnes]	Unsorted urban waste generation [tonnes]	Inhabitants	Per capita urban waste generation [kg/inhab]	Per capita unsorted urban waste generation [kg/inhab]
2008	3.013.721	1.646.404	4.337.966	694,73	379,54
2009	2.987.477	1 571.876	4.377.473[1]	682,47	359,08
2010	3.093.089	1.535.054	4.432.439	697,83	346,32
2011	3.002.771	1 415.337	4.459.246	673,38	317,39
2012	2.893.518	1.334.030	4.471.490	647,10	298,34
2013	2.896.432	1.268.472	4.453.435	650,38	284,83
2014	2.929.953	1.223.344	4.457.115	657,37	274,47
2015	2.962.076	1.165.311	4.454.393	664,98	261,61
2016	2.969.293	1.132.866	4.457.318	666,16	254,16
2017	2.895.720	1.034.832	4.461.612	649,03	231,94
2018	3.011.354	964.693	4.471.485	673,46	215,74

[1] *The geographical area of the region changed in 2009, as 7 Municipalities were annexed from Marche region.*

tic product, is well known. As shown in Figure S2, indeed, the economic crisis influenced waste production both at national and regional level, contributing to the decrease of the total amount of UW generated in 2009 compared to 2008, despite the annexation of 7 new Municipalities to the region.

- the categories of waste assimilated to UW: Emilia-Romagna WM is overall characterized by a high amount of commercial/industrial waste assimilated to UW, also due to a productive network strongly connected to the urban one and to the plant engineering capacity present on the territory. This made it possible to manage the assimilated waste in complete safety and to guarantee its traceability, while containing the tariffs applied to domestic and non-domestic users;
- tourist flows: in Emilia-Romagna in 2018, there were 11.458.497 visitor arrivals and 40.647.799 guests in hotels and complementary structures, with an average length of stay of 3,55 days, mainly in the provinces of Rimini, Ravenna and Forlì-Cesena (for details, see Table S3). The values of the per capita waste generation indicator and per capita unsorted waste generation for these Provinces are among the highest in the region (Figure S3 and S4).

Analysing data on waste generation at municipal level, the average and the standard deviation of per capita total waste generation, per capita sorted and unsorted waste generation for each considered year were calculated (Figure 2). Even if a decrease in the average value of per capita unsorted waste and an increase in the average value of per capita sorted waste occurred, the standard deviation did not decreased and the performances of the Municipalities did not become more homogenous over the years of analysis. The reasons for this can be found in the peculiarities of Municipalities, mainly in terms of assimilation and tourist flows.

The dataset could be more homogeneous (with a lower standard deviation) considering not only the resident inhabitants, but the parameter "Inhabitant Equivalent", introduced by Regional Law 16/2015 in order to make the performance of municipalities comparable in terms of MSW production. For each Municipality, this parameter is calculated, year by year, as the sum of contributions related to different quantities: resident citizens, non-resident households, university students, tourist flows, productive activities.

Moreover, the variation of per capita waste production at municipal level was studied, over the two above-mentioned time frames. The results show that the number of Municipalities which recorded an increase of this indicator (negative variation in x-axis) is higher over the second time frame than over the first one (Figure S5 and S6). The same results are shown if the analysis is repeated on per capita unsorted waste generation, applying the same methodology (Figure S7 and S8). Further research is needed to fully understand the reasons behind these performances.

Within the scope of this research paper, the effects of PAYT schemes on waste production were not studied. The number of Municipalities which implemented PAYT schemes increased from zero to 60 over the 2008-2018 decade.

4.1.2 Separate waste collection

The separate collection rate in the Region increased steadily over the analysed decade, from 45.4% in 2008 to 68% in 2018. The increase was also constant on a national scale, going from 30.6% in 2008 to 58.1% in 2018 (Figure S9).

The reform of the institutional framework in 2011/2012 did not transform the trend, but a significant improvement occurred: the separate collection rate increased 9,4% between 2008 and 2012, while in the second time frame (2013-2018) the increase rate was 15,8%. Given that the more separate collection rate rises, the harder is to achieve further improvements, this data is a really important result and proves the regional commitment on the improvement of separate waste collection.

The separate collection rate over the period 2008-2018

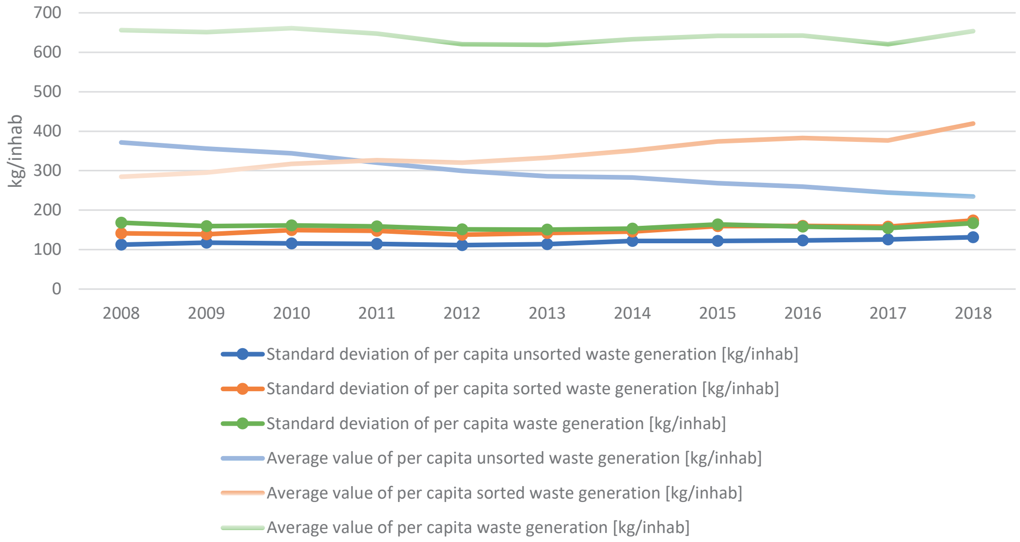

FIGURE 2: Average and standard deviation of per capita sorted and unsorted waste generation at Municipal level. Source: authors' elaboration based on Emilia-Romagna Region data.

in the three homogeneous areas had the same trend as the regional one (Figure S10). Given that the target for this indicator is different for each Municipality according to the relevant homogeneous area the average value and the standard deviation were not analysed.

Furthermore, the percentage variation of separate waste collection rate at municipal level was studied, focusing on two time frames: 2008-2012 and 2013-2018. Considering the availability of data, two samples of 341 and 321 Municipalities were object of analysis in the two time frames respectively. The results show that in the second time frame the number of Municipalities achieving a good performance in separate collection increased, as well as the number of Municipalities with a positive trend of the separate collection rate (Figure 3 and Figure 4, where each dot represents a Municipality). A deeper analysis is required to understand the reasons of the values of outliers Municipalities.

4.1.3 Waste treatment

Even if the collection rate is often used in the assessment of effectiveness of waste service (e.g. CNEL, 2019),

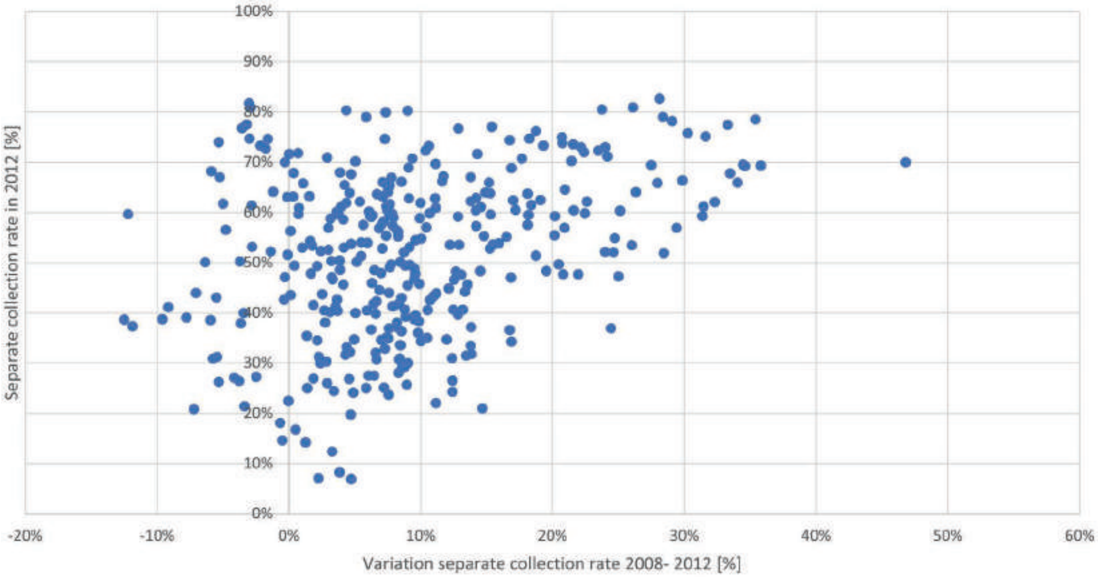

FIGURE 3: Variation of separate collection rate (2008-2012) and separate collection rate (2012), for each Municipality in the Emilia-Romagna region. A negative variation (x-axis) represents a decrease in the separate collection rate over years. Source: authors' elaboration based on Emilia-Romagna Region data.

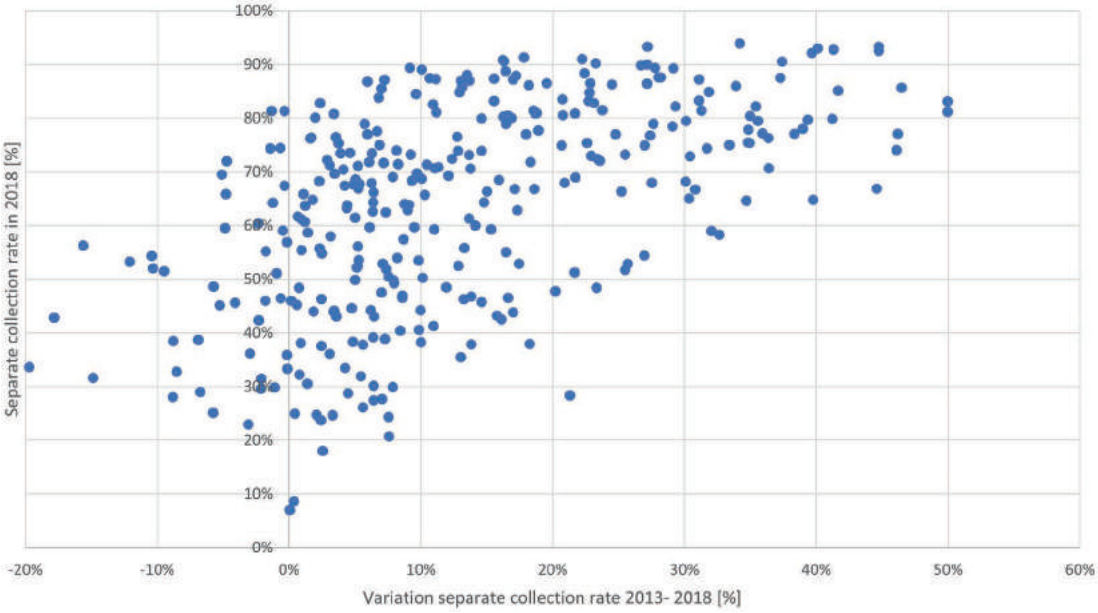

FIGURE 4: Variation of separate collection rate (2013-2018) and separate collection rate (2018), for each Municipality in the Emilia-Romagna region. A negative variation (x-axis) represents a decrease in the separate collection rate over years. Source: authors' elaboration based on Emilia-Romagna Region data.

other aspects of waste service performances should be taken into account in order to assess the compliance with the regulatory framework.

In the region, landfill use constantly decreased over the eleven years analysed: the overall landfill rate in 2018 was 2,79%, already well below the target of 10% by 2035 set out in European Directive 851/2018, and below the Italian rate (Figure S11). No substantial changes in trends can be observed when comparing the time frames 2008-2012 and 2013-2018 (decrease by 12,65% over the first time frame, and by 13,28% over the second one). Landfill diversion was achieved through a combination of strategies, applying the European waste hierarchy and promoting prevention, recycling and WTE plants. As Antonioli and Massarutto (2011) inferred from the analysis of landfill reduction in 8 European Countries over the period 1995-2005, significant diversion rate can be achieved only where all strategies (recycling, indirect material recovery, waste-to-energy) are combined, while strategies concentrated on sole recycling, despite some success at local scale, do not seem to be able to perform at the scale of the overall system.

Emilia-Romagna extensively uses WTE plants, achieving an incineration rate higher than the national one (Figure S12). The amount of UW sent to incineration or used for the production of refuse derived fuel (RDF) in the region increased at a constant rate over the analysed time frame: it increased by 4,89% over the time frame 2008-2012, and by 3,87% over the period 2013-2018.

Considering sorted waste destination/treatment, the regional recycling rate for some waste flows, in the period 2014-2018, and the total recycling rate at regional and national level were analysed (Figure S13). Because of regional data availability, it was not possible to elaborate the indicator for previous years. According to annex 1 of European Commission Decision 2011/753/EU, the calculation method 2 was applied, as also suggested by the Italian Ministry of environment, land and sea. Thus, the ratio between the recycled amount of paper, metal, plastic, glass waste and other single waste streams from households or similar waste stream, and the total generated amount of the same waste streams was calculated. No considerations on waste treatment at municipal level can be drawn.

4.2 Economic assessment

4.2.1 Total cost of waste management

Firstly, the amount of management costs of the urban hygiene service covered by institutional entities (Municipalities, their Consortia and other service managers) were considered, as reported by ISPRA. Because of data availability, the cost of WM has been evaluated in the period 2011-2018. The comparison between the average regional and national costs of WM highlights that Emilia-Romagna has an efficient management of UW, considering the ratio between costs and quantities of waste (Figures S14 and S15). Per capita costs grew, correspondingly to the national average value, while costs per tonne remained fairly constant after 2014. Moreover, the average "per tonne cost of WM" at regional level was lower than the one of almost all the selected Regions in the period 2011-2018 (Figure S16), while the standard deviation of the same indicator assessed at municipal level remained fairly stable, compared to the one of the other Regions (Figure S17).

Secondly, an analysis of the municipal economic-financial plans officially approved by the regulatory authorities was performed, for the time frame 2008-2018, calculating WM cost per resident inhabitant and per tonne of generated waste. Figure 5 shows the evolution of the indicators on regional WM cost. The change in the institutional structure seems to have impact on the growth trend of the total costs of the service, by containing the increase over time: while an increase in the cost per inhabitant and per tonne by 11% and 19% respectively occurred between 2008 and 2012, the indicators increased 7% and 3% respectively in the period between 2013 and 2018.

Then, analysing the costs at municipal level, the values of WM cost per capita and per tonne were calculated. The standard deviations of these datasets for each considered year are outlined in Figure 6. The standard deviation of the cost per tonne indicator had a higher increasing rate before 2012, while after 2013 it seems steadier, showing a higher level of uniformity among the Municipalities. The standard

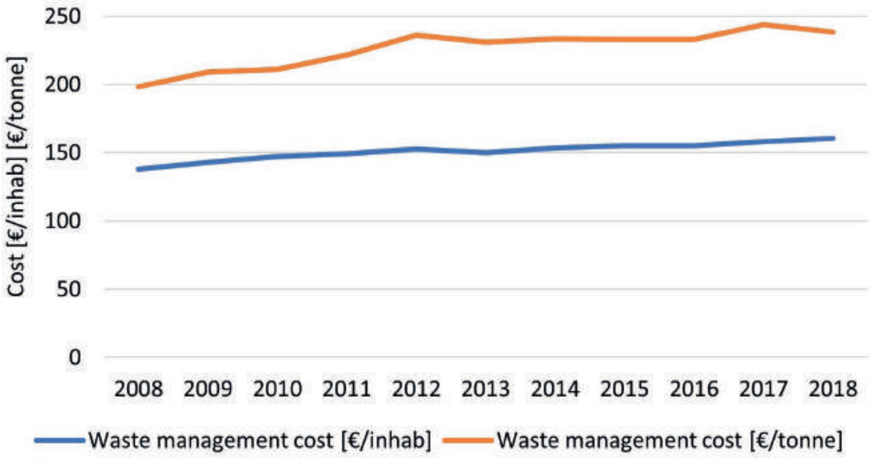

FIGURE 5: Waste management cost in Emilia-Romagna, 2008-2018. Source: authors' elaboration on ATERSIR data.

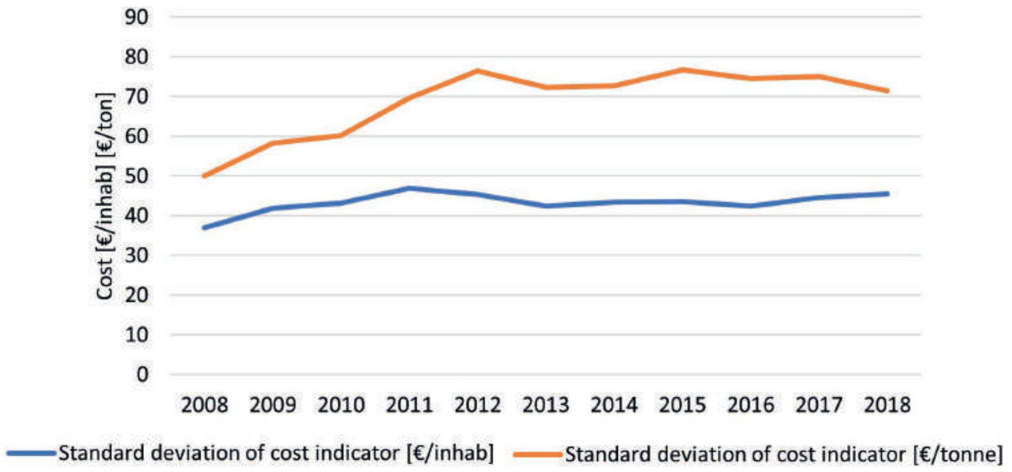

FIGURE 6: Standard deviation of waste management cost in Emilia-Romagna, 2008-2018. Source: authors' elaboration on ATERSIR data.

deviation of the cost per inhabitant indicator was quite constant over the period of analysis.

Moreover, the variation of per capita WM cost at municipal level was studied, focusing on two time frames: 2008-2012 and 2013-2018. The results show that in the second time frame a lower number of Municipalities had considerable variations in the per capita costs (Figures 7 and 8). This proves a higher homogeneity of the per capita WM cost indicator among Municipalities.

In the two time frames analysed, a different distribution of the cost for each municipality is clearly visible (in the figure, each dot represents a municipality). In particular, in the second Figure (Figure 8) the pattern highlights an extremely minor variability (represented by the higher density of the cloud) and a significant number of municipalities switching on the negative values of the x-axis, thus meaning that in those municipalities the total cost has decreased.

4.2.2 Cost of treatment and disposal of unsorted waste (CTS)

According to Biagi and Massarutto (2002), CTS is the less transparent cost item, and the one in which, in all likelihood, the main causes of potential market failures are to be found. Moreover, according to the analysis of empirical data of 6,616 Italian municipalities for a two-year period performed by Di Foggia and Beccarello (2020b), waste-to-energy plants and landfills play a remarkable role in WM cost determination. Thus, this in-deep analysis of CTS was considered relevant by the authors.

The average and the standard deviation of the per tonne cost, analysed at municipal level, show that the trend of both indicators over the eleven years was quite stable (Figure 9).

Unfortunately, regarding the fees for access to treatment facilities, there are no national databases which al-

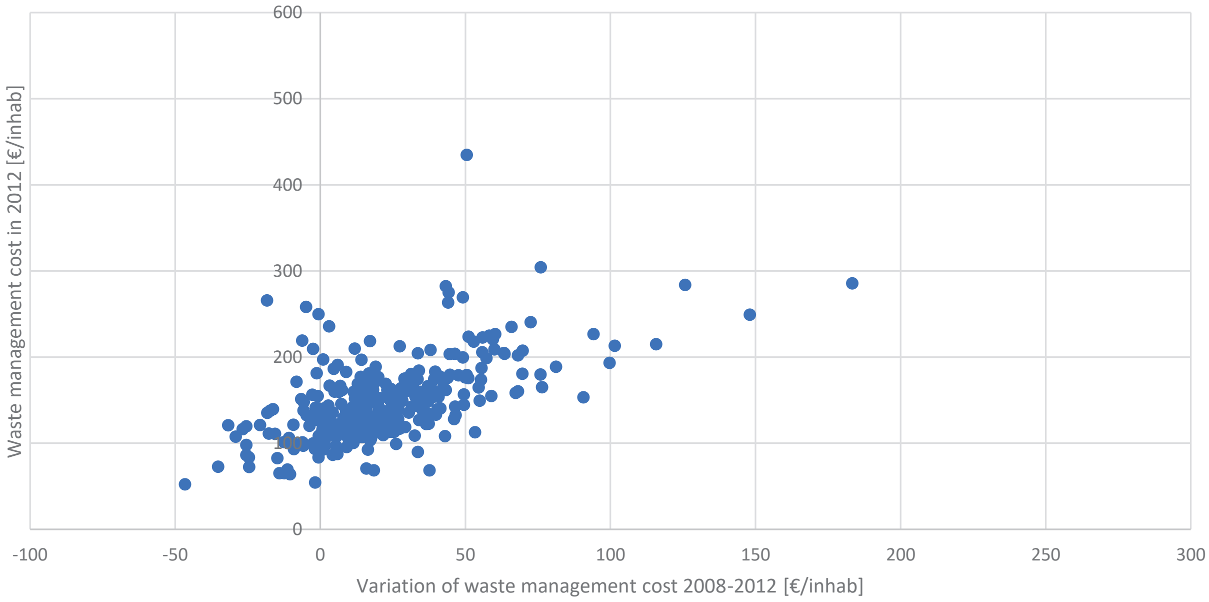

FIGURE 7: Variation of waste management cost (2008-2012) and waste management cost (2012), for each Municipality in the Emilia-Romagna region. A negative variation (x-axis) represents a decrease in the cost over years. Source: authors' elaboration on ATERSIR data.

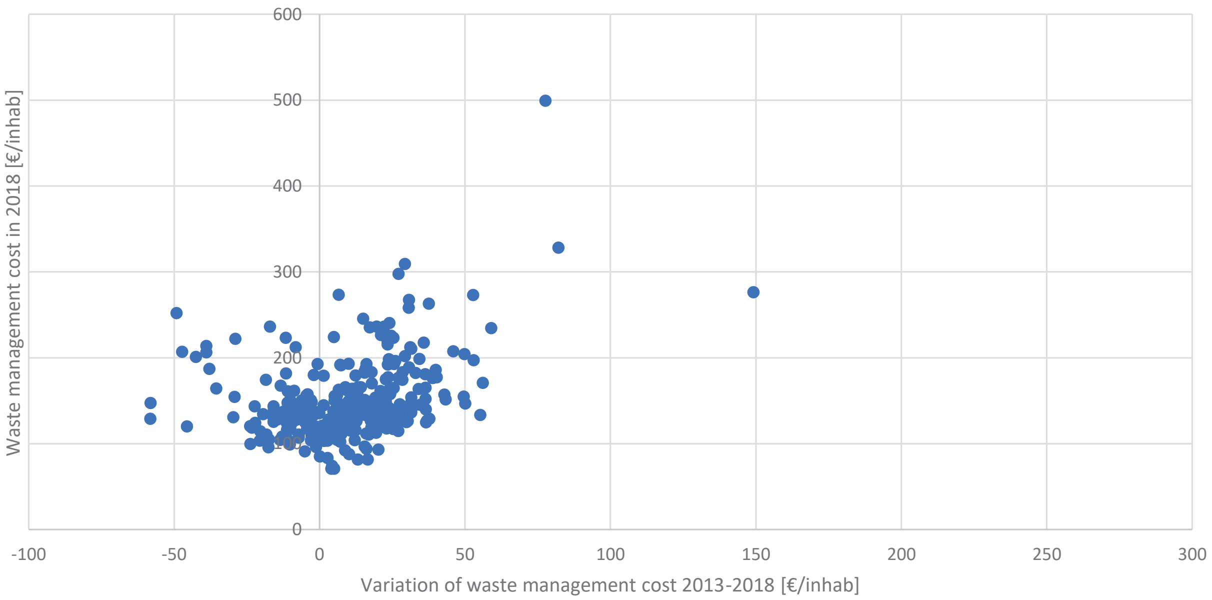

FIGURE 8: Variation of waste management cost (2013-2018) and waste management cost (2018), for each Municipality in the Emilia-Romagna region. A negative variation (x-axis) represents a decrease in the cost over years. Source: authors' elaboration on ATERSIR data.

low one to fully understand how many and which treatment plants are subject to a certain administered rate and how much they apply market prices (ARERA, 2018; Moretto et al., 2019). Therefore, the comparison with the national level was performed by using data from ISPRA. By comparing the standard deviation of the indicator "CTS per tonne" at regional level with the standard deviation of the same indicator assessed in other selected Italian regions, it came to light that in Emilia-Romagna this indicator had a low variability and no peaks (Figure 10). Over the same time frame, the average value was generally lower compared to the one of the other Regions (Figure S18).

Moreover, the variation of per tonne CTS at municipal level was studied. Considering the availability of data, two samples of 299 and 325 Municipalities were object of analysis in the two time frames, respectively. The results show that in the second time frame the variation of CTS was generally and considerably lower.

4.3 Institutional assessment

4.3.1 System of data collection

A positive consequence of establishing a unique regional Agency is surely the higher capability of data acquisition, harmonization and elaboration (Biagini, 2016). On the other hand, a process of homogenisation of information provided by waste service operators and municipalities to the regulatory authority was required, to enable comparisons among different providers. For this purpose, in 2012 the Regional Council Resolution 754/2012 was issued with the aim of making available to ATERSIR a set of technical, management and economic data describing the entire WM cycle, in a uniform manner throughout the territory and according to fixed reporting templates over time, while avoiding as much as possible the duplication of information required to waste operators.

Thus, the acquired information allowed ATERSIR to create the know-how necessary to perform benchmark analyses, thus supporting the decision-making process and the

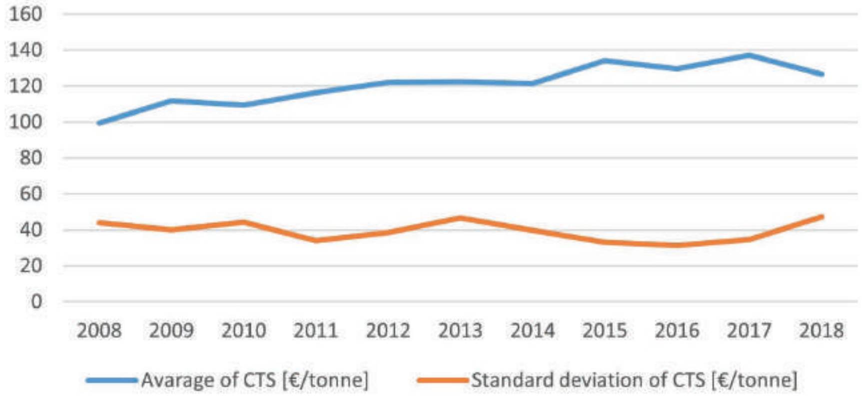

FIGURE 9: Average and standard deviation of cost of treatment and disposal of unsorted waste in Emilia-Romagna, 2008-2018. Source: authors' elaboration on ATERSIR data.

adoption of the best regulatory policies, and to define the disposal fees.

Every single local authority was previously able to analyse data originated from one to maximum four providers (in the Province with the highest degree of fragmentation). Nowadays, the staff of the regional agency can simultaneously analyse data originated by 12 providers and referred to the whole regional territory.

The so-called information asymmetry, which is intrinsic in the interaction between the regulator and regulated firms, was reduced, compared to the previous situation, and the regulatory body was effectively strengthened.

4.3.2 Reporting, access to information and waste traceability projects

Over time, a regulatory approach on a wider scale made

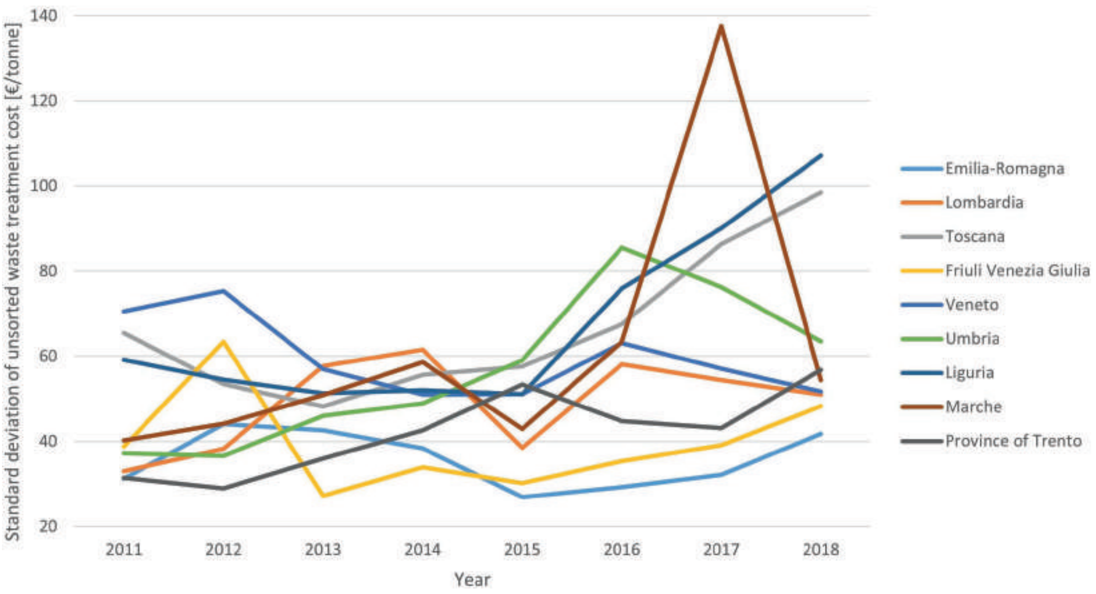

FIGURE 10: Standard deviation of cost of treatment and disposal of unsorted waste (CTS) for some Italian Regions, at municipal level. Source: authors' elaboration on ISPRA, 2019.

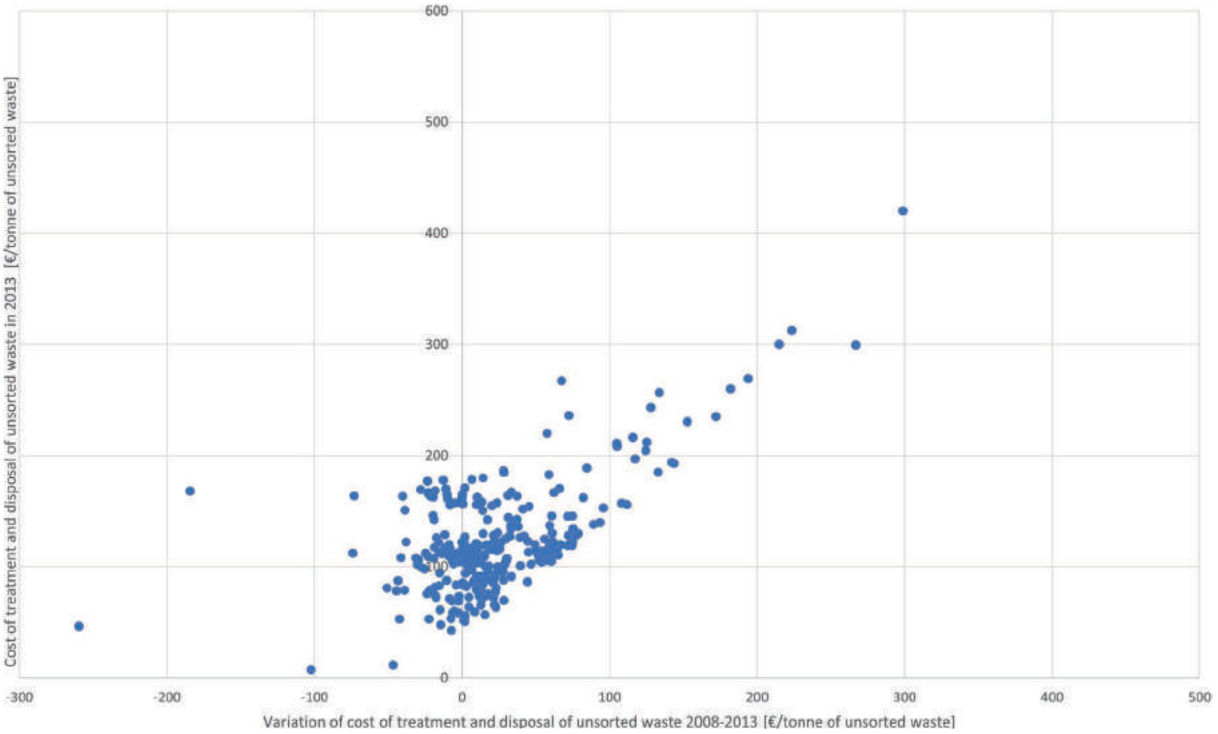

FIGURE 11: Variation of cost of treatment and disposal of unsorted waste (2008-2013) and cost of treatment and disposal of unsorted waste (2013), for each Municipality in the Emilia-Romagna region. A negative variation (x-axis) represents a decrease in the cost over years. Source: authors' elaboration on ATERSIR data.

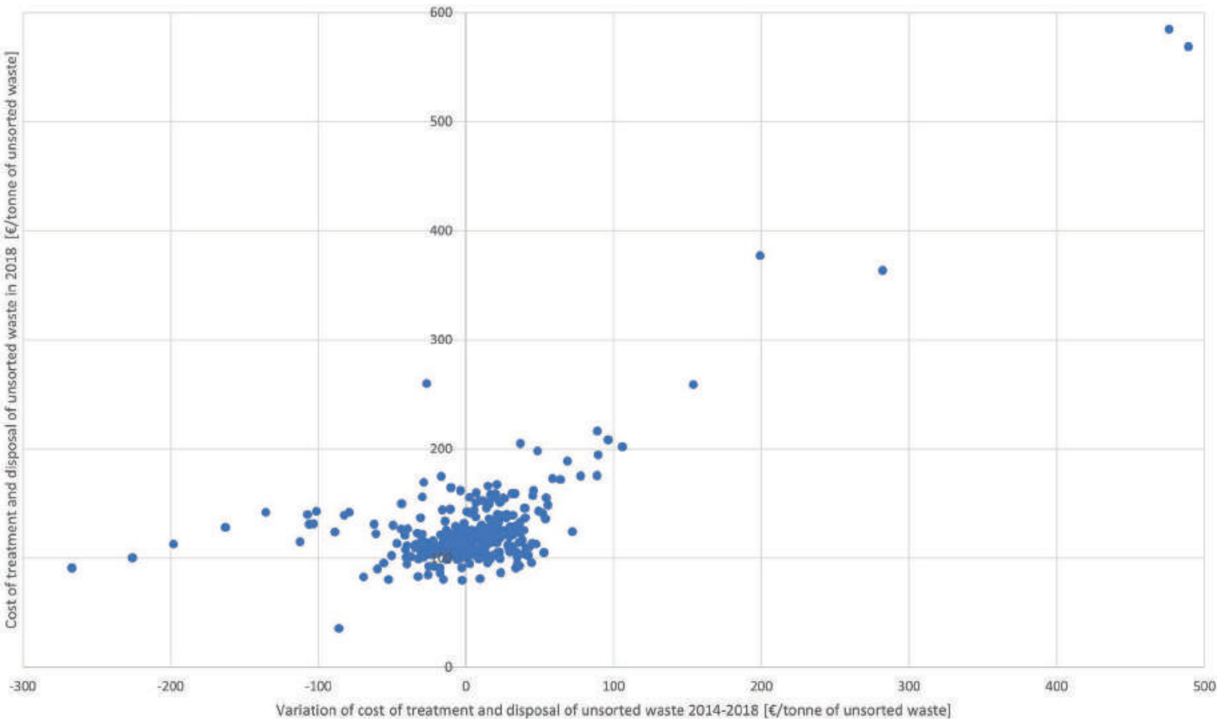

FIGURE 12: Variation of cost of treatment and disposal of unsorted waste (2014-2018) and cost of treatment and disposal of unsorted waste (2018), for each Municipality in the Emilia-Romagna region. A negative variation (x-axis) represents a decrease in the cost over years. Source: authors' elaboration on ATERSIR data.

the different territories comparable, while disclosing uniform information in terms of regional system. The scope of analysis was enlarged, enhancing the reports for citizens with economic analyses.

For example, since 2011, economic analysis of WMS has been included in the annual reports on regional WM, published by ARPAE.

In 2016, the Observatory of WM Plan Cost was set up with the participation of the Region, ATERSIR and ARPAE, with the aim of integrating, monitoring and analysing together territorial, technical and infrastructural information related to the waste service, correlating it to the economic impacts and the effects on users' tariffs.

Another example is the initiative "Chi li ha visti?" ("Who saw them?", translating), which analyses and traces the UW produced annually in the region, retraces the path of some waste flows after separate collection and disseminates data on material recovery, providing a useful tool for monitoring the regional WM plan. The project, carried out by the Region with the collaboration of ARPAE and with the contribution of CONAI (National Packaging Consortium) and based on measured data (Emilia-Romagna region, 2019a), has been conducted since 2009, and was made possible by the regional level of analysis. Emilia-Romagna is the only region where a waste traceability activity is carried out yearly by the public administration.

4.3.3 Rationalization of waste management plants in the region

In the previous phase of provincial WM planning, each Province was required to be self-sufficient in WM. This had resulted in a complete, modern, and efficient system of WM plants in the regional territory. The transition to a regional WM planning has led to an optimised management of the plants, avoiding redundancy, having to guarantee self-sufficiency of WM in a larger area.

From 2014 onwards, the region started to plan individual unsorted waste streams to treatment plants, year by year, in compliance with the European waste hierarchy, achieving coordination between collection and disposal. A substantial rationalisation and reduction of plants was allowed by the interoperability between the provincial territories and the dynamic optimisation of the waste flows to existing plants, in compliance with two fundamental principles: the progressive closure of landfills and the self-sufficiency in the management of urban and special waste generated in the region. Moreover, the surplus capacity of WTE plants at regional level is used to treat commercial and business waste (Laboratorio REF Ricerche, 2020), reaching a balance between the need and the overall plant capacity. Thus, Emilia-Romagna is one of the few regions in Italy achieving self-sufficiency. Since the use of WM facilities within the territory (with the exception of mechanical biological treatment) is a significant cost reduction driver (Di Foggia and Beccarello, 2020b), self-sufficiency in WM capacity contributes also to the economic sustainability of the service.

4.3.4 Waste management service entrustment

One of the main tasks of ATERSIR is to contract out the waste service, according to the methodologies accepted by the European legislation, i.e. in-house providing society, public-private partnership and international call for tender.

Particularly, the decision to proceed to public international competitive tendering procedures for service entrustment is an important innovation in this field, considering the traditional in-house management of these services, directly provided by public-owned companies.

All the decisions about service entrustment, included the form of the concession commitment, have been carried out in close agreement with the municipalities.

While almost all the previous service concession contracts lasted until the end of 2011 and the end of 2014, and nothing had been decided by the former 9 provincial authorities about how to entrust the service, ATERSIR in the years between 2012 and 2018 arranged three different new in-house providing concessions and published three calls for tender, concerning three different areas. Still, as this paper is being written, the Agency is trying to entrust the MSW service in all the basins where the concession contract is expired.

Probably, the previous, local, administrations (grouped in the former provincial authorities) could have been weaker in contracting out the service, in designing a tender or in analysing and managing the real effectiveness and efficacy of self-owned companies, demonstrating the concrete risk of the regulatory capture both by the political instances and industrial interests (Armstrong and Sappington, 2006).

4.3.5 Standard guidelines for the Municipalities

After 2012, the regional and centralised governance allows the development of standardised guidelines and operating procedures, with practical implications for Municipalities. For example, in 2018 standard rules for the definition of the waste fees in PAYT systems were issued (ATERSIR, 2018; Emilia-Romagna Region, 2018; Emilia-Romagna Region, 2019b). The Region has also regulated the activity of reuse centres through the guidelines for municipal and non-municipal reuse centres (Emilia-Romagna Region, 2017). Moreover, in 2019 uniform procedures to facilitate the removal of small amounts of asbestos-containing material from a compact matrix were defined (Emilia-Romagna Region, 2019c).

5. CONCLUSIONS

This study provides an integrated analysis of waste services in the Emilia-Romagna region over the period 2008-2018, considering three dimensions of sustainability (environmental, economic, and institutional). Environmental and economic data were gathered and analysed at regional and municipal level, considering different time frames, before and after the institutional reform which unified the provincial ATOs in one unique regional ATO.

The institutional assessment has taken into account the different institutional levels and how this new division of roles has impacted on decision-making and planning processes.

Since the providers of waste services are the same over the analysed period, it can be assumed that the variations have to be ascribed to changes in policies, regulations, and governance.

While some improvements in environmental performances might be a consequence of a stricter regulation at European and national level, the authors claim that better and more homogeneous performances resulted from a centralised regulation at regional level, pursuing circular economy strategies. Furthermore, according to the paragraph 4.3.3, the dynamic optimisation of the waste flows to existing plants allowed a substantial rationalisation and reduction of plants, besides a considerable minimisation of landfill use as final destination, favouring the shift towards energy recovery of unsorted waste.

Thanks to the economic regulation, an increased homogeneity of economic variables among Municipalities was fulfilled, both considering total WM cost and CTS, according to a specific regional regulation. This result is confirmed by the comparison with other Italian regions. The authors claim that the increased territorial scope of the regulatory authority contributed to this result, by enabling data collection from different data sources and benchmark among a higher number of waste service providers.

As far as the contracts granted to MSW providers are concerned, the regional level seems to be more suitable to these kinds of decisions than the previous provincial one. The decision process about the waste service entrustment guarantees the safeguard of municipalities instances, but in full compliance with a stronger independence of the regulatory agency.

Thanks to the centralisation of the regulation, more complete reports on waste management were made available for citizens and uniform information were disclosed to public.

The results of the environmental assessment show that prevention of UW is not completely fulfilled at regional level, although the commitment of the region to prevention and reuse is strong. This evidence suggests that waste prevention is a long-term process, affected by multiple exogenous factors (firstly, cultural habits and industrial design, in the authors' opinion). Long term policies and step-by-step improvements are required to achieve actual results.

A focus on the transition to PAYT systems should be done by policy makers, in this framework about prevention policies. Even though the first results from the municipalities in which PAYT systems have been implemented suggest that per capita waste production is 10%-15% lower than the average production in the region, the regional target on waste production (set in the regional WM plan) is far from being achieved. Moreover, the effectiveness of PAYT schemes on waste reduction seems to be mostly affected by how the system is designed (Magrini et al., 2020).

On the other hand, waste production and treatment concern regional policies which have to be incentivised and integrated in different production sectors and actors, firstly including waste service providers. Moreover, thanks to the transition to a regional OTA, the policies on WM and planning might be better integrated in policies about other topics of regional responsibility, e.g. industrial policy, urban policy, tourism management, in a circular economy approach. National guidelines could help in this process, leading to a more standardized policy framework. As already claimed by OECD (2020), to achieve circular economy goals, subnational governments need to be supported by an enabling framework that national governments can establish effectively.

Besides the existing target at Municipal level on separate waste collection rate, monitoring waste production at Municipal level could be useful to increase the awareness of the local level on waste prevention. The parameter "Inhabitant Equivalent" might be used in order to make comparable the different Municipalities, to provide a more detailed analysis and to support the policy-makers.

A national multidisciplinary approach could also lead to the chance of comparing different regions, both for analyses concerning waste production and prevention, and for economic and financial analyses. In this respect, the implications from the recent institution of ARERA (in 2018) should be discussed in further research studies. Indeed, different regulatory models are in place in Italy: this highlights that efficiency and economies of scale/scope/density are not the only criteria impacting on this decision, but the political context plays a role.

Despite the institutional transition described in this paper, economic and waste service characteristics are still quite affected by decisions at municipal level because of the legislative competences of Municipalities in some fields, such as in the tariffs management, the essence of users charges and the adoption of municipal guidelines. The balance of these competences with a more industrial approach to the MSW management is still challenging.

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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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CISA

Research to Industry and Industry to Research

CHALLENGES AND PERSPECTIVES OF DIRECT TEST METHODS FOR ASSESSING WASTE HAZARDOUS PROPERTIES (HP)

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Waste generation is a global problem, not only environmentally but also the economic loss it represents. Annual waste generation is projected to increase by 70% by 2050 (OECD, 2019a). Consequently, waste management should be planning and managing a circular economy, to ensure that resources used remain in the EU economy for as long as possible, while ensuring the best degree of environmental safety (European Commission, 2020). In this context, hazard waste classification plays a crucial role.

Hazard waste classification entails safe handling and disposal of discarded materials, with significant impacts on waste producers' budgets, their legal conduct, and public perception. The regulatory criteria should be realistic and scientifically sound ensuring full transparency while providing a level-playing field for all industrial sectors. According to European regulations, waste is defined as hazardous if satisfies at least one of the 15 hazard properties (HP) or contains concentrations of certain persistent organic pollutants over specific legal thresholds (European Commission, 2014; European Parliament and European Council, 2019). Equally, wastes are classified as hazardous according to the 6-digits codes enlisted in the European Waste Catalog, established by the European Commission, (2000). Accordingly, among "absolute non-hazardous" waste, "absolute hazardous" waste, and the so-called "mirror entries" (i.e., waste streams potentially classified as hazardous by their composition), only these latter require an effort to assess specific HPs.

HPs can be assigned by an "indirect" approach, from the total content of hazardous substances (selected according to "expert judgment"), or a "direct" approach, which relies on outcomes of single HP-specific laboratory tests (European Commission, 2014). Based on widespread analytical methods, the "indirect" approach is cheap and currently the most adopted. Notably, it is characterized by some challenges: the subjectivity of the "expert judgment," the impossibility of detecting all substances and elements that compose the waste material, and the so-called "worst-case" approach, which considers the waste constituents detected as in the most hazardous form (Bishop and Hennebert, 2021; Hennebert, 2019). These drawbacks have been limited by the development of non-targeted organic and mineral analyses, giving an analytical mass balance > 90% (Hennebert et al., 2013), and the speciation of so-called "worst-case with information" pre-calculated approaches (Hennebert, 2019). However, the classification as hazardous can sometimes be judged as incomplete or unrealistically conservative. In these cases, specific testing methods to evaluate "directly" (i.e., without further assumptions) different HP-related effects, closely associated with the real speciation (and environmental fate) of waste constituents. The European legislator suggested the latter approach given the information about the waste composition is not sufficient for a correct evaluation. The European legislation affirms that direct test results will prevail over the results from chemical composition analyses (European Commission, 2018). The EU law-maker also suggests the methods used to be guided by the CLP regulations for performing direct testing, toward the harmonization of products and wastes law frameworks (European Council, 2008).

There are still some challenges to be faced:

- A limited number of laboratories are accredited for the methods available, increasing in costs but not in use;
- The methods designed for classifying products under CLP Regulation (European Council, 2008) can be unsuitable for testing wastes;

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TABLE 1: Approaches available for assessing waste Hazard Properties.

Hazard Property		Evaluation approach
HP 1	Explosive	Direct
HP 2	Oxidising	Direct
HP 3	Flammable	Direct
HP 4	Irritant – skin irritation and eye damage	Indirect and Direct
HP 5	Specific Target Organ Toxicity	Indirect
HP 6	Acute Toxicity	Indirect
HP 7	Carcinogenic	Indirect
HP 8	Corrosive	Indirect and Direct
HP 9	Infectious	Not available
HP 10	Toxic for the reproduction	Indirect
HP 11	Mutagenic	Indirect
HP 12	Release of an acute toxic gas	Direct
HP 13	Sensitising	Indirect
HP 14	Ecotoxic	Indirect and Direct
HP 15	Waste capable of exhibiting a hazardous property listed above not directly displayed by the original waste	Indirect

- For some HPs, there is no EU-harmonized list of direct test methods;
- The need to validate in-vitro methods as alternatives to in vivo and ex-vivo animal testing.

Experts from analytical and industrial sectors, with representatives of environmental research institutions, discuss the state-of-the-art direct test procedures while building an efficient and correct waste classification framework. The meeting happened during a specific workshop session held virtually during the SUM Symposium (Fifth Symposium on Urban Mining and Circular Economy Virtual Event/18–20 November 2020). Subsequent meetings involved experts to further widen the main topics discussed during the symposium and are presented here.

This paper describes the most discussed direct decision trees, namely, the methods for assessing HP 4 (Irritant), HP 8 (Corrosive), and HP 14 (Ecotoxic). These HPs can be assessed with the "indirect" and the "direct" approaches, while the remaining properties are evaluated using the calculation method or direct testing only (Table 1). Finally some suggestions prompting the introduction of validated test methods for HP 10 (Toxic for reproduction) are included as an example of the research path taken to provide data for future regulation updates.

HP 4 (irritant) and HP 8 (corrosive)

Both HP 4 and HP 8 have potentially negative effects on human tissues (i.e., skin and eyes), but with an increasing degree of severity, from reversible damages for HP 4 to irreversible injuries in the case of HP 8 (European Commission, 2014). According to experts interviewed, the procedure for evaluating HP 4 and HP 8 using direct testing is harmonized and well-established at the EU level, which is based on validated in-vitro test methods that are in the EU technical guidelines for waste classification (European Commission, 2018).

The decision tree consists of a combined assessment of outcomes from the conventional acid/alkali reserve test and *in-vitro* assays. Practically, only wastes characterized by inherent pH ≤ 2 or ≥ 11.5 can be classified with HP 8 or HP 4. In this case, if the buffer capacity is high (i.e., (pH - 1/12 acid reserve (g NaOH/100g)) < -0.5 or ((pH + 1/12 alkaline reserve)) > 14.5; or, equivalently, (pH - 1/3 buffer capacity (mol H+/kg)) < -0.5 or (pH + 1/3 buffer capacity (mol H+/kg)) > 14.5) the waste is classified with HP 8. Instead, waste characterized by inherent pH ≤ 2 or ≥ 11.5 but showing low buffer capacity can undergo further in-vitro testing for HP 4 and/or HP 8 classification. Both pH and buffer capacity are measured following the instructions of OECD 122 (OECD, 2013a). In particular, for solid wastes, the established method requires inherent pH and acid/alkali reserves to be measured in a 1% (w/v) (i.e., 10 g/L) solution with distilled or deionized water (OECD, 2013a). Another approach is to use the classical batch waste leaching test according to EN 12457 series, typically with 100 g/L (CEN, 2004).

A stepwise process was proposed to decrease the costs related to performing in-vitro tests (Figure 1). In-vitro assays related to the "irritation and eye damage" endpoint (i.e., HP 4) should be performed before skin corrosiveness tests (i.e., HP 8). If waste is not classified as HP 4, it cannot be classified also as HP 8, while the contrary is not true. This procedure ensures that mirror entries not requiring any classification for HP 4 and HP 8 undergo just one round of in-vitro tests (i.e., for HP 4 assessment).

The in-vitro test method for assessing skin irritation (i.e., OECD 439) is performed with a time-limited application of the material tested on models of reconstructed human Epidermis (RhE) (OECD, 2013b). Any irritation potential is identified by the loss of cell viability over a specified threshold, by indirectly measuring the enzymatic activity of the RhE model after exposure. Similarly, in-vitro assays for skin corrosion are conducted according to OECD 430, from the exposure of epidermal surfaces of rat skin disks (OECD, 2013c) or, more preferably, by OECD 431, through

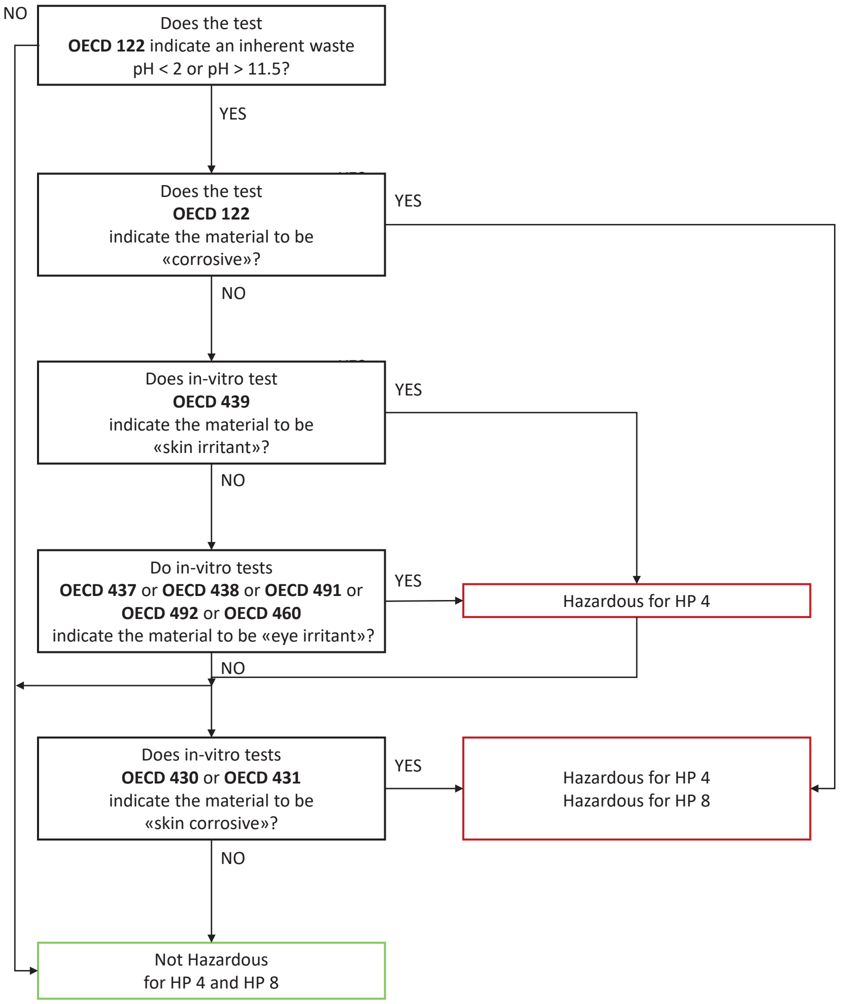

FIGURE 1: Proposed decision tree for waste HP 4 and HP 8 classification by direct tests.

exposing RhE models (OECD, 2015). Both methods measure the consequent response in the loss of cell viability and integrity. Specifically, in-vitro tests for evaluating skin irritation can use the same RhE models used to test in-vitro skin corrosion. However, skin corrosion is assessed by different procedures and classification limits than skin irritation.

Furthermore, the proposed approach includes in-vitro tests for assessing "eye irritation or serious eye damage" since waste having such effects falls within the definition of HP 4 (European Commission, 2014). However, the methods suggested are excluded in either the EU technical guidelines on waste classification or the regulation of analytical methods allowed for CLP (ECHA, 2017; European Council, 2008). However, to strictly adhere to the definition of HPs and in the absence of alternative tests, five valid OECD in-vitro test methods are used to address "eye irritation or serious eye damage," and are consistently suggested in this study (Figure 1). All in-vitro assays considered are performed by placing the waste test portion on biological layers reconstructed and cultured from bovine cornea cells as in OECD 437 (OECD 2020a), chicken eyes as in OECD 438 (OECD 2018), rabbit cornea cells as in OECD 491 (OECD, 2020), human cornea-like epithelium as in OECD 492 (OECD, 2019a), and canine kidney as in OECD 460 (OECD, 2017). Waste classification can then be assigned by comparing the chemical-induced damage measured at the end of each test.

HP 10 (toxic for reproduction)

Waste is classified as HP 10 if it has negative effects on the reproductive functions of adults or the sexual development of offspring. No strategy for directly assessing this property is within the EU waste law framework. In particular, technical guidelines for waste classification state that "there are limited options for testing reproductive toxicity properties in-vitro." (European Commission, 2018).

From experts, several in-vitro tests are validated and suited to classify chemical products for the category "reproductive toxicity." Other in-vitro methods are under investigation with a view to full regulatory acceptance. A full list of in-vitro tests useful for the classification of substances and information on their acceptance status is in the EURL ECVAM dataset on alternative methods to animal experimentation (DB-ALM) (European Commission and Joint Research Center, 2019). However, pending the transposition to the related waste regulations, only the calculation method should be applied (European Commission, 2014).

A possible solution is a direct decision tree based on already validated in-vitro tests. With the approach developed for chemical products, a multi-step analytical strategy should be performed for a reliable classification of reproductive toxicity. In particular, the biological concept of "reproductive cycle" has been broken down into three elements that are estimated by the combined assessment of three biometabolic effects, namely, loss of male fertility, decrease of implantation capacity (i.e., female fertility), and hindered prenatal development (Hareng et al., 2005). A specific battery of in-vitro assays can also be proposed for waste, whose results cover the sub-endpoints range identified.

Research that focuses on solving the critical issues that occur when applying the validated in-vitro methods for waste characterization is our interest. The latter relates to the adequacy and adaptability of methods to the heterogeneous nature of wastes compared to the more homogeneous features of chemical products. Solubility mechanisms and solid wastes extraction conditions appear the most challenging to be solved before full adoption of available in-vitro assays.

Similar considerations are also valid for direct testing of HP 7 (carcinogenic) and HP 11 (mutagenic) classification of wastes.

HP 14 (ecotoxicity)

Despite the extensive debate on the ecotoxicity classification of wastes, there is no agreement among scientific and industrial communities on the most suitable decision tree to be applied. In this regard, efforts toward a widespread agreement and harmonization at the EU level are essential, since HP 14 "Ecotoxic" is the most frequent HP classifying mirror entries as hazardous (Hennebert et al., 2014). Consequently, the EU regulation does not precisely define the direct test methods but considers methods for CLP appropriate (i.e., European Council, 2008), with "other internationally recognized test methods and guidelines" (European Council, 2017). This has contributed to to establish a non-harmonized framework of regulations among EU Member States.

Each decision tree for the ecotoxicological classification of waste is built on three main elements: the sample preparation procedure such as the leaching tests for preparing liquid samples from solid test portions; the battery of bioassays to be performed; finally, the reference thresholds triggering hazardous classification. If at least one ecotest is non-compliant with the limits, the waste is classified as "ecotoxic". Two main approaches emerged from the discussion.

First, the "CLP-based approach," based on a protocol issued by the Italian Institute of Environmental Protection (ISPRA, 2018) and included in the guidelines for waste classification (SNPA, 2020), entails only methods and classification limits developed and validated for chemical products within the CLP regulations, regarding exclusively aquatic toxicity (ECHA, 2017; European Council, 2008). The method for liquid test sample preparation from solid samples (i.e., leaching test) was originally developed for dissolving metal compounds to an aqueous medium, i.e., OECD 29 (OECD, 2001). Solid waste eluates, termed Water Accommodated Fractions (WAF), are produced through a leaching test characterized by loading rates of 100 mg/L, 10 mg/L, and 1 mg/L and tested individually (i.e., without further dilutions) using the battery of biotests. Thus, the concentration limits, indicated in terms of EC50 and NOEC, are expressed in terms of "loading rate" (i.e., mg/L). The bioassays that are composed of the test battery proposed for this approach are listed in Table 2 together with the corresponding concentration limits triggering an HP 14 classification. A potential decision tree adopted following this approach is also depicted in Figure 2.

Second, the "Waste-based approach," adopts internationally acknowledged methods developed and validated specifically for waste testing, unlike CLP-related methods (CEN, 2005; Moser and Römbke, 2009; Pandard and Römbke, 2013). The test battery proposed includes terres-

TABLE 2: "CLP-based approach." Battery of biotests and concentration limits used within the testing strategy complying with the CLP Regulation.

Organism	Type	Standard	Classification Criteria (waste is hazardous for HP 14 if)	Source
Algae	Acute-Chronic ***	OECD 201 * (Freshwater Alga and Cyanobacteria, Growth Inhibition Test)	Acute LC50 ≤ 100 mg/l	(OECD, 2011)
	Chronic	OECD 221* (Lemna sp. Growth Inhibition Test)	Chronic NOEC ≤ 1 mg/l	(OECD, 2006)
Crustacean	Acute	OECD 202 * (Daphnia magna, Acute Toxicity Test)	Acute LC50 ≤ 100 mg/l	(OECD, 2004)
	Chronic	OECD 211 * (Daphnia magna, Chronic Toxicity Test)	Chronic NOEC ≤ 1 mg/l	(OECD, 2012)
Fish	Acute	OECD 203 * (Fish, Acute Toxicity Test)	Acute LC50 ≤ 100 mg/l	(OECD, 1992)
	Chronic	OECD 210 ** (Fish, Early-Life Stage Toxicity Test)	Chronic NOEC ≤ 1 mg/l	(OECD, 2013)

** Method included in the list of tests validated within the scope of the CLP regulation (European Council, 2008) and SNPA, (2020).*
*** Test not reported within European Council, (2008) and SNPA, (2020), but present in ECHA, (2017).*
**** According to ECHA, (2017) "The algal growth inhibition test is a short-term test that provides both acute and chronic endpoints. However, EC50 is treated as an acute value for classification purposes."*

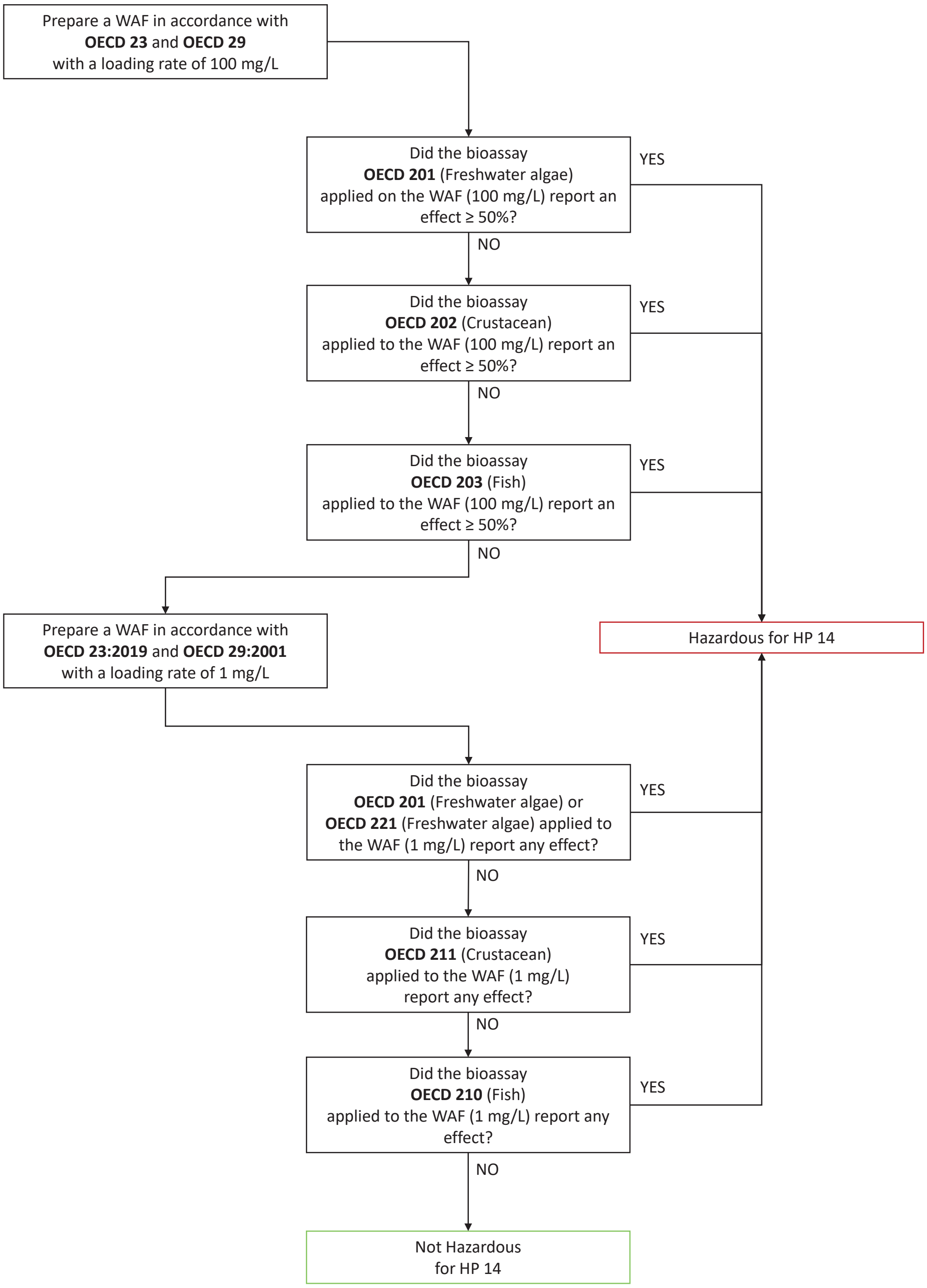

FIGURE 2: "CLP-based approach." Proposed decision tree for HP 14 classification of waste with methods validated for CLP. WAF: Water Accommodated Fractions. OECD 23: Guidance Document on Aquatic Toxicity Testing of Difficult Substances and Mixtures; OECD 29: Guidance document on transformation/dissolution of metals and metal compounds in aqueous media; OECD 201, 202, 203, 210 and 211: see Table 2.

trial and aquatic tests, listed in Table 3. Test portions are prepared following the waste-specific standard EN 14735 (CEN, 2005). In particular, aquatic bioassays are conducted on several volumetric dilutions of solid waste eluates from a leaching test adopting a loading rate of 100 g/L, which corresponds to a Liquid-to-Solid ratio of 10 L/kg, conducted as prescribed in EN 12457–2 (CEN, 2004). Concentration limits, expressed as EC50 (i.e., %vol/vol), were proposed for this approach by Hennebert, (2018). A proposed decision tree is shown in Figure 2. A practical application to the HP 14 classification of typical mirror entry waste (i.e., Automotive Shredder Residue) is also proposed in Pivato et al., (2020).

A discussion occurred among the participants about the comparison between the two approaches.

In particular, it is worth mentioning that the water extracts are derived according to two distinct sets of applied conditions (L/S ratio, particle size, test duration, leaching media etc.), simulating a wide range of occurred leaching conditions.

The size and the granulometry of the solid test portions indicated by the leaching methods were discussed: despite the significant efforts to reduce particle size up to 1 mm and massive subsampling (from the laboratory sample to 100 mg test portion), test portions can seldom be thought of as representative of the waste batch to be classified. From this, the application of the validated standard EN 14735, recalling the instructions laid down in EN 12457–2, can solve the issues discussed.

Finally, other challenges were specified during the discussion, whose solutions can lie within the outcomes of future research in the field:

- Waste water extracts frequently feature extreme pH values, both lower and higher than the survival range of organisms used in the test batteries suggested (i.e., pH range of 6–9). Therefore, the results of bioassays can be predicted without performing the tests. Further research to identify more roles of pH (and its adjustment in the second set of repeated tests) on aquatic toxicity mechanisms for each matrix tested. Regulations and specific technical standards are open to adjusting the pH of the sole test portion (i.e., not the dilutions) if extreme pH influenced the outcomes of the biotests performed. Nonetheless, pH adjustment should be reported in any test reports produced, together with results of biotests without pH adjustment.
- From Table 2, several methods to obtain chronic endpoints are suggested only by the guideline on the application of CLP criteria (ECHA, 2017), but are neither included in the list of methods validated for CLP (European Council, 2008) nor the Italian guideline for waste classification (SNPA, 2020), which applies a CLP-consistent approach for HP 14 assessment. Similarly, no chronic endpoints can be assessed by the abovementioned testing approach described in Table 3 and Figure 3, consistent with the propositions of Pandard and Römbke, (2013). This is due to the efforts required by the main laboratories for chronic testing that hindered its use for conventional purposes like waste classification.
- High intra-laboratory and inter-laboratory variability characterize outcomes of the aquatic ecotoxicological bioassays of the test batteries. No data on repeatability and reproducibility are available in the scientific literature or institutional reports for terrestrial tests (Hennebert and Beggio 2021). This issue is crucial for adopting a conventional harmonized decision tree for the HP 14 classification of waste. Besides the intrinsic variability of the method, the low reproducibility and repeatability can be related to the preparation of the test portion analyzed, highlighting the influence of the specific leaching test adopted. This issue was evident in the algae test conducted on waste WAF prepared following the CLP-leaching test. In particular, an Italian institutional report, resulting from inter-laboratory tests conducted according to ISO 5725–2 (ISO, 2019) at 23 laboratories on a prepared 100 mg/L WAF, calculated relative repeatability and reproducibility values for the algae test as 78% and 104%, respectively (ISPRA, 2017). If the results of repetitions of that test are normally distributed, the confidence interval of the mean with a 95% probability level [mean – 1.96*standard deviation; mean + 1.96*standard deviation] includes zero and negative values. Therefore, the results cannot be significantly different from zero nor used for regulatory purposes.

Conclusions and future trends

In the current work, a state-of-the-art method on the most important direct tests for waste classification is presented. Drawbacks are discussed and proposals suggested in a systematic and organic approach.

The recommended test(s) to assess a given hazard property, which covers all aspects of that specific hazard

TABLE 3: "Waste-based approach." Eco tests and corresponding limits for HP 14 classification of waste according to (Hennebert, 2019; Hennebert, 2018; Pandard and Römbke, 2013; Moser and Römbke, 2009).

Organism	Type	Standard	Classification Criteria (waste is hazardous for HP 14 if)[1]	Source
Aquatic Bacteria	Acute	EN ISO 11348-3 (Determination of the inhibitory effect of water samples on the light emission of Vibrio fischeri (Luminescent bacteria test) - Part 3: Method using freeze-dried bacteria)	EC50 < 15%(vol/vol) (original data 15.8%)	(ISO, 2008)
Algae	Acute	EN ISO 8692 (Fresh water algal growth inhibition test with unicellular green algae)	EC50 < 10%(vol/vol) (original data 7.03%)	(ISO, 2012a)
Crustacean	Acute	EN ISO 6341 (Determination of the inhibition of the mobility of Daphnia magna Straus)	EC50 < 10%(vol/vol) (original data 7.95%)	(ISO, 2012b)
Soil Bacteria	Acute	EN ISO 18187 (Contact test for solid samples using the dehydrogenase activity of Arthrobacter globiformis)	EC50 < 5%(w/w) (original data 2.25%)	(ISO, 2018)
Plants	Acute	EN ISO 11269-2 (Determination of the effects of pollutants on soil flora - Part 2: Effects of contaminated soil on the emergence and early growth of higher plants)	EC50 < 15%(w/w) (original data 13.7%)	(ISO, 2013) (ISO, 2013) (ISO, 2013)
Soil Invertebrates	Acute	ISO 17512-1 (Avoidance test for determining the quality of soils and effects of chemicals on behavior. Test with earthworms)	EC50 < 5%(w/w) (original data 3.75%)	(ISO, 2020)

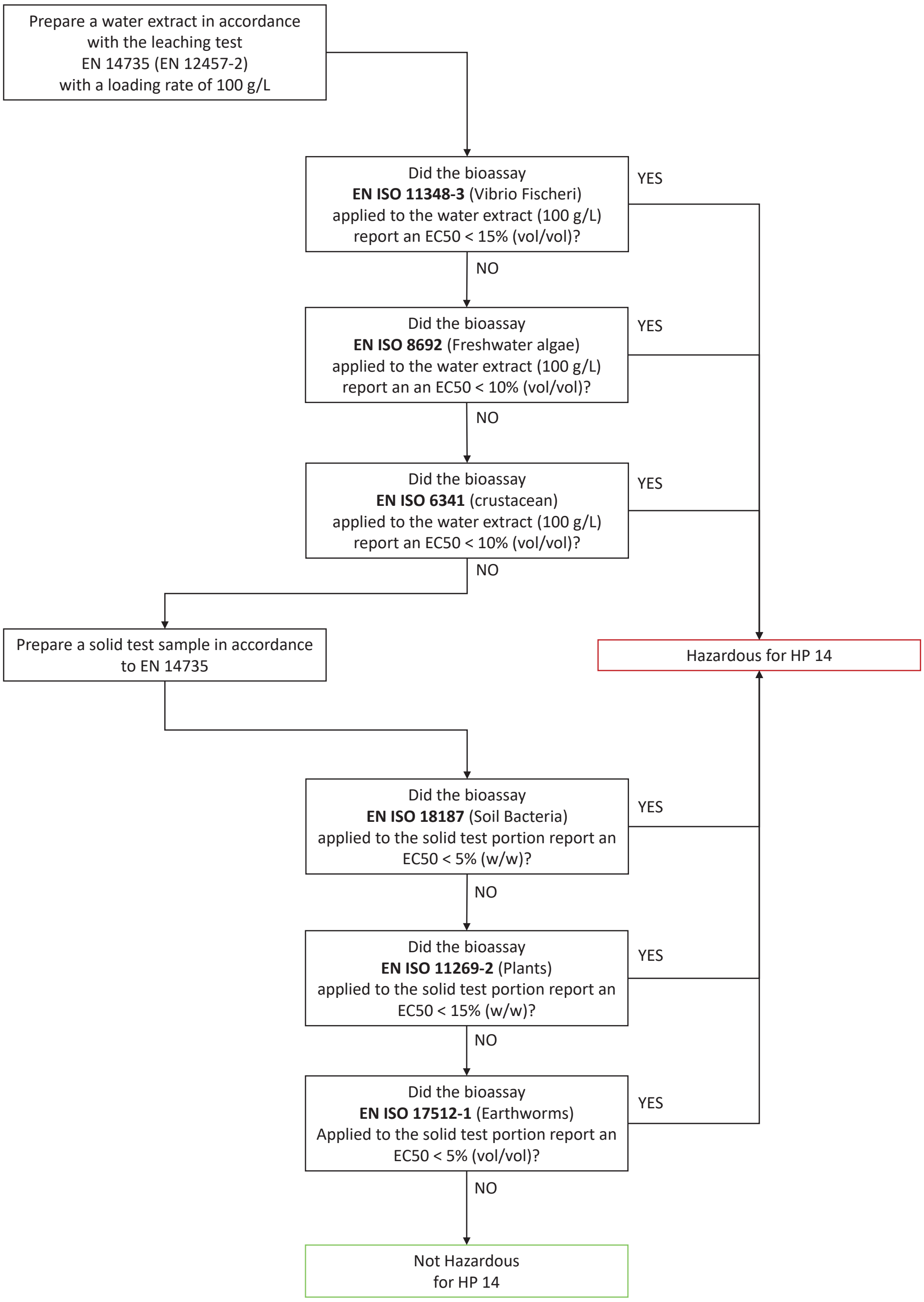

FIGURE 3: "Waste-based approach." Proposed decision tree for HP 14 classification of waste according to the international standards proposed in Pandard and Römbke, (2013) and the classification limits established in Hennebert (2018, 2019).

is agreed upon by lab specialists. For instance, the six tests proposed for HP 14 are non-correlated, indicating different functional toxic actions in the living organisms. The choice of concentration limits or thresholds that trigger the hazard classification is a distinct subject. Limits must be harmonized and conventional, when dealing with intrinsic hazard and not with risk. They cannot be derived for a given scenario (i.e., emission-transfer-target) of a given use or fate of the waste. Site-specific scenarios are different, and the corresponding thresholds would be multiple too. Thus, the objective is not just to achieve a "zero effect" but to consider an "acceptable" effect. The best solution is achieved by aligning the concentration limits or thresholds of the tests to match the resulting classification with an existing and well-established reference. This has been done for pH and buffering capacity for irritancy and corrosiveness by the UK industry of soaps and detergents (Young et al. 1988, 1994) and for HP 14 (Hennebert 2018). This approach is also valid when proposing ecotoxicity classification of fertilizers from the circular economy (e.g., digestates, composts). For this latter, an ecotoxic effects at the agronomic rate can be considered acceptable if its results are not higher than that caused by millenary-used manure, organic matter, and some minerals at their respective agronomic rates. From this standpoint, a consensus can be reached, and the implementation of the regulations improved.

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

by Rainer Stegmann

Artists seldom provide an interpretation of their own work; they leave this to the observer. Each of us will have his/her own individual view of a specific piece of art, seeing different contents and experiencing a range of own feelings and emotions. Bearing this in mind, I created this page where you will find regularly selected masterpieces from different epochs and I express my thoughts on what the work conveys to me personally. My interpretation will refer specifically to the theme "Environment". Any comments or suggestions regarding this column should be addressed to stegmann@tuhh.de.

HUBERT ROBERT / Demolition of houses on The Pont au Change (1788).

A colleague who read my Art Column made me aware of this painting. It is different from the ones I presented before because it shows directly its connection to waste. I regard it as a painting showing recycling of construction and demolition waste (C+D waste). We see some remnants of the demolished houses covered with rubble. As materials wood beams and larger stone plates were separated and prepared for transport and reuse.

Recycling of construction materials has a long tradition going back to high cultures some thousand years BC. An example from younger periods is a reused Byzantine capital from post antique times adjacent to a Hafsidic one in the Moscque of Kairouen, Tunesia, which was rebuilt from the 11- Century.

In Germany after the 2. World war huge amounts of materials from destroyed buildings were sorted by the so-called "Rubble Women" as a basis for rebuilding damaged houses. Later in the 70s -90s less C+D waste was re- used; it was mainly co-disposed with residential waste which filled up the landfills quite fast. To divert C+D waste from landfills the disposal prices were significantly increased, which made recycling more economical attractive.

In many countries C+D waste recycling is common practice where up to 70 to 80% is recycled. The materials are mainly used for noise protection walls, road construction and concrete production. Before buildings are destroyed doors, windows, insolation material, electrical devices and plastic materials are removed. The concrete, bricks and tiles are crushed and sieved in several steps and mainly mineral materials of different grain size are produced; unwanted materials are often manually removed.

For recycling of walls, floors, roof tiles, bricks and other installations prefabricated houses should be constructed in a way considering future dismantling. Brunner, 2011 calls such an approach urban mining. Due to the implementation of the prefabricated building concept in the former DDR entire concrete walls could be removed and were partly reused.

Since C+D waste makes up in many countries the largest waste fraction and since the availability of construction materials as e.g. sand is decreasing C+D waste recycling is coming more in the focus of construction industry.

Coming back to the painting and to our situation today the main driver for recycling are shortage economy, cost and availability of raw materials as well as availability of landfills. Unfortunately, reasons as environmental protection and saving natural resources are seldom the reasons.

REFERENCE

Brunner, P. H., 2011, Urban mining as contribution to reindustrializing the city, Journal of Industrial Ecology

The next artwork will be a photo installation of the photographer Fabrice Monteiro and the fashion designer Jah Gal from Senegal: "The Prophecy #1", 2013. Monteiro was born in 1972 in Belgium and has a Benin father and a Belgium mother, he lives in Dakar , Sengal.

FABRICE MONTEIRO AND JAH GAL / The Prophecy #1 (2013).

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CONTENTS

Editorial

Biowaste management

Construction and demolition waste

WEEE management

Plastic recycling

Pyrolysis

Landfilling

Waste management practices

Columns

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