Università degli Studi di Padova

Head Office: Università degli Studi di Padova

Department of Civil, Environmental and Architectural Engineering

---

Ph.D. COURSE IN: Sciences for Civil, Environmental and Architectural Engineering

CURRICULUM: Risk, Vulnerability, Environment, Health, and Territory

SERIES: XXXV

TOOLS FOR SUSTAINABLE LANDFILLING IN THE FRAMEWORK OF CIRCULAR ECONOMY

Coordinator: Prof. Carmelo Maiorana

Supervisor: Prof.ssa Maria Cristina Lavagnolo

Co-Supervisor: Dr. Valentina Grossule

Ph.D. student: Qingliu Luo

SEDE AMMINISTRATIVA:	Università degli Studi di Padova Dipartimento di Ingegneria Civile, Edile, Ambientale
CORSO DI DOTTORATO DI RICERCA IN:	Scienze per l'ingegneria civile, edile, ambientale 35^{th} Cycle
PhD Programme Coordinator	Prof. Carmelo Maiorana DICEA-Department of Civil, Environmental and Architectural Engineering University of Padova
Supervisor:	Prof.ssa Maria Cristina Lavagnolo DICEA-Department of Civil, Environmental and Architectural Engineering University of Padova
Co-supervisor	Dr. Valentina Grossule DICEA-Department of Civil, Environmental and Architectural Engineering University of Padova
Reviewers:	Prof. Mario A. Gandini Facultad de Ingeniería Universidad Autónoma de Occidente Calle 25 No. 115-85 Cali - Colombia PBX: +57-2-3188000 Ext. 11315 Correo electrónico: magandini@uao.edu.co Prof.ssa Cristina Trois School of Engineering University of kwaZulu-Natal Centenary Bld Howard College Campus Durban, 4041, South Africa Tel. +27 (0)31 260 3065 Email: troisc@ukzn.ac.za; cristina.trois@gmail.com
Candidate:	Ing. Qingliu Luo DICEA-Department of Civil, Environmental and Architectural Engineering University of Padova

Abstract

Sustainable landfilling plays a fundamental role either in providing an economic option for municipal solid waste management or in closing the materials loop of the Circular Economy (CE). A landfill is considered sustainable when the emission potential generated from landfills poses no environmental risk and the quality of emissions achieves the Final Storage Quality (FSQ) within 30 years. The goal of this thesis was to pre-treat waste before landfilling for shortening the time achieving the FSQ and assess the sustainability of a landfill. Thus, a three-year research programme with painstaking efforts was carried out based on two main lab-scale works: washing pre-treatment and column leaching test. Waste washing study evaluated the effectiveness of waste washing prior to landfilling on reducing the emission potential and long-term impacts of three different kinds of residues from municipal solid waste treatment on the environment. Results gained from washing tests demonstrated that washing pre-treatment could stabilise the landfilling waste by removing readily leachable contaminants and reduce long-term emissions as well as shorten the time reaching FSQ limits significantly. Column leaching tests investigated the influence of irrigation quantity and frequency on time achieving the FSQ and emission potential. Results obtained revealed that values from column leaching tests could provide more representative information in assessing the sustainability of a landfill and could simulate the real emissions occurring under landfill conditions. Irrigation frequencies related to column leaching tests had a significant impact on the reduction of waste emission potential, and it could be taken into account to improve the in-situ treatments.

Table of Contents

LIST OF FIGURES

LIST OF TABLES

1. Background

1.1 The Role of Sustainable Landfilling in Circular Economy

Circular Economy (CE) has replaced the traditional linear economy (take-make-waste) to provide a global strategy for current solid waste management (SWM). The linear traditional approach based on the extraction of raw materials, production, use, wasting and landfilling is obsolete and progressively abandoned (Tisserant et al., 2017; Zeiss et al., 2021). The circular approach primarily arises from a growing need for primary raw materials, as a consequence of global economic development. Attention is currently shifting from the limited and fixed stocks of raw materials to the increasing anthropogenic stocks of materials. This creates the base for the development of different strategies for recovering of resources from waste (urban mining, circular economy, etc.), mainly aiming at reducing the consumption of non-renewable resources, recirculating the materials and minimising waste generation. In the concept of CE, the circular approach of turning the generated waste into resources by using proper methods solves the issues about the accumulative generation of residues that could not be addressed in a traditional linear approach.

The emphasis placed in CE mainly focuses on the role of recycling, turning residues into resources depending on the recovery of energy and material to close the material loop, which is commonly described as a perfect cycle. Although the close cycle where the different life stages of a product are represented: production – distribution – consumption - reuse/repair - recycling of secondary raw materials to production, can be described as a perfect solution for addressing the issues about resources and waste, a critical problem is ignored in the loop: where is the destination of non-renewable residues, namely, the role of landfilling is completely missing.

In the waste hierarchy, a top-down triangle hierarchic set of actions which consists in: Prevention, Preparation to Reuse, Recycling and Final disposal, demonstrating a highlight on the first three types of waste management, but a quite ignoring attitude towards final disposal. Consequently, the underestimated attitude toward final disposal leads to misinformation of public opinion: landfill is no longer needed in circular waste management. In particular, landfilling has been regarded as an “obsolete and polluting” system that should be abandoned (Cossu et al., 2020a), which totally denies its fundamental but important role in waste management.

Although landfills play the role of a dustbin in the system and even the final dustbin is missing in the closed material loop of CE, the negligence leads the politicians and the population to underestimate a series of aspects dramatically, such as:

1. not all the waste can be effetely converted into recycling resources;
2. endless waste recirculation is not possible;
3. even recycling activities generate residues;
4. hazardous substances accumulate in recycled materials;

Obviously, landfills continue to play a fundamental role in deposing the non-recycling residues in the waste management system. However, traditional landfills can no longer meet the desire for pursuing the sustainability principle due to their both short and long-term emissions. Building a sustainable waste management strategy is emergently needed not only in terms of technology but also in the needs of practices. Thus, measurements should be taken on the regulatory level to improve the sustainability of the system, assuring a final sink to close the material loop and immobilize the contaminants.

Both in the concept of CE and waste hierarchy, the recycling process is highlighted due to focusing on converting waste into resources and reducing the consumption of raw resources. Recycling process in waste management can be outlined in four distinct stages: extraction, selection, refinement and production, and each stage is able to generate various types of wastes (gaseous, liquid and solid), leading to an enormous environmental burden (Demirbas, 2011; Paritosh et al., 2017; Romano et al., 2019). In particular, a considerable amount of the solid non-renewable residues generated in those stages eventually end up in a final sink: landfills, even though landfilling of residues is not considered in the close loop of CE. All those residues generated in the different stages immensely need to be treated in order to immobilize the potential pollutants and avoid contaminating the environment.

Since it has been well-known that "zero waste" is impossible, and at least "zero waste" to date cannot come true in terms of technology and theory, landfills as the final "dustbin" collecting all non-recycling residues should be designed to achieve the sustainability principle. Even though the final sink could be a "temporary sink" instead of a "permanent sink" due to the possibility of reusing the "buried final resource" in the future, the concept of sustainability must be applied to build a landfill in the waste management system. Thus, Back to the Earth Sites (BES) needs to be identified. BES is intended to store the residues generated by the circular economy in a non-mobile and stable form to close the material loop (Cossu, 2016).

The role of the final sink in the waste management hierarchy must be taken by the landfill. To achieve the goal of closing the material loop, sustainable landfills should replace the ‘‘nasty and unsightly” landfills to keep the waste materials back to the earth sites in a safe and sustainable way. As shown in **Figure 1. 1**, the role of sustainable landfill should be presented in closing this circular approach instead of being missing.

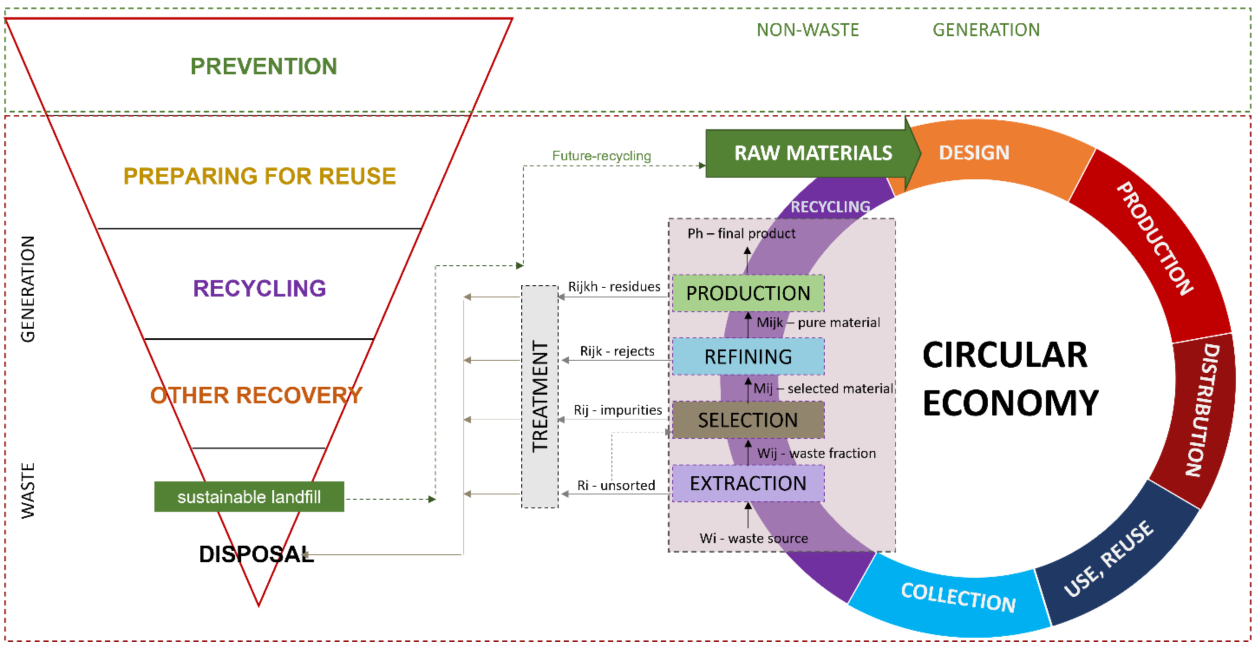

Figure 1. 1. The important role of sustainable landfills in circular economy, modified from the previous studies (Cossu et al., 2020a; Mangesh et al., 2015) (W: Waste, M: Valuable materials, R: Residues, P: Products) (i: different waste source, j: individual recovered fraction, k: pure materials from the individual recovered fraction, h: individual type of product)

The concept of sustainability applied to landfills aims to provide a safe sink to store the “temporary waste”, and also close the loop of CE by returning waste to their non-mobile state, as they were before they were extracted from the ground to be used as raw materials. The final storage quality (FSQ) is taken into the consideration in sustainable landfills to assure sustainability principle. In sustainable landfills, treatment measurements (pre-treatments, in-situ and on-site treatments, etc.) are taken on non-recycling residues to remove or reduce leachable contaminants in order to enhance the quality of landfilling wastes. Multi-barriers (physical barriers) are taken into consideration in landfill design to prevent long-term emissions in order to reduce the burden on surrounding environments. Real-time monitoring and aftercare measurements are applied to keep landfills running in a sustainable way.

The abovementioned aspects demonstrate the importance of applying sustainable concept to building the final sink, and also imply the indispensable role of sustainable landfills in closing materials loop in CE.

1.1.1 Waste Hierarchy and the Limits of Circular Economy

The concept of waste hierarchy principle has existed for approximately 40 years, which is prioritizing reduction, recycling and reuse of waste over treatment or disposal, and the concept was first proposed in the Dutch parliament in 1979 (Pires & Martinho, 2019). Around 30 years later (in 2008), the European Union approved the Waste Framework Directive (EU 2008/98) which introduced and defined some concepts and definitions concerning the waste management, which consists in the waste hierarchy. Since then, waste hierarchy has been a strategy guiding a clear way in the holistic waste management system, which makes recommendations on the treatment of not only end of life waste but also recommends a "waste hierarchy" that is applicable across the 28 member states of the European Union. As shown in Figure 1. 2, the hierarchy of actions consists in: reduction of waste production (prevention), preparation to reuse, recycling (material recovery), energy recovery and, lastly, final disposal or landfilling.

According to the top-down triangle rank, prevention was deemed the perfect way to eliminate waste, and it aims to reduce the generation of waste from the source. The main goal of the action is to reduce the amount of waste generated that could be possibly taken in each step of the life of a product, and is also to reduce their negative effects on the environment and human health, and the amount of hazardous substances in the products. The concept of prevention not only affects producers' way to generate production, but also can it have the influence on the way to consumption by consumers. On the one hand, to accomplish the goal of prevention, producers take lots of measures, for example, over-packaging will be abandoned, and eco-production materials are taken more into account for packages stuff. On the other hand, consumers can prefer reusable and refillable goods, buy only what is needed, share goods and buy second-hand products. Besides, political and legislative decisions are the most important factor affecting the implementation of action prevention, for example, decrees and legislation could extend producer responsibility, and fomulation of regulations of penalties and rewards could encourage both producers and consumers to follow a reduction in waste.

To some extent, prevention is the most perfect strategy in waste management due to the reduction from the source. According to triangle rank, prevention is a priority in the waste hierarchy. However, it is not easy to prevent waste generation in real life, especially in the condition of exploding population. Apart from the factor of population, factors of social, economic and resources, would result in harsh progress in the prevention of waste. Thus, prevention could not possibly become the most practical method in the holistic system, though it continues to the top hierarchy in the waste management system.

Preparation of reuse is followed by prevention, and ranks in the second hierarchy. According to EU 2008/98, preparation of reuse is to "control, clean, dismantle and repair objects that became waste, so that they can be reused without further pre-treatments". Although this procedure seems to be an approach to addressing the issues of waste generation, actions to promote and implement it are missing; indeed, these actions about preparation or reuse are carried out by organizations different from the ones that manage the waste.

Recycling not only plays an important role in waste management, but also is a critical step in closing the material loop in CE. Recycling aims to recover both materials and energy generated from discarded waste in order to obtain new products, materials or substances instead of consuming natural resources. According to the European Directives 851 and 852 of 2018, by 2030 the target for recycling of municipal solid waste is set to 60% of the total waste produced (EU 2018/851); this recycling rate is set to 70% concerning packaging waste, with different goals depending on the type of material. For instance, the recycling rates for plastic, paper and glass are respectively 70%, 85% and 75% (EU 2018/852). Although these targets are very encouraging and ambitious, and the data seems to be very powerful for the citizens, the matter of waste itself is not taken into consideration, in terms of infinite material lifespan and recycled times. Thus, recycling efficiency should be taken into account when it comes to the recycling step. Recycling efficiency is defined by three important indicators in terms of material, energy and quality efficiency (Cossu et al., 2020a), simply illustrated in **Figure 1. 2**.

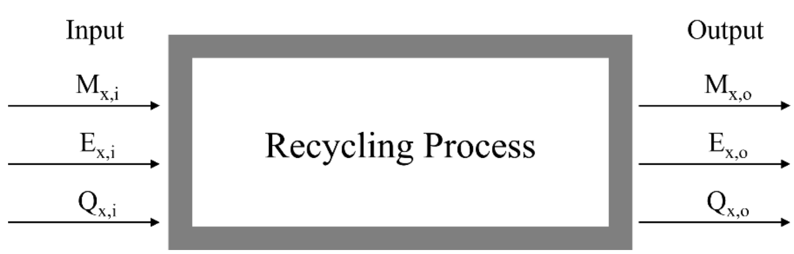

Figure 1. 2. Recycling effiency affected by material, energy and quality efficiency. (M: material, E: energy; Q: quality. x: different types of resource, i: input, and o: output)

Material efficiency (η_M) is the ratio between the material output (m_o) from recycling process and the material input (m_i) the recycling process, and the equation is as follows:

$$\eta_M = m_o/m_i \qquad (1.1\text{-}1)$$

Energetic gain (E_g) is the difference of the specific energy consumed for the production of the virgin product (e_v) and for the recycled one (e_r), divided by the specific energy used to produce the virgin product:

$$E_g = e_r - e_r/e_v \qquad (1.1\text{-}2)$$

Quality efficiency (η_Q) is the ratio between the quality of the product obtained from recycling (Q_r) and the product from virgin material (Q_v):

$$\eta_Q = Q_r/Q_v \qquad (1.1\text{-}3)$$

Both material efficiency and energetic gain can be quantified according to some collected data, while quality efficiency is not easy to be defined. Because in the recycling process, the quality of the recycled materials mainly depends on their application, and the same recycled materials will be required with different levels of quality due to different applications or practices.

Apart from material recovery, energy recovery is also very important and necessary in waste management. It mainly aims to gain both materials with high energy value and heat generated by the waste treatment process, such as heat from the incineration process, biogas from anaerobic digestion and biofuel from the biomass gained from organic waste (Consonni et al., 2011; Ma et al., 2018; Yaman et al., 2020). Those thermal processes can not only accomplish the goal of energy recovery but also can they reduce the waste volume, consequently reducing the final disposal of waste volume. However, energy recovery is a hot topic but not so widespread due to the need for high-cost investment and specific technologies. Besides, the incineration ash and gases produced during the thermal process will inevitably enter the final disposal step.

In the triangular hierarchic representation, final disposal ranks at the bottom of the top-down triangle, which is deemed as the least preferred option. With the term disposal is intended landfills as well as every biological, physical, chemical and thermal pre-treatment prior to landfilling (EU 2008/98). Landfilling is viewed as a sort of a dustbin in the system, in which to deposit all unavoidable and non-recyclable wastes, together with residues from previous treatment, namely all the residues from each previous step of the hierarchical waste management. Although landfills are regarded as the final sink can accept all the residues which could be treated in the previous steps of waste hierarchy, the long-term emissions from the landfills are always the most concerning issues. Long-term emissions, including odour, gases and leachates, have a severe impact on the surrounding environment, eco-system and citizens nearby. Thus, reducing the long-term emissions and keeping landfills sustainable is important in terms of both technology and policy.

Current waste management practices are strongly influenced by the waste hierarchy, and each phase of it is necessary to implement and important. The circular economy (CE) is a part of this integrated system and should include every step of the hierarchy defined by EU 2008/98. In 2015, the Circular Economy Strategy from EU COM/2015/0614 (EU Commission, 2015) defended the role of waste hierarchy.

Circular economy is currently a popular concept promoted by the EU, by several national governments and by many businesses around the world. Unlike the traditional linear economy, recycling the practical policy and business-orientated CE approach emphasizes product, component and material reuse, remanufacturing, refurbishment, repair, cascading and upgrading as well as solar, wind, biomass and waste-derived energy utilization throughout the product value chain and cradle-to-cradle life cycle (Corvellec et al., 2022; Korhonen et al., 2018; Parajuly et al., 2020). However, the content of the CE concept is not comprehensive in terms of both scientificity and framework. The concept of CE mainly focuses on developing environmental sustainability, and it contributes to closing the material loop by highlighting the recycling process. For this reason, the concept seems to be perfect in the circular approach in terms of addressing resource issues. The emphasis placed subsequently on the recycling of waste has promoted throughout Europe a marked increase of separate collection, often perceived by politicians and citizens as the definitive solution for any waste disposal problem.

Despite the recycling process closing the material loop, it ignored the disposal of unrenewable materials and dramatically underestimated the importance of landfilling disposal. As mentioned before, residues would be produced from all the stages in waste hierarchy management. Even though, recycling contributes to converting waste resources into second resource (e.g. material recovery and energy recovery), "by-products waste" is generated during the recycling process, which is missing in the CE concept, consequently, the disposal is missing. Besides, in the concept of CE, many key questions are still open. Material flows exceed man-made boundaries and the complexity will increase when new uses are found for the existing flows, the basic idea of CE. Furthermore, the utilization of bio-based materials and biofuels will have an important role in CE. But the assessments of the actual environmental impacts of biofuels (Holma et al., 2013; Mattila et al., 2010), biomaterials (Weiss et al., 2012) or various types of eco-efficiency initiatives (Huppes & Ishikawa, 2009) still face many unresolved methodological and other limitations, e.g. those concerning the common method of environmental life cycle assessment (LCA) in these types of cases. Controversy about the concept has continued since its inception, and critiques about it have also existed, in terms of environmental, economic and social sustainability (Corvellec et al., 2022; Cossu et al., 2020b; Hobson, 2021; Korhonen et al., 2018). As shown in **Table 1. 1**, the limitations and challenges of CE were summarized in terms of two aspects: scientific sense and framework.

Table 1. 1. Limitations and challenges of the circular economy concept, modified from (Cossu et al., 2020a; Korhonen et al., 2018)

Limitations in terms of scientific sense	Limitations in terms of framework
▪ Not every material or waste generated can be recycled	▪ Thermodynamic limits
▪ Recyclable materials cannot be recycled an infinite number of times	▪ System boundary limits
▪ Recycling activities generate residues as well	▪ Limits posed by physical scale of the economy
	▪ Limits posed by path-dependency and lock-in
▪ Recycling activities cause the accumulation of hazardous substances in the final products, thus decreasing the quality of the goods produced	▪ Limits of governance and management
	▪ Limits of social and cultural definitions

Even if CE is recognised today as a necessary cornerstone of sustainable development, we are still far from the "Zero-waste" idea, which remains a theoretical objective; moreover, it is not prepared to manage the residues generated by recycling activities. Thus, to accomplish the goal of reaching the real closing loop, final disposal or landfilling must be taken into consideration. Furthermore, to reach the target of final sustainability in society, economy and environment, measurements must be done on a regulatory level to promote sustainability of the system and allow it to act as a virtuous sink to close the material loop and immobilize the abovementioned contaminants.

1.1.2 Role of Landfilling as a Sink for Closing Materials Circular Loop

The concept of circular economy (CE) has been introduced to the holistic waste management strategy and it is imperative arises from the huge and still growing material turnover of modern societies. CE mainly focuses on recycling to achieve resource availability in order to accomplish the goal of closing materials circular loop. However, recycling processes are fed with materials that contain not only useful and valuable but also hazardous substances. Numerous substances are potentially shifted to recycling materials and pose risks to human health and environmental safety. Although when it comes to recycling, lots of praise could be seen and little analysis describes the fate of chemicals throughout the lifecycle of materials ending up in recycled products, the adverse impact of detrimental substances on recycled goods does exist, for instance: carcinogenic substances contaminate the asphalt cycle, heavy metals contaminate recyclable grit, and brominated flame retardants and phthalates contaminate the plastic cycle (S. J. Chen et al., 2009; Pivnenko, et al., 2016a, 2016b; Pivnenko & Astrup, 2016). Because of the accumulative contaminates, those recycled materials could not be recycled infinitely, which is meant the final disposal of those un-recyclable waste must be taken into consideration.

Moreover, the recycling procedure itself can generate residues. Recycling processes, in the same way as procedures used in the processing of natural resources, can be outlined in four distinct stages: extraction, selection, refinement and production, as shown in the recycling part of **Figure 1. 1**. Each of these stages produces residues that will subsequently need to undergo treatment in order to render innocuous or immobilise potential contaminants; lastly, a final sink will need to be identified in which to store the wastes safely and sustainably over an extended period of time, as is the case with any type of natural cycle. According to **Figure 1. 1**, the total amount of generated residue generated (R_{tot}) is thus obtained from the difference between the diverse waste flows (W_i) and the final products (P_h), as described by the following mass balance equation (Cossu et al., 2020a):

$$R_{tot} = \sum_{i,j,k,h} \left(R_i + R_{i,j} + R_{i,j,k} + R_{i,j,k,h}\right) = \sum_i W_i - \sum_h P_h \qquad (1.1\text{-}4)$$

Considering the waste generation produces in each stage of the recycling procedure, thermal treatment plants are deemed as an ideal final waste treatment option because they can not only accomplish energy recovery but also considerably reduce the waste volume. Inclusion of the production of Solid Recovered Fuel (SRF) or Refuse Derived Fuel (RDF) could make most of these materials ultimately end up in thermal treatment plants, while some are co-fired in other facilities (coal power plants, cement plants, etc.) (Hilber et al., 2007; Y. Yang et al., 2021). For these reasons, it becomes a very popular treatment in developed countries. For instance, countries like Germany, Japan or Denmark seem to have completely abandoned the use of landfills, reducing it to the 2-3%; whilst they do not consider the amounts of residues generated from waste incineration, for example, fly ash and bottom ash, which are often landfilled or exported to other countries (Cossu et al., 2020a). To make a clear understanding of the percentage of waste recycled, statistical data should take into consideration all the flows of residues coming from other treatment options. Cossu et al. 2020 made some assumptions in order to calculate in a more accurate way the percentages of waste recycled, incineration and thermal treatment, which are:

- 60% of the recovered waste fractions become new products;
- 20% of the residues from the treatment of waste are thermally treated;
- 20% of the residues from recycling processes are landfilled;
- The residues from thermal treatment are landfilled for 20% and recycled for 10%;
- No material is recovered from landfills.

Considering these assumptions, it is possible to describe a completely different situation with respect to the one obtained from statistical data. A large proportion of residues will finally end in landfills instead of being recycled as we expected. Thus, the addition of final sink for closing materials loop in CE is imperative and necessary. In **Figure 1. 3**, the non-renewable residues generated from recycling and recovery procedures can be deposited in presence of the final sink. As shown in **Figure 1. 3**, the amount of residues ending up in landfill is equal to the fraction FS and the total amount of waste disposed in a landfill would be equal to the sum of the fractions from recycling (FS_{RR}), the one from thermal treatment (FS_{TT}) and the one directly landfilled (FS). In a word, the disposal of waste is actually achieved through the production of end products in recycling, together with gasification of material in thermal treatment and permanent depositing in landfilling.

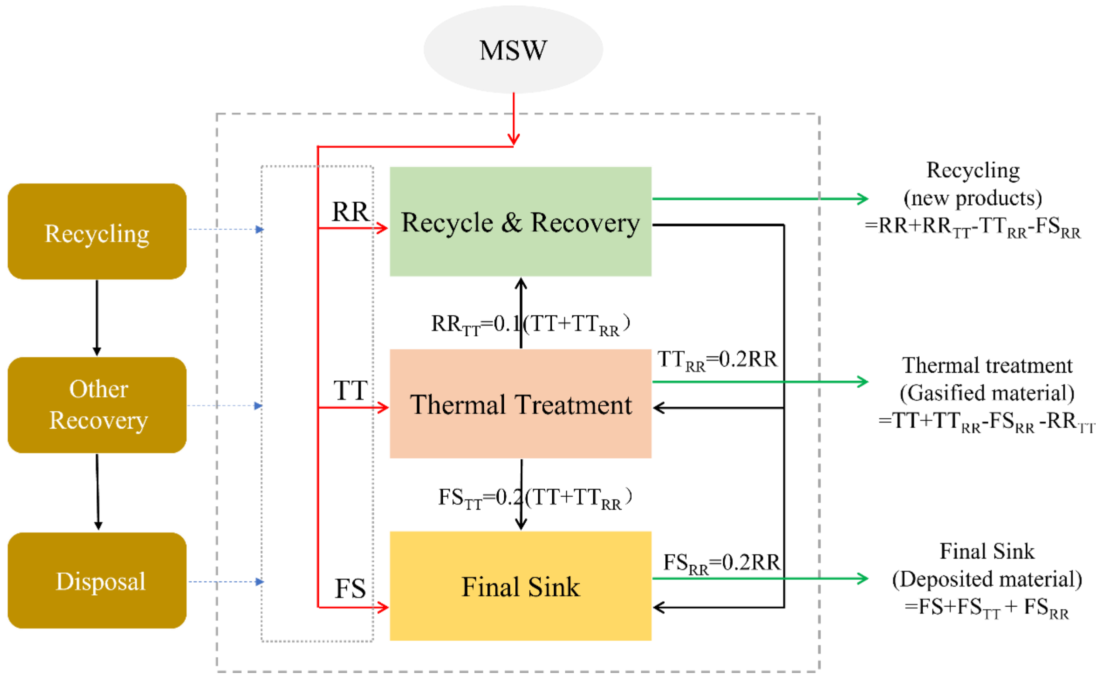

Figure 1. 3. Flows of residues among the main management options (modified from Cossu et al., 2020a). (FS_{RR} :fractions from recycling; FS_{TT}: fractions from thermal treatment; FS: the fractions directly enter into the final sink)

Moreover, another important role of the landfill in CE is the reduction of diffuse pollution (Cossu & Stegmann, 2018). In natural conditions, environmental protection measures are heavily reliant on dilution. However, with time the concentration of pollutants in the water, air and soil will increase. Many pollutants will accumulate over lengthy periods in the environment and create a toxic local, regional, as well as global environment. Moreover, little is known about the long-term behavior of nanoparticles, endocrine substances, etc. in the different media: air, water and soil. Heavy metals remain indefinitely and may react with other elements, be adsorbed/absorbed to particles, solubilized or chemically bound; even in a less mobile state, they may be mobilized when the chemical/physical conditions change. Besides, biologically stable compounds such as aromatics, chlorinated hydrocarbons and other xenobiotics, which may also be produced during thermal processes, are often of low water-solubility; they will be adsorbed/absorbed to soil particles or other surfaces and are distributed in the environment. Some of the compounds will be degraded after longer periods of time, whilst others may remain or at times be converted into even more hazardous compounds. Abovementioned facts demonstrate how dilution has never been, and indeed never will represent a long-term solution. The ultimate goal is to prevent dilution as far as possible and aim to achieve the concentration and isolation of final emissions from the environment. Although the circular approach of CE tried to prevent diffusion by recycling and recovering materials and energy and diffusion of contaminants did reduce, diffusion of contaminants continues to represent a problem and modern landfilling may represent, contrary to the generally held belief, a tool for use in reducing the problem.

Therefore, the matter cycle including emission control strategies and final materials sink, should be taken into consideration, as described by the scheme in **Figure 1. 4**, which was modified from (Cossu & Stegmann, 2018).

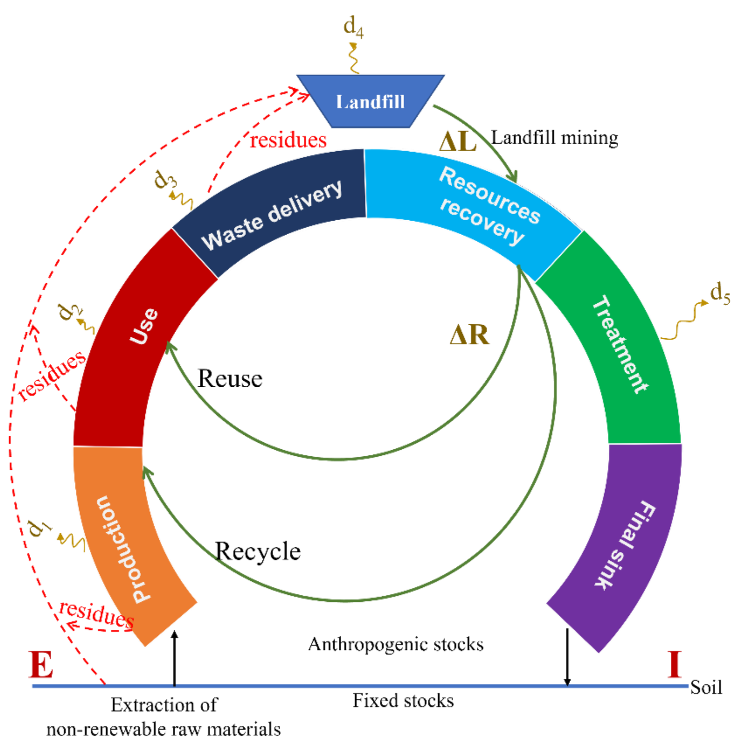

Figure 1. 4. Role of the landfill in the circular economy, modified from (Cossu & Stegmann, 2018)

Firstly, non-renewable raw materials are extracted from the soil for producing new production; the extraction process mobilizes the substances that were stored in a non-mobile form and generates residues. The final fate of a product is being disposed in a landfill after being used. As mentioned before, all the waste management stages generate residues and those diffuse emissions as well as are discovered at every stage of life of the product. The excavation of waste from old landfills (landfill mining) provides additional resources whilst inevitably generating emissions. In the end, the final waste generated needs to be treated to avoid the spreading of toxic substances that accumulated during the previous treatments. Indeed, contaminants must be immobilized and a final sink must be identified to close the material loop.

The following mass balance can be determined considering this circular system (Cossu & Stegmann, 2018):

$$E = \varDelta R + \varDelta L + \varSigma d_i + I \qquad (1.1\text{-}5)$$

Where:

E: extracted raw materials;

$\varDelta R$: recycled and reused materials (secondary raw materials);

$\varDelta L$: recovered materials from landfill mining;

d_i: diffuse emissions of each process;

I: immobilized materials in the final sink.

Rearranging the equation (1.1-5), it is possible to make clear which are the actions necessary to minimize and control the diffuse emissions ($\varSigma d_i$), obtaining the following equation:

$$\varSigma d_i = E - \varDelta R - \varDelta L - I \qquad (1.1\text{-}6)$$

In order to minimize the diffuse emissions, the actions to carry out are:

Minimization of the extraction of raw materials (E);

Maximization of reuse, recycle and recovery of residues ($\varDelta R; \varDelta L$);

Maximization of immobilization of materials (I) in a final sink.

As for any cycle of chemical elements (e.g., biogeochemical cycles of carbon, nitrogen and phosphorus), it is necessary to put back the material in a stable form. Although cycles exist in natural environments in various ways depending on material forms, such as the biogeochemical cycles the storage can be found in the marine sediments or in the atmosphere, for the residues of the circular economy the final sink is the sustainable landfill.

As mentioned above, all human activities undertaken will result in the production of gaseous, liquid and solid emissions which, will finally go back to the earth sites (BES) in different ways or forms, and in the majority of cases, those BES substances will exert a detrimental effect on the environment. CE leads an ideal circular approach to close materials loop by recycling the waste and reducing the consumption of raw resources. However, it cannot exist without the management of the residues; in order to truly close the loop of materials, a final sink is necessary. Thus, building a sustainable final sink to store non-renewable residues and prevent the diffusion of contaminates to immobilize the potential pollutants and avoid the contamination of the environment, is the correct way to truly close materials loop.

1.2 Landfill reactor

Although landfill ranks at the bottom of the waste hierarchy strategy and has negative side effects on the environment, it remains the most common method for waste disposal worldwide and continues to be considered a reliable and low-cost alternative to final municipal solid waste disposal (Torrente-Velásquez et al., 2020). To date, landfills have developed from uncontrol-emission and polluted open dumps to modern highly engineered facilities with sophisticated control measures and monitoring routines. However, in spite of all new approaches and technological advancements, the landfill still is a potential risk in the long term due to long-term emissions as the consequence of reactions taking place inside (Kjeldsen et al., 2002; Laner et al., 2011, 2012; Sekhohola-Dlamini & Tekere, 2020), as shown in **Figure 1. 5**. Much of current landfill design and technology has been introduced as a reaction to problems encountered at actual landfills.

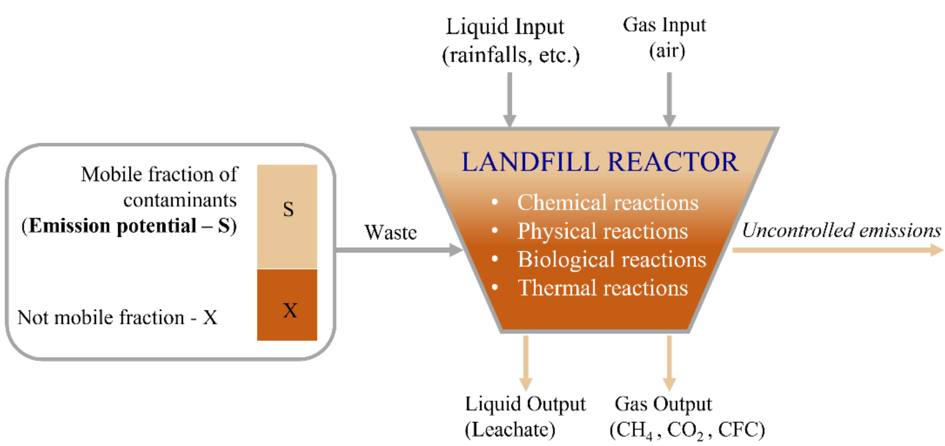

Figure 1. 5. The concept of general landfill reactor. (S: emission potential and X: non-mobile fractions)

In general, landfills receive a variety of waste categories and types, including demolition waste, industrial/commercial waste, and contaminated soil, but municipal solid waste with a high content of organic waste always is a significant fraction (Christensen, 2011). The mix of received waste varies over a wide range within and between landfills and will lead to large amounts of reactions taking place in landfills. The principal reactions and processes that may occur are biological decay, precipitation and dissolution of inorganic constituents, sorption of chemical constituents, leaching of sediments, ion exchange, generation and diffusion of gases and movement of dissolved materials. According to the reaction principle, landfilling reactions could generally be categorized into four main types, including chemical reactions (e.g. oxidation and precipitation), physico-chemical reactions (e.g. dissolution, leaching, adsorption, desorption, etc.), biological reactions (e.g. anaerobic and aerobic) and thermal reactions (e.g. combustion and pyrolysis) (Christensen, 2011; Cossu & Stegmann, 2018; Kjeldsen et al., 2002). Thus, landfill is a kind of reactor where chemical, physical, biological and thermal reactions are producing.

Landfill can be categorized into two main types of reactors in terms of technology, conventional reactor and bioreactor, while bioreactor can be further subdivided into five types of bioreactor: anaerobic bioreactor, aerobic bioreactor, hybrid bioreactor, semiaerobic bioreactor and flushing bioreactor. As shown in **Figure 1. 6**, compared with conventional reactor which are described with respect to the time-dependent development in gas and leachate quality as well as quantity, the other five types of reactors are described with respect to depending on new technologies and proper approaches to improving the quality of gas and leachates as well as utilizing the gas and leachates.

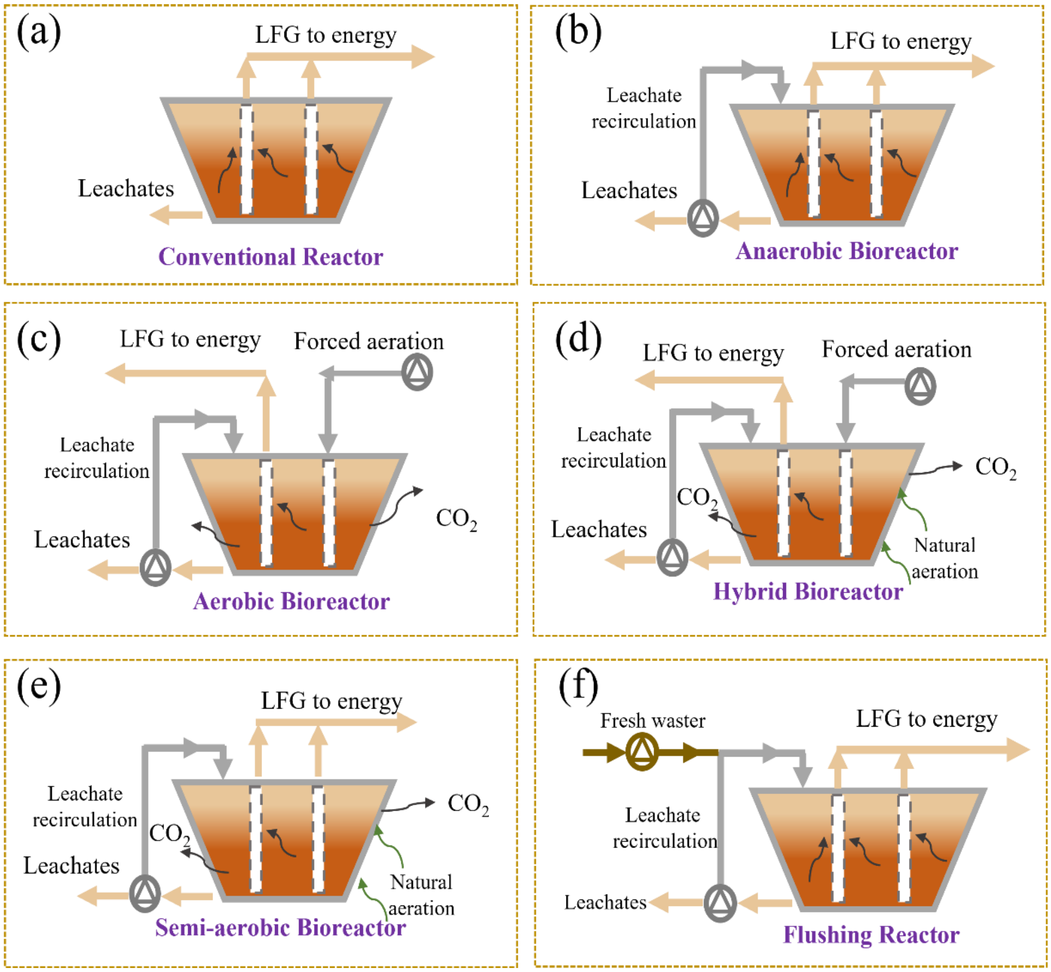

Figure 1. 6. Main technical differences of the various landfill reactors, modified from (Christensen, 2011; Grossule et al., 2018).

- Conventional reactor

Conventional landfill reactor generally receives a mix of waste without hazardous waste and the buried waste is typically compacted to a wet density of 0.7 - 1.0 t/m^3 (Christensen, 2011). The reactor is equipped with leachate and gas management systems or collection systems in order to monitor and control the landfilling emissions. Because of lack of pretreatment, heterogeneity of the waste, lack of mixing and very often low moisture content, the reactions taking place inside landfill are very complex and can last for decades years or even centuries, consequently, landfills constitute a long-lasting "reactor".

In the conventional reactor, the degradation of organic matter is predominantly anaerobic (Adhikari et al., 2008; Erses et al., 2008; Ma et al., 2018). Although the initial degradation of organic matter is aerobic in presence of oxygen, the aerobic degradation could not last a long time (several days) as the oxygen contained in the air entrapped with the landfilled waste is used up. However, the subsequent anaerobic degradation could not be described as one or two simple processes due to a series of reactions, as well as the movement and mix between the old and young leachates. With respect to gas and leachate composition the phases of the conventional landfill are categorized into: (1) Initial aerobic phase, which lasts for a short period in presence of oxygen; (2) Acidic phase, where the pH of leachate is lower less than 6 and volatile fatty acids (VFA) and CO_2 start being produced; (3) Initial methanogenic phase, where niches of near neutral pH value exist, a balance between the acid formers and the methanogens evolves and CH_4 starts appearing; (4) Stable methanogenic phase, where the rate of landfill gas generation peaks and CH_4 generates more than CO_2, (5) Air intrusion phase, where air begins to intrude the outskirts of the landfill and N_2 will appears in the gas; (6) Methane oxidation phase, where N_2 and CO_2 concentrations are increasing, whilst CH_4 is at low concentration; (7) Carbon dioxide phase, where CO_2 is still produced but in less quantity, and N_2 is now the dominant component in the gas; (8) Soil air phase, where the organic matter is stable and gas with high concentration of nitrogen, 10 - 15% O_2 and 5 - 10% of CO_2 (Christensen, 2011).

All the phases are defined in a theoric way, the real reactor could have more complex reactions and some of the phases could last longer or shorter terms mainly depending on the real conditions and wastes.

The bioreactor is typically defined as a system purposely planned and operated for the in-situ treatment of degradable waste with the aim of enhancing or accelerating conversion processes (Grossule et al., 2018). The definition of bioreactor highlight the fundamental role of water and/or air injection, leachate recirculation, enhanced cultivation and other combinations of in situ treatments designed for allowing biochemical kinetics control, nitrification, pH, redox conditions and moisture content adjustment (Christensen, 2011; Erses et al., 2008; Townsend et al., 2015) Consequently, taking care of initial cultivation of bioreactor, controlling extraction of leachate and gas, managing moisture content, recirculating process liquids and injecting air in waste body can be the active actions efficiently applicable.

In bioreactor, enhancement of landfill cultivation can be accomplished by mixing the highly-degradable organic materials with construction and demolition waste for diluting the acidity of landfill in order to build a suitable biochemical process (Christensen, 2011). The moisture control in the bioreactor is very important for supporting the metabolic processes, nutrients and microorganisms movement (Bolyard & Reinhart, 2016; Norbu et al., 2005). Thus, the injection of treated leachate fractions, wastewater, wastewater treatment sludge as well as freshwater can be theoretically useful to homogenize the reactor, control pH, redox conditions of landfills. Furthermore, the application is proved to have positive effects in degradation kinetics and methane production capacity (Mali Sandip et al., 2012; Norbu et al., 2005). Besides, air injection in the landfill body can consistently speed up the biological processes due to the presence of oxygen (Berge et al., 2006; Morello et al., 2016; Ritzkowski & Stegmann, 2013). The addition of oxygen in the waste body causes the inhibition of methane production and the creation of a different biochemical condition, involving different mircoogranisms and kinetic.

There are five main types of landfill bioreactors in terms of technology and function (Christensen, 2011).

Anaerobic bioreactor is the most common application of bioreactor systems where biological degradation is enhanced by means of leachate recirculation, as shown in **Figure 1. 6** (b). It is similar to the conventional reactor, is managed under anaerobic conditions for improving methane generation rate and recovering LFG but does not produce a significant impact on ammonia removal, and degradation remains slow (Calli et al., 2005; Christensen, 2011)

Aerobic bioreactor (**Figure 1. 6** (c)) is the system within the waste mass through forced air injection. In the aerobic bioreactor, air circulation is promoted, increasing biochemical degradation kinetics of organic substances, for example, increasing ammonia and COD removal kinetics up to 10-fold anaerobic ones, and inhibiting methane generation. The oxygen presence allows also nitrification process, which can contribute to biological ammonium ion reduction.

Hybrid bioreactor, seen in **Figure 1. 6** (d), is a system with a combination of aerobic and anaerobic conditions to achieve the benefits of both of them (Cossu et al., 2016; Long et al., 2009; Xu et al., 2014; Y. L. Yang et al., 2021). In the hybrid system, air injection can be controlled to adjust the need for energy recovery or treatment of nitrogen compounds.

Semi-aerobic bioreactor is based on the passive aeration of the landfill, in which natural flow of the external air into the waste mass is moved through the leachate collection pipes by a temperature gradient between the inside and outside of the landfill (Huy et al., 2020).

Flushing Bioreactor is a bioreactor in which supplementary liquids are added to the waste mass for enhancing biochemical processes and favouring the release of soluble substances (primarily ammonium, but also salts and hard COD) (Bolyard & Reinhart, 2016; Christensen, 2011). This system can be anaerobic, aerated or hybrid. The addition of moisture can be fresh water, or treated leachate (Bolyard & Reinhart, 2016; Morello et al., 2016). However, the flushing bioreactor requires a performance-based management and constant monitoring of the system which is expensive. Thus, to date, there have been no full-scale applications published.

Although bioreactor landfills require a more sophisticated degree of management and monitoring than a conventional reactor, it can have several advantages over conventional landfills, from both an economic and environmental point of view:

1) Reduce the environmental burden due to the management of landfilling emissions (collection of landfill gas and improvement of leachates);
2) Shorten the aftercare time and reduce the aftercare costs;
3) Enhance LFG in anaerobic bioreactor;
4) Increase the rate of settlement providing more capacity or a more stable surface for final use of the site (Christensen, 2011; Grossule et al., 2018).

Despite the significant advantages of bioreactor, it can also have some disadvantages mainly derived from the enhancement of biochemical process:

1) increased odours;
2) physical instability of waste mass due to increased moisture and density;
3) instability of liner systems, surface seeps, and landfill fires.

Considering practical application, the operation of full-scale bioreactors should take account of the comprehensive factors in terms of technological, economic, environmental as well as social influence.

1.2.1 Multibarrier Concepts

Landfill as a final sink for storing non-renewable residues should be designed taking into account the isolation of waste and the environment, in particular, the isolation of emissions generated from the biological, chemical, and physico-chemical reactions taking place inside the landfill. Emissions from landfills are mainly gas and leachate. While gas emissions are regarded as the largest human-caused source of methane in the atmosphere and significantly contribute to global warming, landfill leachate leakage –which contains high amount of organic matter, chloride, ammonium and some hazardous compounds, which can severely pollute the surrounding environment. Indeed, contamination of surface and groundwater by landfill leachate represents the major environmental concern associated with the landfilling of waste (Rubinos & Spagnoli, 2018). To reduce environmental concerns and keep the sustainability of landfills, barrier systems have to be installed to minimize emissions. In this sense, the concept of multibarrier has been introduced in planning or designing a landfill mainly contributes to reducing long-term landfill emissions.

In the design of a landfill, multibarrier consists of not only physical barriers, but aslo sitting and morphology, due to the functions of barriers being both prophylactic and technical installations (**Figure 1. 7** (a)). The physical barriers are commonsense barriers, which include top cover, collection systems (collection of biogas and leachate), as well as lining, with the function of controlling, monitoring and aftercare. With respect to sitting, the barriers that should be considered are (Christensen, 2011): sitting should be built distant from important water resources and avoiding physical damage to the landfill body as well as in geological strata providing subsurface attenuation capacity in case minor leakage should take place. Concerning morphology, three main types of landfill morphologies are used in the practice: below-ground-level landfill (typical design of landfilling in exhausted pits), “slope” landfill and above-ground landfill. Each type of landfill is designed for operation mainly depending on the local geography and geomorphology.

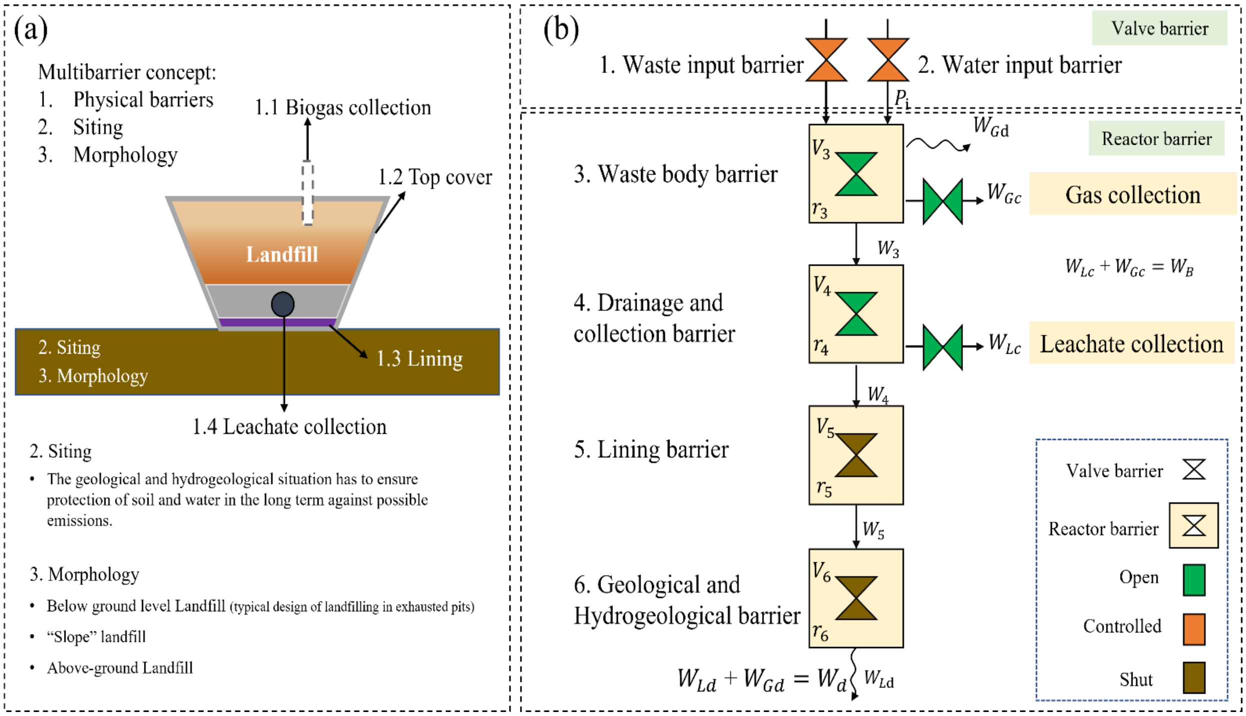

Figure 1. 7. The scheme of multibarrier concept and different barriers and contaminant loads in a landfill system, modified from (Cossu & Stegmann, 2018).

In addition to the traditional interpretation of multibarrier, the landfill itself can also be considered as a barrier to the isolation of the emissions, and it is a cascade of different kinds of barriers: a combination of valve barrier and reactor barrier, as seen in **Figure 1. 7** (b).

Valve barrier consists of waste input barrier and water input barrier, in which no change in contaminant quality occurs and the flow is practically convective (Cossu & Stegmann, 2018). With respect to waste input barrier, the flow of contaminant contained in the waste to be landfilled, W_R, can be regarded as a convective mass flow entering the system and can be expressed as follows:

$$w_R = \sum_{R-1}^{n} c_R Q_R \tag{1.2-7}$$

Where c_R represents concentration of contaminant in the waste; Q_R represents amount of landfilled waste per day; n is number of different types of waste.

Water input barrier can be deemed as the percolation of rainfall water, which moves through the top cover system under the given climatic conditions, with the role of causing hydrolysis of complex biodegradable organics, assisting the movement of nutrients and bacteria in the waste mass and removing the soluble compounds. A hydrological balance in the system can be described as follows (Cossu & Stegmann, 2018):

$$P_i = P + J + R^* - R - ET \pm \Delta U_s \tag{1.2-8}$$

where P is precipitation; J is irrigation or leachate recirculation; R is surface runoff; R^* represents runoff from external areas; ET and U_s represent actual evapotranspiration and water content in top cover soil, respectively.

Reactor barriers can consist of four types of barriers, as shown in **Figure 1. 7** (b): waste body barrier, drainage and collection barrier, lining barrier, as well as geological and hydrogeological barrier. Among those barriers, waste body barrier can be further subdivided into three subbarriers: (1) the reactor barrier, (2) the hydraulic waste barrie and (3) the daily cover barriers.

(1) the reactor barrier is the transformation process of contaminants within the waste body, and the mass balance for the transport of contaminants within the landfill reactor can be expressed by the following equation (Cossu & Stegmann, 2018):

$$V_3 \frac{d_c}{d_t} = \sum_{R=1}^{n} c_R Q_R - c_{3g} Q_g - c_3 Q_3 - r_3 V_3 \tag{1.2-9}$$

Where c_3 is the concentration of contaminants; Q_3 is the leachate flow, and V is the given volume.

(2) the hydraulic wate barrier with the function of physically controlling the liquid flow through the waste, the Darcy law is a proper way to be used to describe the flux of leachate through the waste layer (Cossu & Stegmann, 2018), as follows:

$$v_3 = \frac{Q_3}{A} = k_R i_3 = k_R \frac{h_3}{L_3} \tag{1-10}$$

Where, k_R is the hydraulic conductivity of waste; v_3 is the Darcy velocity; Q3 is the leachate flow; A is the surface of the waste layer; i_3 is hydraulic gradient in the waste layer, h_3/L_3; h_3 is the leachate head in the waste layer; L_3 is the thickness of the waste layer.

(3) the daily cover barrier is built to control the liquid flow and transformation processes within the cover layers.

Biogas collection system is a fundamental role in a landfill management system due to energy recovery and LFG collection. The following equation could be used to describe the flow of contaminants associated with landfill gas emissions (Cossu & Stegmann, 2018).

$$w = c_{3g} Q_g \tag{1-11}$$

Where w_g is the contaminant mass flow; c_{3g} represents gas-phase concentration of the considered contaminant; and Q_3 is the gas flow rate.

The drainage system typically consists of perforated collection pipes laid directly over the barrier, covered by a drainage layer of coarse granular material. The barrier is generally sloped so that a ridge or drainage divide is located midway between adjacent parallel collection pipes. Liquids percolate downward into the saturated zone directly overlying the barrier, and then flow downslope to the collection pipes (Bruce et al., 1989; Koda et al., 2016).

To prevent leachate from leaking out of landfill, the lining barrier is the fundamental system in landfill design. In general, a lining system consists of either geomembrane and a compacted clay liner or a geomembrane, a geosynthetic clay liner and a soil liner (Tang et al., 2018). Besides, a leachate collection system is required in linning barrier design, in order to collect leachate, discharge the leachate at defined points out of the disposal area, as well as avoid an accumulation of leachate above the bottom liner (Cossu & Stegmann, 2018).

Although the concept of multibarrier has been introduced in modern landfill design, one thing should be noticed that is not all barriers would be feasible in every case. Several factors should be taken into the design of barriers: types and qualities of waste, surrounding environment in terms of both geological and humanistic environment, monitoring and aftercare system, as well as maintenance and contingency plan (Christensen, 2011).

1.2.2 Evolutions of Barriers

Evolutions of barriers are developed with the increasing need for sustainability and the long-term safety of landfills. The barriers conceived have marked the technical development of landfills (**Figure 1. 8**), following the cultural development of modern society and reflecting the growing social and political awareness of environmental issues.

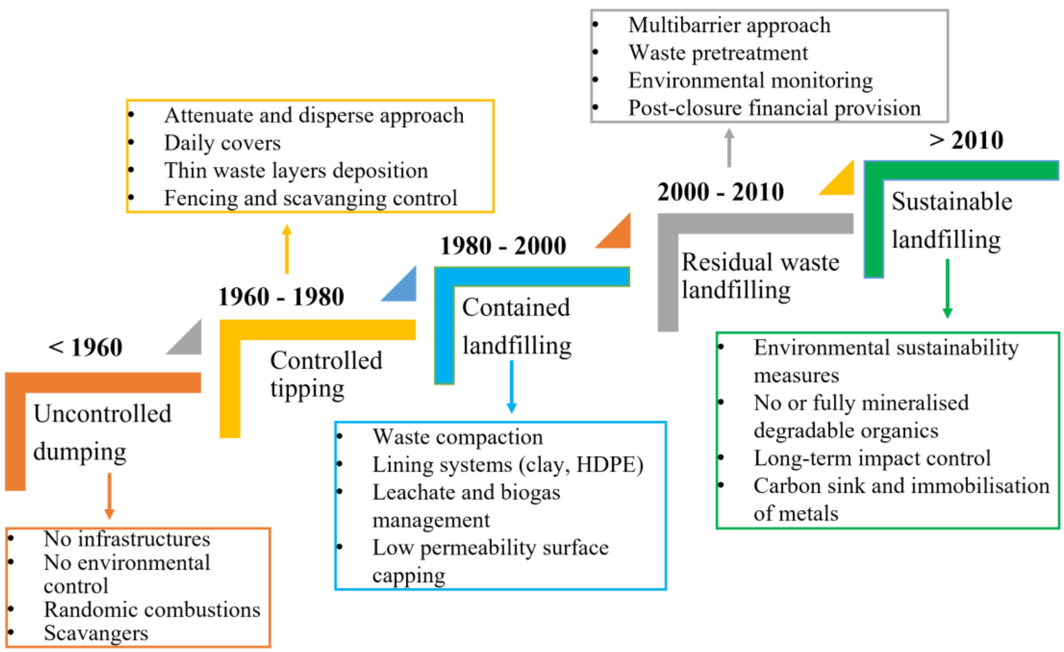

Figure 1. 8. The development of concepts, technologies, and management procedures for landfill, modified from (Cossu, 2010).

Open dumping of MSW poses high risks to human health and the ecosystem as toxic compounds are released into the atmosphere and the environment, including dioxins and dioxin-like compounds (polychlorinated dibenzo-para-dioxins and polychlorinated dibenzofurans) (Prateep Na Talang & Sirivithayapakorn, 2021). As shown in **Figure 1. 8**, around half a century ago, uncontrolled dumping was the dominant waste disposal way. Although nowadays this form of waste management has been regarded as obsoleted waste management and it is not supported by means of environmental control, it remains to be applied in many countries, in particular in developing countries (Ajibade et al., 2021; Prateep Na Talang & Sirivithayapakorn, 2021).

With the increasing concerns about environmental safety, in the 1960s, the utilization of uncontrolled dumping was gradually taken place by controlled tipping in industrial countries. In form of controlled tipping waste management, barriers started to be introduced to prevent the diffusion of contaminants, monitor leachate and control scavenging. However, in the controlled tipping leachate and biogas were not collected and treated, most of them were released into surrounding environments, resulting in pollution in groundwater and the atmosphere.

When simple physical barriers could not meet the demands of prevention of emissions (e.g. biogas, leachates and odours), new barriers with more functions were introduced into the landfill design. A series of management systems were built in contained landfill consisting of lining systems, leachate and biogas collection systems, as well as surface capping, in order to accomplish the goal of preventing and treating emissions. In the contained landfill, collected leachates were subjected to subsequent treatment and disposal dictating the need for specific technologies, in order to remove the high organic, and ammonia content and the wide quality variation linked to landfill ageing. Biogas was exacted for being flared or energy recovery.

However, contained landfill was discovered with the problems of management costs and long-term accumulative contaminants, implying the need for developing more functional landfills. Residual landfill was developed based on a hierarchical view of waste management and rapidly became an international reference strategy. Although residual landfill continues to keep the characteristics established for the contained landfill, it rather constitutes a deposit for residual wastes. Based on the waste hierarchy concept: waste prevention, material reuse and recycling, and energy Recovery, the establishment of a residual landfill is to achieve the final aim of reducing the waste volume, minimizing the LFG generation, and lowering environmental impacts and risks.

Although residual landfill with the multibarrier approach to addressing the environmental and treatment as well as post-closure issues, landfill as a final sink storing a large amount of residues itself becomes a problem. Moreover, residual landfill did not develop at the same pace as environmental sustainability requirements, based on avoiding leaving future generations to manage unacceptable environmental burdens. Thus, in the concept of the modern landfill, a landfill must be built with the sustainability principle, which is meant the landfill can reach an equilibrium with the surrounding environment over one generation, stabilising the landfilling waste and immobilizing uncontrolled emissions, as well as closing the material cycle.

A simple physical commonsense barrier is far from getting landfill to achieve environmental sustainability. For this reason, the multibarrier concept introduced can not only include a series of preventing, collecting and monitoring systems, but also include pretreatments (e.g. washing, thermal treatment and mechanical biological treatment) (Cossu & Lai, 2012; Danthurebandara et al., 2015; Fei et al., 2018) and in-situ treatment measures (e.g. flushing, irrigation and aeration) (Bolyard & Reinhart, 2016; Lü et al., 2012; Raga & Cossu, 2013). Furthermore, both pretreatment and in-situ treatment can significantly enhance the quality of landfilling waste to achieve Final Storage Quality which is an important value to evaluate landfill sustainability.

Barriers are one of the most fundamental and important parts of landfill design, due to their role in reducing the release of environmental-impacted emissions. Considering the long life of the landfill and the emissions from the continuous reaction inside, the lifespan of barriers, the control efficiency of barriers, as well as relevant maintenance should be taken into account in landfill design strategy.

1.2.3 Problems of Traditional Landfilling

Although landfills are regarded as an obsolete system due to issues in terms of land consumption and long-term impacts on the environment, landfill continues to play an important role in waste management, in particular in closing material loop in a circular economy, providing in line with the Back to Earth concept, a sink for all those substances and materials that would otherwise remain dispersed in the biosphere, thus adding to diffuse environmental pollution (Cossu, 2016). Landfills as a final sink for the storage of non-renewable residues must be designed in a sustainable and safe way to guarantee long-lasting safety. However, in many cases, landfills are seen as a simple and economical means of disposing of waste, and from a political, legislative and technical viewpoint they are frequently denied the attention devoted to other engineering works, lacking adequate financial investment to cover the costs required to ensure a sustainable landfill system (Grossule & Stegmann, 2020).

In a traditional landfill, received waste generally contains untreated or partly treated organic matter and thus produces gas as well as contaminated leachate. The term reflects the fact that the presence of organic matter in the landfill and the degradation reactions that it undergoes make the landfill a reactor that must be engineered in order to control the gas and leachate that it produces. Physical barriers are regarded as the most commonsense technology applied to keep the safety of a traditional landfill.

Barriers are engineered in the design of the traditional landfill, graphically depicted in **Figure 1. 9**. In a landfill, potential waste contaminants are essentially present in either a mobilizable or non-mobilizable form. To control the emissions and risk of contamination in both the short and the long term, barriers are used. As shown in **Figure 1. 9** (b), the mobile contaminant fractions generated from open dumps without any barriers are continuously released into the surrounding environment at the peak during the operation time. The quality of emissions (gases, odours and leachates) could not reach equilibrium with the environment even after a certain long time (30 years) of landfill closure, which is meant open dump keeps polluting environments in the short time and the long time due to non-protection measurements. Compared with the open dump, traditional landfills with barriers can effectively prevent releasing emissions. However, traditional landfills could not guarantee long-lasting safety due to the limited lifespan of physical barriers Click or tap here to enter text.. (Laner et al., 2011, 2012; Rowe, 2005)Although barriers can prevent the mobile contaminant fractions releasing during the landfill operation period, the condition of the barrier system can deteriorate over time, once the barriers break, all the contaminants can be released into the surrounding environment and cause severe pollution. As seen in **Figure 1. 9**, untreated odours, gases and leachates will be released to the atmosphere, groundwater and soil, and the quality of emissions will break the safe limited levels and cause damage to the surrounding environment.

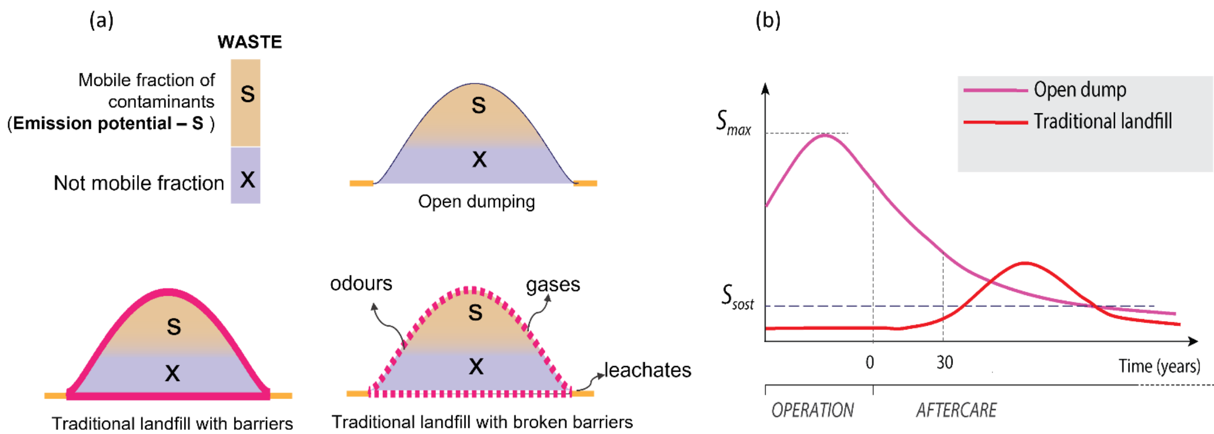

Figure 1. 9. Main issues associated with the open dump and traditional landfill (a); time trend of the emission potential for release of contaminants from a landfill (b), modified from (Grossule & Stegmann, 2020). S_{max}: the maximum amount of releasing a possible mobile fraction of contaminants; S_{sust}: the emission potential reaches a value in equilibrium with the environment.

Moreover, the presence of barriers could not improve the quality of landfilling waste, to some extent, barriers could even cause deterioration of the inside environment of landfills due to the accumulation of moisture and changes in pH value of accumulative leachates. Besides, in terms of financial sense and lifespan of capacity, a traditional landfill is costly(Al-jaf & Al-ameen, 2021; Berge et al., 2009). Because once the landfill reaches permit capacity, the waste input will stop and so will the company's opportunity to recoup their expenditure. At this point, the company will need to increase their expenditure on securing an engineered cap to keep out moisture and conduct landfill gas collection, maintenance and monitoring for decades. At the same time the company will be looking for a new landfill, and if successful the procedure begins again, whilst all the time the waste continues to be generated and is in need of safe disposal and treatment (Read et al., 2001).

For those reasons, a traditional landfill could not achieve the sustainability of a final sink, consequently, it could not truly close material loop in a circular economy.

Accordingly, to achieve the principle of environmental sustainability and reach the equilibiruim with the environment, establishing a landfill should take into account not only the barrier systems, but also the quality of landfilling waste (pre-treatment) as well as the landfilling conditions (in-situ treatment) (Grossule & Stegmann, 2020). A sustainable landfilling concept should be also applied to the design of a landfill.

1.3 Sustainable Landfilling

Landfilling is considered the least preferable option in solid waste management according to the waste hierarchy. Nevertheless, landfills continue to play an important role in solid waste management worldwide with respect to cost and technology. Especially in some low-income countries, landfills are still the main disposal method of waste management (Dhokhikah & Trihadiningrum, 2012; Zohoori & Ghani, 2017). However, in a modern waste management system, a landfill with the simple function of storing waste is far from the social and environmental goals due to the environmental issues arising from traditional landfills, in particular the long-term emissions (gas and leachate). The design of landfill must be safe and sustainable, in order to provide a final sink for non-renewable residues from the circular economy and close the material loop. But the sustainability principle in a traditional landfill can not be achieved due to the lack of sustainable design in landfills. The physical barriers system can not prevent long-term emissions or improve the quality of landfilling waste, consequently, gas and leachate generated from landfills will leak into the surrounding environment due to the broken barriers and cause lots of pollution.

Accordingly, one of the major issues in the landfilling of waste is compliance with environmental sustainability in terms of control of long-term emissions (gas and leachate) and compliance with final storage quality (FSQ) of the landfill (Cossu & Stegmann, 2018; Heimovaara et al., 2014; Laner et al., 2012). To achieve sustainability in the final deposal of waste, sustainability must be introduced into landfill design. Physical barriers system continues to be the fundamental part of sustainable landfilling, contributing to preventing the leak of emissions and assuring the safety of the surrounding environment during a certain period (depending on the lifespan of barriers). Apart from the traditional physical barriers, active barriers with a combination of treatments (pre-teatments and in-situ treatments) should be implemented throughout the different life stages of a sustainable landfill, in order to stabilize the waste and reduce the long-term mobility of pollutants. In this case, the sustainable landfill can keep safety both in a short time (operation period) and a long time (post-closure care period).

The application of sustainable landfilling aims to not only keep the long-term safety of a landfill, but also plan the current economic and social development with the purpose of guaranteeing the same possibilities of the present generation to the future ones. Thus, achieving sustainability in landfilling is an essential need for both environmental and social development.

1.3.1 Definition of Sustainable Landfilling

As above mentioned, landfilling constitutes an unavoidable final step in waste management, being aimed at bringing no-renewable residues back to the stable form they were in before the extraction from ground and their use as raw materials (Cossu, 2016). The application of the sustainability principle into landfilling mainly aims to guarantee environmental protection and health safety during operation and post-closure care of the landfill. Thus, sustainable landfilling of waste should keep waste in a stable state in terms of chemical and biological senses, in order to obtain equilibrium with the surrounding environment as well as a closure of material loop. For those reasons, a sustainable landfill can be defined as a landfill, both in operation and post-closure period, having a stable structure and poses no environmental risk (Vaverková, 2019; Westlake, 1997).

To comply with the task of sustainability, the sustainable landfill can not rely only on physical barriers but on further measures of protection. These measures consist in the multibarrier system which is composed by different levels of protection formed by physical barriers, active barriers and control of the quality of input waste. Measures applied to landfills should ascertain the whole life phase of a landfill is in line with sustainability, both in the operation and post-closure . Thus, actions aimed to reach sustainability should be taken in each stage of landfilling waste, as shown in **Figure 1. 10**.

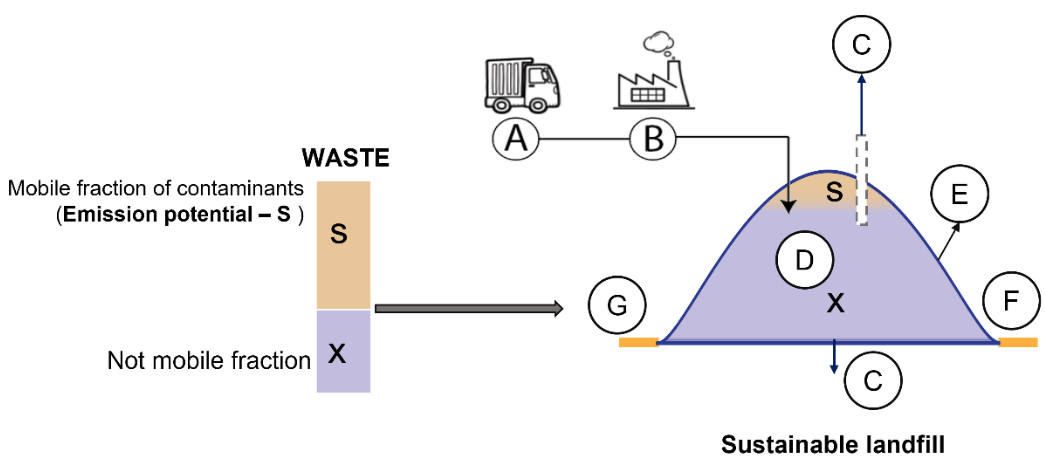

Figure 1. 10. The multibarrier concept in the sustainable landfill. A: waste minimization; B: pre-treatment; C: biogas and leachate management system; D: in-situ treatment; E: top cover; F: lining; G: siting and morphology, modified from (Grossule & Stegmann, 2020).

Landfills store huge quantities of mobile or non-mobile waste. The mobilizable fractions contained in landfilled wastes and exposed to the atmosphere, groundwater or soil are in line with their characteristics of degradability and leachability, which pose a high risk to the environment (Laner et al., 2011; Sayadi et al., 2015). Physical barriers are one of the most effective instalments in controlling those mobile contaminants fractions. Physical barriers consist of top covers, clay lining, drainage systems as well as gas and leachate collection systems, which mainly aim to collect and treat the leachate and gas to avoid uncontrolled emissions spreading in the environment.

Apart from avoiding the emissions spreading, enhancing the quality of buried residues is also an important method of achieving landfilling sustainability. Both pre-treatments and in-situ treatments can be applied to achieve this goal. Pre-treatments can minimize the emission potential of waste prior to landfill by means of physical, biological and thermal methods, in order to comply with the final storage quality and equilibrium with the surrounding environment. Moreover, physical treatments may consist in shredding and sieving in order to homogenize the waste mass; whereas biological treatments consist in aerobic or anaerobic degradation processes to stabilize the organic putrescible fractions (Grossule & Lavagnolo, 2020; Vallini et al., 1991). Boosting the biochemical stabilization of landfilling waste and guaranteeing an adequate FSQ can be also achieved by means of in-situ treatments, for instance, flushing, aeration, etc. These measures can be implemented either during the operational phase of the landfill or during the post-closure phase. Due to the types of in-situ treatment, landfills can be classified into several bioreactors: anaerobic landfill, aerated landfill, semi-aerobic landfill as well as hybrid landfill. Boreactor landfills can lead to faster mineralization and stabilization of municipal solid waste by accelerating anaerobic degradation of biodegradable components (Nanda & Berruti, 2021). Achievement of the aims of environmental sustainability is guaranteed in those bioreactors by multiple benefits afforded by in-situ treatments, including reduced time frame for interventions and lower post-management costs, shorter duration of environmental responsibility for the landfill management, accelerated reduction of emission potential (due to an increased degradation and leaching) and mechanical stabilisation of the waste mass (Grossule & Stegmann, 2020).

The sustainable landfilling concept comprises the above-mentioned actions and measures to achieve the goal of ensuring that emissions can be ascertained within a safe level during operation and post-closure.

1.3.2 Role of Waste Treatment

In order to comply with the sustainability concept, a landfill should reach an acceptable equilibrium with the environment within one generation time (30 years) (Cossu & Stegmann, 2018). A series of technologies and tools are applied in practice for reaching sustainability. One of the most important management methods is waste treatment, due to its immobilization of contaminants and enhancement of waste quality.

Landfilling waste poses a potential risk to the environment mainly depending on the mobile fractions of contaminants. The mobile contaminants fractions can transform and pass from different phases due to their characteristics of degradability and leachability, consequently can lead to pollution both in atmosphere, groundwater as well as soil (Grossule & Stegmann, 2020). Waste treatment is regarded as the effective method of controlling or removing the mobile contaminants fraction in order to reach the environmental equilibrium. Different waste treatments can be applied during different life phases of the landfill to achieve FSQ, for instance, pre-stabilization of waste prior to landfilling and in-situ treatment of enhancing biodegradation during landfilling or the aftercare phase, as shown in **Figure 1. 11**.

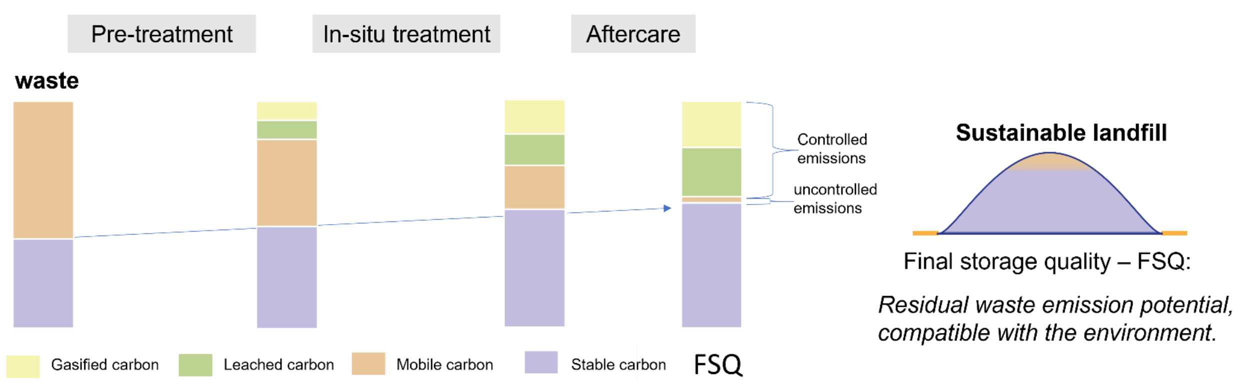

Figure 1. 11. Mobile contaminants are removed by means of waste treatment in order to achieve the final storage quality in sustainable landfills.

Based on the life phase of the landfill, treatment processes can be classified into pre-treatment, in-situ treatment and aftercare treatment.

- Pre-treatment

Considering treatment processes, pre-treatments can be classified as physical, physico-chemical, chemical-physical, biological as well as thermal treatments. Each form of pre-treatments corresponds to different operations and gains different effects, as shown in **Table 1. 2**.

Table 1. 2. Classification and description of unit applied in waste pre-treatment, adopted from (Grossule & Stegmann, 2020).

Process	**Unit operations**	**Aim**
Physical	• Shredding • Sieving	• Pre-treatment, recovery of refuse-derived fuel (RDF)
Biological	• Anaerobic digestion • Aerobic stabilisation • Composting	• Biological stabilisation • Resource recovery (methane, hydrogen, compost)
Thermal	• Combustion • Pyrolysis • Gasification • Thermal incineration	• Reduction of waste volume • Destroying of organic contaminants • Energy recovery • Recycling of ashes
Physico-chemical	• Washing (with or without chemical agents) of inorganic waste	• Removal of contaminants • Reduction of emission potential
Chemical-physical	• Solidification by both inorganic (e.g. Hydraulic binder) and organic (e.g. Thermoplastic materials) reagents.	• Reducing leachability • Increasing mechanical stabilisation • Resource recovery (recycling of stabilised material)

Among those treatments, both biological treatments and thermal treatments are applied in practice and both of them aim to obtain biological stabilisation of waste and reduce the volume of residues. In general, a combination of treatments is often applied to achieve the goal of minimization and reduction of waste economically, for instance, biological pre-treatment is combined with mechanical processes to afford the mechanical biological treatment (MBT), the aim of which is to stabilise undifferentiated wastes prior to landfilling and compact volume of landfilling waste (Ponsá et al., 2010; Soyez & Plickert, 2002.). Moreover, the combination of thermal treatments and co-processing methods may not only reduce the waste volume before landfilling but also recycle or reuse the waste resources, for example, combustion co-processes with physicochemical washing treatment can be able to gain excellent contaminants removal and can recycle the residues as the road construction materails or cement production (A. A. Abbas et al., 2009; Di et al., 2006; Trebouet et al., 2001).

- In-situ treatment

With respect to economical and technical issues, waste pre-treatment might be insufficient in achieving long-term stabilisation of waste and leaching contaminants. Due to the long-lasting reactions taking place inside landfills, leachate and gas can be unavoidably generated over time. To address emission issues during the routine operation of a landfill, in-situ treatment must be implemented in landfills.

All in-situ treatments are applied by means of biological processes to obtain the biological stabilisation of landfilling waste. Based on the different unit operations of in-situ treatment adopted in landfills, landfills can be categorized into several types of bioreactors corresponding with specific aims, which are described in **Table 1. 3**.

Table 1. 3. Classification and description of landfill bioreactors, modified from (Grossule & Stegmann, 2020).

Types of bioreactor	Unit operations	Aim
Anaerobic landfill	Leachate recirculation	• Improvement of biodegradation and leachate quality • Enhancement of biogas in a shorter time • Better removal of soluble compounds
Aerated landfill	Forced aeration	• Improvement of biodegradation kinetics • Reduction of methane dispersion • Removal of nitrogen
Semi-aerobic	Natural aeration	• Acceleration of biodegradation kinetics • Removal of nitrogen • Low cost

The objective of treatment methods of any life phases applied to waste forwarded to final sustainable disposal should be that of removing, transforming or immobilising potential contaminants into a stable form. The combination of pre-treatments and in-situ treatment can be suitable for the quality of emissions to reach an acceptable effect on the environment and to achieve a sustainable state of landfilled municipal solid waste.

1.3.3 Final Storage Quality

A landfill may be considered sustainable if emissions do not significantly modify the quality of the surrounding environmental compartments: air, water, and soil (Cossu & Hans Albert, 2014; Westlake, 1997; Widomski et al., 2017). This status should be reached over the time frame of one generation. Accordingly, the final storage quality (FSQ) can be defined as waste quality reached by emissions and waste at a specific point in time, when all active control measures can be safely removed (Cossu & Stegmann, 2018). In order to guarantee sustainability, the equilibrium with the environment should be reached within a certain time, commonly taken to be around 30 years after the closure of a landfill.

Although perpetual care of a landfill to ascertain environmental safety seems to be reasonable in terms of theory, perpetual care is rarely applied and could not fit with the economic and sustainability principles. In general, the time based aftercare termination is used methodology prescribing to terminate the aftercare after 30 years with respect to the landfill owner's duties. However, it can not guarantee that the landfill has reached environmental acceptability within so-called aftercare termination. Therefore, the design of a sustainable landfill should be based on the kind and amount of landfill emissions achieving the FSQ in a sustainable period, subsequent to which the aftercare phase can be truly terminated. With the respect to the cost, long-term emissions should be maintained within the FSQ values in order to the consequent cost of environmental remediation. The financial provisions are set to cover the aftercare costs for 30 years according to the environmental legislation (Directive 1999/31/EC and D.Lgs. n.36/2003), thus, the pollution caused by emissions after the termination of aftercare, financial support probably will not cover this part of remediation anymore. Therefore, landfilling emissions achieving the FSQ is critical based on the need not only for long-lasting safety, but also for reducing the cost of remediation.

To achieve sustainability targets, a combination of treatments (pre-treatments and in-situ treatments) is required to control the trend of emissions over time. Various types of barriers are implemented to pursue the FSQ objective, to reduce the emission potential (S) in terms of leachate and gas generated, as graphically represented in **Figure 1. 12**.

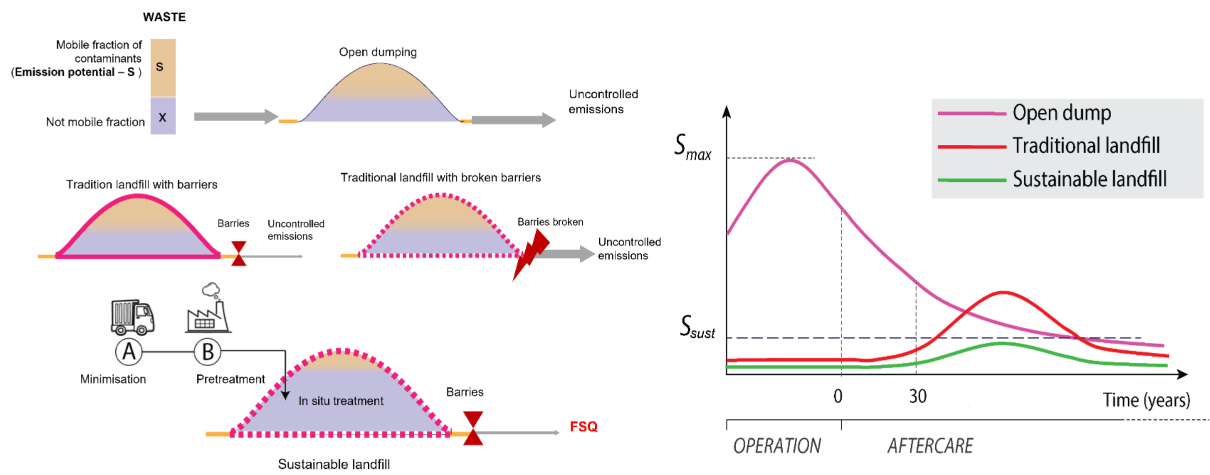

Figure 1. 12. Potential emission of pollutants according to different landfills, modified from (Cossu & Stegmann, 2018), Ssust: the emission potential reaches a value in equilibrium with the environment.

As above mentioned, the sustainability of a landfill is defined by discharge reaching FSQ within one generation, however, up to date, FSQ has not been included in any environmental criteria. In general, the emission potential of the landfill body during the aftercare period may be monitored by means of qualitative and quantitative analysis of biogas, leachate, and deposited waste. Therefore, some of the parameters commonly tested and used as indicators of emission quality may be more significant in evaluating the degree of landfill stabilization and verifying the achievement of FSQ. In **Table 1. 4**, a set of values to define the FSQ according to the Lombardia Region (Regione Lombardia, 2014), those values are given to define the quality of long-term emissions, including biogas, leachates as well as disposed waste.

Table 1. 4. Values of goal parameters for the definition of FSQ modified from (Regione Lombardia, 2014)

	Parameter	Unit	FSQ Value
Leachate	COD	mg_{O2}/l	1500
	BOD_5/COD	-	0.1
	NH_4-N	mg_N/l	50
	Al	mg/l	1
	As	mg/l	0.5
	B	mg/l	2
	Cd	mg/l	0.02
	Cr; Cr (VI)	mg/l	2; 0.2
	Cu	mg/l	1
	Fe	mg/l	2
	Mn	mg/l	2
	Ni	mg/l	2
	Pb	mg/l	0.2
	Zn	mg/l	3
	CN^-	mg/l	0.5
	SO_4^{2-}; SO_3^-	mg/l	1000; 1
	F^-	mg/l	6
	NO_3^-	mg/l	20
	Total hydrocarbon	mg/l	5
	phenols	mg/l	0.5
	Aromatic organic solvents	mg/l	0.2
	Nitrogenous organic solvents	mg/l	0.1
	Total pesticides	mg/l	0.05
	Phosphorous pesticides	mg/l	0.1
	Chlorinated solvents	mg/l	1
Biogas	CH_4	NL CH_4 /m^2 h	0.5
Disposed waste	IR_4	mg_{O2}/g_{TS}	2
	IRD	$mg_{O2}/kg_{VS}/h$	100
	GP_{21}	NL/kg_{TS}	5

Termination of the aftercare phase of a landfill mainly depends on the environmentally acceptable emissions, which is meant landfilling emissions achieve the FSQ before aftercare termination. A sustainable methodology for evaluating FSQ and assessing the aftercare termination has been purposed by Cossu et al. 2007, as seen in **Figure 1. 13**.

The FSQ methodology procedure starts when a landfill has achieved a stable state. Then the first analysis will be carried out for the biogas indexes. Relevant parameters of biogas will be valued for assessing the FSQ. Once values of biogas achieve FSQ limits, leachate indexes will be taken into consideration. In general, the long-term potential pollution of leachates mainly comes from the presence of persistent compounds. Therefore, in-situ treatments for improving the biological process must be applied in landfill to remove those persistent compounds. The last procedure is conducted on the FSQ indexes in solid material. In this step, FSQ indexes are mainly useful to quantify the possible presence of biologically active compounds in landfills not reached by humidity for years. When all the values of each procedure step achieve the FSQ limits, the aftercare can be considered by FSQ methodology to be terminated safely.

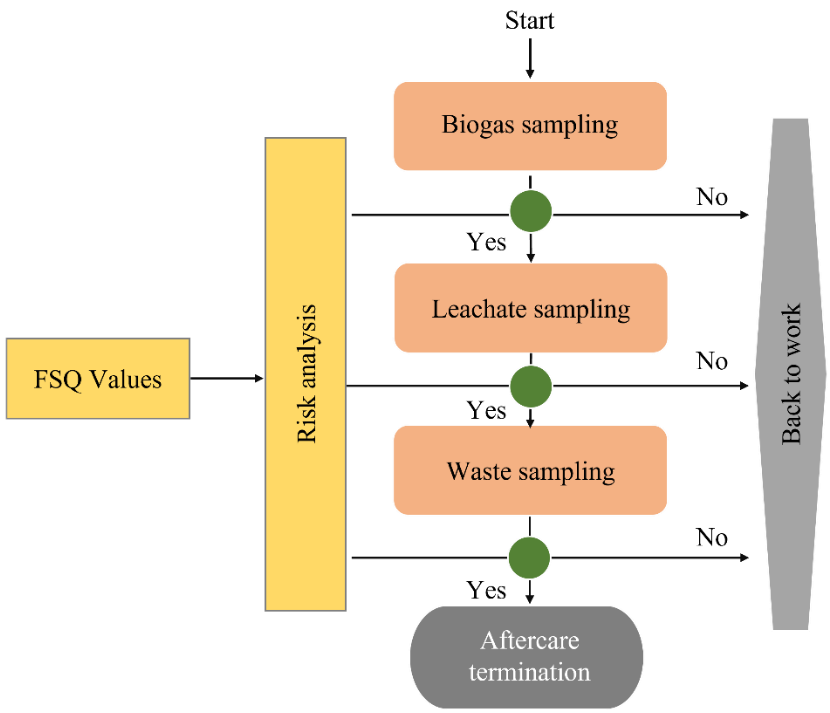

Figure 1. 13. Sketch of the procedure proposed to assess Final storage quality, modified from (Cossu et al., 2007)

References

Abbas, A. A., Jing, S. G., Ping, L. Z., Ya, P. Y., &Al-rekabi, W. S. (2009). Review on landfill leachate treatments. *Journal of Applied Sciences Research*, 5(5), 534–545.

Abbas, Z., Partovi, A., & Steenari, B. (2003). Release of salts from municipal solid waste combustion *residues.Waste Management, 23*, 291–305.

Abumaizar, R. J., & Smith, E. H. (1999). Heavy metal contaminants removal by soil washing. *Journal of Hazardous Materials*, *70*(1–2), 71–86. https://doi.org/10.1016/S0304-3894(99)00149-1

Adhikari, B., Dahal, K. R., & Nath Khanal, S. (2008). A Review of factors affecting the composition of municipal solid waste landfill leachate. *Certified International Journal of Engineering Science and Innovative Technology (IJESIT)*, *9001*(5).

Ajibade, F. O., Adelodun, B., Ajibade, T. F., Lasisi, K. H., Abiola, C., Adewumi, J. R., & Akinbile, C. O. (2021). The threatening effects of open dumping on soil at waste disposal sites of Akure City, Nigeria. *International Journal of Environment and Waste Management*, *27*(2), 127–146. https://doi.org/10.1504/IJEWM.2021.112947

Alam, Q., Florea, M. V. A., Schollbach, K., & Brouwers, H. J. H. (2017). A two-stage treatment for Municipal Solid Waste Incineration (MSWI) bottom ash to remove agglomerated fine particles and leachable contaminants. *Waste Management*, *67*, 181–192. https://doi.org/10.1016/j.wasman.2017.05.029

Al-jaf, A., & Al-ameen, J. (2021). Comparison of a traditional landfill and a mechanically-biologically treated waste landfill (Case study; Kirkuk landfill). *Tikrit Journal of Engineering Sciences*, *28*(2), 73–79. https://doi.org/10.25130/tjes.28.2.06

Arya, L. M., Leij, F. J., Shouse, P. J., & van Genuchten, M. Th. (1999). Relationship between the Hydraulic Conductivity Function and the Particle-Size Distribution. *Soil Science Society of America Journal*, *63*(5), 1063–1070. https://doi.org/10.2136/sssaj1999.6351063x

Bandara, Y. W., Gamage, P., & Gunarathne, D. S. (2020). Hot water washing of rice husk for ash removal: The effect of washing temperature, washing time and particle size. *Renewable Energy*, *153*, 646–652. https://doi.org/10.1016/j.renene.2020.02.038

Beiyuan, J., Tsang, D. C. W., Ok, Y. S., Zhang, W., Yang, X., Baek, K., & Li, X. D. (2016). Integrating EDDS-enhanced washing with low-cost stabilization of metal-contaminated soil from an e-waste recycling site. *Chemosphere*, *159*, 426–432. https://doi.org/10.1016/j.chemosphere.2016.06.030

Beiyuan, J., Tsang, D. C. W., Valix, M., Baek, K., Ok, Y. S., Zhang, W., Bolan, N. S., Rinklebe, J., & Li, X. D. (2018). Combined application of EDDS and EDTA for removal of potentially toxic elements under multiple soil washing schemes. *Chemosphere*, *205*, 178–187. https://doi.org/10.1016/j.chemosphere.2018.04.081

Beiyuan, J., Tsang, D. C. W., Valix, M., Zhang, W., Yang, X., Ok, Y. S., & Li, X. D. (2017). Selective dissolution followed by EDDS washing of an e-waste contaminated soil: Extraction efficiency, fate of residual metals, and impact on soil environment. *Chemosphere*, *166*, 489–496. https://doi.org/10.1016/j.chemosphere.2016.09.110

Berge, N. D., Reinhart, D. R., & Batarseh, E. S. (2009). An assessment of bioreactor landfill costs and benefits. *Waste Management*, *29*(5), 1558–1567. https://doi.org/10.1016/j.wasman.2008.12.010

Berge, N. D., Reinhart, D. R., Dietz, J., & Townsend, T. (2006). In situ ammonia removal in bioreactor landfill leachate. *Waste Management*, *26*(4), 334–343. https://doi.org/10.1016/j.wasman.2005.11.003

Bogush, A. A., Stegemann, J. A., & Roy, A. (2019). Changes in composition and lead speciation due to water washing of air pollution control residue from municipal waste incineration. *Journal of Hazardous Materials*, *361*(February 2018), 187–199. https://doi.org/10.1016/j.jhazmat.2018.08.051

Bogush, A. A., Stegemann, J. A., Zhou, Q., Wang, Z., Zhang, B., Zhang, T., Zhang, W., & Wei, J. (2020). Co-processing of raw and washed air pollution control residues from energy-from-waste facilities in the cement kiln. *Journal of Cleaner Production*, *254*, 119924. https://doi.org/10.1016/j.jclepro.2019.119924

Bolyard, S. C., & Reinhart, D. R. (2016). Application of landfill treatment approaches for stabilization of municipal solid waste. *Waste Management*, *55*, 22–30. https://doi.org/10.1016/j.wasman.2016.01.024

Bramryd, T., & Binder, M. (2007). Environmental and Efficiency Evaluation of Different Pre-Stabilization Techniques for Solid Wastes. *Sustainable Land Filling*.

Bruce M.McEnroe & Member ASCE. (1989). Study drainage or landfill covers and bottom liners. *Journal of Environmental Engineering*, 115(16): 1114-1122

Calli, B., Mertoglu, B., Inanc, B., & Yenigun, O. (2005). Methanogenic diversity in anaerobic bioreactors under extremely high ammonia levels. *Enzyme and Microbial Technology*, *37*(4), 448–455. https://doi.org/10.1016/j.enzmictec.2005.03.013

Chen, S. J., Ma, Y. J., Wang, J., Chen, D., Luo, X. J., & Mai, B. X. (2009). Brominated flame retardants in children's toys: Concentration, composition, and children's exposure and risk assessment. *Environmental Science and Technology*, *43*(11), 4200–4206. https://doi.org/10.1021/es9004834

Chen, W. S., Chang, F. C., Shen, Y. H., Tsai, M. S., & Ko, C. H. (2012). Removal of chloride from MSWI fly ash. *Journal of Hazardous Materials*, *237–238*, 116–120. https://doi.org/10.1016/j.jhazmat.2012.08.010

Chen, X., Bi, Y., Zhang, H., & Wang, J. (2016). Chlorides Removal and Control through Water-washing Process on MSWI Fly Ash. *Procedia Environmental Sciences*, *31*, 560–566. https://doi.org/10.1016/j.proenv.2016.02.086

Cheng, S., Lin, Q., Wang, Y., Luo, H., Huang, Z., Fu, H., Chen, H., & Xiao, R. (2020). The removal of Cu, Ni, and Zn in industrial soil by washing with EDTA-organic acids. *Arabian Journal of Chemistry*, *13*(4), 5160–5170. https://doi.org/10.1016/j.arabjc.2020.02.015

Chiang, K. Y., & Hu, Y. H. (2010). Water washing effects on metals emission reduction during municipal solid waste incinerator (MSWI) fly ash melting process. *Waste Management*, *30*(5), 831–838. https://doi.org/10.1016/j.wasman.2009.12.009

Chimenos, J. M., & Ferna, A. I. (2005). *Optimizing the APC residue washing process to minimize the release of chloride and heavy metals*. *25*, 686–693. https://doi.org/10.1016/j.wasman.2004.12.014

Christensen, T. H. (2011). *Solid waste technology & management*. Wiley.

Colangelo, F., Cioffi, R., Montagnaro, F., & Santoro, L. (2012). Soluble salt removal from MSWI fly ash and its stabilization for safer disposal and recovery as road basement material. *Waste Management*, *32*(6), 1179–1185. https://doi.org/10.1016/j.wasman.2011.12.013

Consonni, S., Giugliano, M., Massarutto, A., Ragazzi, M., & Saccani, C. (2011). Material and energy recovery in integrated waste management systems: Project overview and main results. *Waste Management*, *31*(9–10), 2057–2065. https://doi.org/10.1016/j.wasman.2011.04.016

Corvellec, H., Stowell, A. F., & Johansson, N. (2022). Critiques of the circular economy. *Journal of Industrial Ecology*, *26*(2), 421–432. https://doi.org/10.1111/jiec.13187

Cossu. (2016). Back to Earth Sites: From "nasty and unsightly" landfilling to final sink and geological repository. In *Waste Management* (Vol. 55, pp. 1–2). Elsevier Ltd. https://doi.org/10.1016/j.wasman.2016.07.028

Cossu, R. (2010). Technical evolution of landfilling. In *Waste Management* (Vol. 30, Issue 6, pp. 947–948). https://doi.org/10.1016/j.wasman.2010.03.003

Cossu, R., Grossule, V., & Lavagnolo, M. C. (2020). What about residues from circular economy and role of landfilling? In *Detritus* (Vol. 9, Issue March, pp. 1–3). Cisa Publisher - Eurowaste Srl. https://doi.org/10.31025/2611-4135/2020.13920

Cossu, R., & Hans Albert, van der S. (2014). *Sustainable Landfilling*. CISA.

Cossu, R., & Lai, T. (2012). Washing of waste prior to landfilling. *Waste Management*, *32*(5), 869–878. https://doi.org/10.1016/j.wasman.2011.12.005

Cossu, R., & Lai, T. (2013). Washing treatment of automotive shredder residue (ASR). *Waste Management*, *33*(8), 1770–1775. https://doi.org/10.1016/j.wasman.2013.04.007

Cossu, R., & Lai, T. (2015). Automotive shredder residue (ASR) management: An overview. *Waste Management*, *45*, 143–151. https://doi.org/10.1016/j.wasman.2015.07.042

Cossu, R., Lai, T., & Pivnenko, K. (2012). Waste washing pre-treatment of municipal and special waste. *Journal of Hazardous Materials*, *207–208*, 65–72. https://doi.org/10.1016/J.JHAZMAT.2011.07.121

Cossu, R., Morello, L., Raga, R., & Cerminara, G. (2016). Biogas production enhancement using semi-aerobic pre-aeration in a hybrid bioreactor landfill. *Waste Management*, *55*, 83–92. https://doi.org/10.1016/j.wasman.2015.10.025

Cossu, R., & Stegmann, R. (2018). *Solid Waste Landfilling: Concepts, Processes, Technology*. Elsevier.

Danthurebandara, M., van Passel, S., Machiels, L., & van Acker, K. (2015). Valorization of thermal treatment residues in Enhanced Landfill Mining: Environmental and economic evaluation. *Journal of Cleaner Production*, *99*, 275–285. https://doi.org/10.1016/j.jclepro.2015.03.021

Davis, B. A. P., & Singh, I. (1995). Washing of Zinc (II) from contaminated soil column. *Journal of Environmental Engineering.* 121(2), 174–185.

Demirbas, A. (2011). Waste management, waste resource facilities and waste conversion processes. *Energy Conversion and Management*, *52*(2), 1280–1287. https://doi.org/10.1016/j.enconman.2010.09.025

Deng, L., Zhang, T., & Che, D. (2013). Effect of water washing on fuel properties, pyrolysis and combustion characteristics, and ash fusibility of biomass. *Fuel Processing Technology*, *106*, 712–720. https://doi.org/10.1016/j.fuproc.2012.10.006

Dhokhikah, Y., & Trihadiningrum, Y. (2012). Solid waste management in Asian developing countries: Challenges and opportunities. *J. Appl. Environ. Biol. Sci*, *2*(7), 329–335. https://www.researchgate.net/publication/284942823

Di, C., Ramadori, R., & Lopez, A. (2006). *Combined biological and chemical degradation for treating a mature municipal landfill leachate*. *31*, 118–124. https://doi.org/10.1016/j.bej.2006.06.002

di Gianfilippo, M., Costa, G., Verginelli, I., Gavasci, R., & Lombardi, F. (2016). Analysis and interpretation of the leaching behaviour of waste thermal treatment bottom ash by batch and column tests. *Waste Management*, *56*, 216–228. https://doi.org/10.1016/j.wasman.2016.07.034

Dijkstra, J. J., Van Der Sloot, H. A., & Comans, R. N. J. (2006). The leaching of major and trace elements from MSWI bottom ash as a function of pH and time. *Applied Geochemistry*, *21*(2), 335–351. https://doi.org/10.1016/j.apgeochem.2005.11.003

Erses, A. S., Onay, T. T., & Yenigun, O. (2008). Comparison of aerobic and anaerobic degradation of municipal solid waste in bioreactor landfills. *Bioresource Technology*, *99*(13), 5418–5426. https://doi.org/10.1016/j.biortech.2007.11.008

Ettler, V., & Johan, Z. (2014). 12 years of leaching of contaminants from Pb smelter slags: Geochemical/mineralogical controls and slag recycling potential. *Applied Geochemistry*, *40*, 97–103. https://doi.org/10.1016/j.apgeochem.2013.11.001

Evangelista, R. A., & Zownir, A. P. (1989). Lead contamination removal by soil washing. In *Proceedings - A&WMA Annual Meeting* (Vol. 6).

Faravash, R. S., & Ashtiani, F. Z. (2007). The effect of pH, ethanol volume and acid washing time on the yield of pectin extraction from peach pomace. *International Journal of Food Science and Technology*, *2001*, 1177–1187. https://doi.org/10.1111/j.1365-2621.2006.01324.x

Fei, F., Wen, Z., Huang, S., & de Clercq, D. (2018). Mechanical biological treatment of municipal solid waste: Energy efficiency, environmental impact and economic feasibility analysis. *Journal of Cleaner Production*, *178*, 731–739. https://doi.org/10.1016/j.jclepro.2018.01.060

Freyssinet, P., Piantone, P., Azaroual, M., Itard, Y., Clozel-Leloup, B., Guyonnet, D., & Baubron, J. C. (2002). Chemical changes and leachate mass balance of municipal solid waste bottom ash submitted to weathering. *Waste Management,* 22,159-174.

Galvín, A. P., Ayuso, J., Jiménez, J. R., & Agrela, F. (2012). Comparison of batch leaching tests and influence of pH on the release of metals from construction and demolition wastes. *Waste Management*, *32*(1), 88–95. https://doi.org/10.1016/j.wasman.2011.09.010

Garrido, Y., Marín, A., Tudela, J. A., Allende, A., & Gil, M. I. (2019). Chlorate uptake during washing is influenced by product type and cut piece size, as well as washing time and wash water content. *Postharvest Biology and Technology*, *151*(February), 45–52. https://doi.org/10.1016/j.postharvbio.2019.01.014

Grathwohl, P. (2014). On equilibration of pore water in column leaching tests. *Waste Management*, *34*(5), 908–918. https://doi.org/10.1016/j.wasman.2014.02.012

Grathwohl, P., & Susset, B. (2009). Comparison of percolation to batch and sequential leaching tests: Theory and data. *Waste Management*, *29*(10), 2681–2688. https://doi.org/10.1016/j.wasman.2009.05.016

Grossule, V. (2020). Final quality of a sustainable landfill and postclosure management. *Detritus*, *13*(September), 148–159. https://doi.org/10.31025/2611-4135/2020.13999

Grossule, V., & Lavagnolo, M. C. (2020). Lab tests on semi-aerobic landfilling of MSW under varying conditions of water availability and putrescible waste content. *Journal of Environmental Management*, *256*. https://doi.org/10.1016/j.jenvman.2019.109995

Grossule, V., Morello, L., Cossu, R., & Lavagnolo, M. C. (2018). Bioreactor landfills: Comparison and kinetics of the different systems. *Detritus*, *3*, 100–113. https://doi.org/10.31025/2611-4135/2018.13703

Grossule, V., & Stegmann, R. (2020). Problems in traditional landfilling and proposals for solutions based on sustainability. *Detritus*, *12*, 78–91. https://doi.org/10.31025/2611-4135/2020.14000

Gu, N., Liu, J., Ye, J., Chang, N., & Li, Y.-Y. (2019). Bioenergy, ammonia and humic substances recovery from municipal solid waste leachate: A review and process integration. *Bioresource Technology*, *293*, 122159. https://doi.org/10.1016/J.BIORTECH.2019.122159

Gudka, B., Jones, J. M., Lea-Langton, A. R., Williams, A., & Saddawi, A. (2016). A review of the mitigation of deposition and emission problems during biomass combustion through washing pre-treatment. *Journal of the Energy Institute*, *89*(2), 159–171. https://doi.org/10.1016/j.joei.2015.02.007

Gupta, N., Gedam, V. v., Moghe, C., & Labhasetwar, P. (2019). Comparative assessment of batch and column leaching studies for heavy metals release from Coal Fly Ash Bricks and Clay Bricks. *Environmental Technology and Innovation*, *16*. https://doi.org/10.1016/j.eti.2019.100461

Heimovaara, T. J., Cossu, R., & van der Sloot, H. A. (2014). State of the art and perspectives for sustainable landfills. In *Sustainable Landfilling*. CISA.

Hilber, T., Maier, J., Scheffknecht, G., Agraniotis, M., Grammelis, P., Kakaras, E., Glorius, T., Becker, U., Derichs, W., Schiffer, H. P., de Jong, M., & Torri, L. (2007). Advantages and possibilities of solid recovered fuel cocombustion in the European energy sector. *Journal of the Air and Waste Management Association*, *57*(10), 1178–1189. https://doi.org/10.3155/1047-3289.57.10.1178

Hobson, K. (2021). The limits of the loops: critical environmental politics and the Circular Economy. *Environmental Politics*, *30*(1–2), 161–179. https://doi.org/10.1080/09644016.2020.1816052

Holma, A., Koponen, K., Antikainen, R., Lardon, L., Leskinen, P., & Roux, P. (2013). Current limits of life cycle assessment framework in evaluating environmental sustainability - Case of two evolving biofuel technologies. *Journal of Cleaner Production*, *54*, 215–228. https://doi.org/10.1016/j.jclepro.2013.04.032

Huppes, G., & Ishikawa, M. (2009). Eco-efficiency guiding micro-level actions towards sustainability: Ten basic steps for analysis. *Ecological Economics*, *68*(6), 1687–1700. https://doi.org/10.1016/j.ecolecon.2009.01.007

Huy, V. Q., Kohata, Y., & Yoshida, H. (2020). The elevated temperature and gas component within an operating semi-aerobic landfill. *Detritus*, *10*(June), 147–159. https://doi.org/10.31025/2611-4135/2020.13945

Irfah, N., Pauzi, M., Omar, H., & Misran, H. (2010.). Settlement model of waste soil for dumping area in Malaysia. *Electronic Journal of Geotechnical Engineering.* 15: 1917-1929. https://www.researchgate.net/publication/228509175

Iskander, S. M., Zhao, R., Pathak, A., Gupta, A., Pruden, A., Novak, J. T., & He, Z. (2018). A review of landfill leachate induced ultraviolet quenching substances: Sources, characteristics, and treatment.

In *Water Research* (Vol. 145, pp. 297–311). Elsevier Ltd. https://doi.org/10.1016/j.watres.2018.08.035

Jha, A. K., Singh, S. K., Singh, G. P., & Gupta, P. K. (2011). Sustainable municipal solid waste management in low income group of cities: a review. *Tropical Ecology*, *52*(1), 123–131. www.tropecol.com

Jiang, Y., Xi, B., Li, X., Zhang, L., & Wei, Z. (2009). Effect of water-extraction on characteristics of melting and solidification of fly ash from municipal solid waste incinerator. *Journal of Hazardous Materials*, *161*(2–3), 871–877. https://doi.org/10.1016/j.jhazmat.2008.04.033

Jiao, F., Zhang, L., Dong, Z., Namioka, T., Yamada, N., & Ninomiya, Y. (2016). Study on the species of heavy metals in MSW incineration fly ash and their leaching behavior. *Fuel Processing Technology*, *152*, 108–115. https://doi.org/10.1016/j.fuproc.2016.06.013

Ju, Y., Zhang, K., Yang, T., Deng, D., Qiao, J., Wang, P., You, Y., Du, L., Chen, G., Kołodyńska, D., & Dionysiou, D. D. (2020). The influence of a washing pretreatment containing phosphate anions on single-mode microwave-based detoxification of fly ash from municipal solid waste incinerators. *Chemical Engineering Journal*, *387*(January). https://doi.org/10.1016/j.cej.2020.124053

Keulen, A., van Zomeren, A., Harpe, P., Aarnink, W., Simons, H. A. E., & Brouwers, H. J. H. (2016). High performance of treated and washed MSWI bottom ash granulates as natural aggregate replacement within earth-moist concrete. *Waste Management*, *49*(2016), 83–95. https://doi.org/10.1016/j.wasman.2016.01.010

Khan, L. I., & Abumaizar, R. (1996). Laboratory Investigation of Heavy Metal Removal by Soil Washing. *Journal of the Air and Waste Management Association*, *46*(8), 765–768. https://doi.org/10.1080/10473289.1996.10467512

Kim, K., Yoon, S., Kwon, H. ah, & Choi, Y. (2020). Effects of treatment agents during acid washing and pH neutralization on the fertility of heavy metal-impacted dredged marine sediment as plant-growing soil. *Environmental Pollution*, *267*, 115466. https://doi.org/10.1016/j.envpol.2020.115466

Kim, S. Y., Matsuto, T., & Tanaka, N. (2003). Evaluation of pre-treatment methods for landfill disposal of residues from municipal solid waste incineration. *Waste Management and Research*, *21*(5), 416–423. https://doi.org/10.1177/0734242X0302100504

Kjeldsen, P., Barlaz, M. A., Rooker, A. P., Baun, A., Ledin, A., & Christensen, T. H. (2002). Present and long-term composition of MSW landfill leachate: A review. In *Critical Reviews in Environmental Science and Technology* (Vol. 32, Issue 4, pp. 297–336). https://doi.org/10.1080/10643380290813462

Koda, E., Miszkowska, A., & Stępień, S. (2016). Quality control of Non-Woven geotextiles used in a drainage system in an old remedial landfill. In Proceedings. *Geoscience (Chicago) 2016 GSP 271*. 254–263. https://doi.org/10.1061/9780784480144.025

Komonweeraket, K., Cetin, B., Aydilek, A. H., Benson, C. H., & Edil, T. B. (2015). Effects of pH on the leaching mechanisms of elements from fly ash mixed soils. *Fuel*, *140*, 788–802. https://doi.org/10.1016/j.fuel.2014.09.068

Korhonen, J., Honkasalo, A., & Seppälä, J. (2018). Circular Economy: The Concept and its Limitations. *Ecological Economics*, *143*, 37–46. https://doi.org/10.1016/j.ecolecon.2017.06.041

Laner, D., Crest, M., Scharff, H., Morris, J. W. F., & Barlaz, M. A. (2012). A review of approaches for the long-term management of municipal solid waste landfills. *Waste Management*, *32*(3), 498–512. https://doi.org/10.1016/j.wasman.2011.11.010

Laner, D., Fellner, J., & Brunner, P. H. (2011). Future landfill emissions and the effect of final cover installation - A case study. *Waste Management*, *31*(7), 1522–1531. https://doi.org/10.1016/j.wasman.2011.02.022

Liaw, S. B., & Wu, H. (2013). Leaching characteristics of organic and inorganic matter from biomass by water: Differences between batch and semi-continuous operations. *Industrial and Engineering Chemistry Research*, *52*(11), 4280–4289. https://doi.org/10.1021/ie3031168

Lin, W. Y., Heng, K. S., Nguyen, M. Q., Ho, J. R. I., Mohamed Noh, O. A. bin, Zhou, X. D., Liu, A., Ren, F., & Wang, J. Y. (2017). Evaluation of the leaching behavior of incineration bottom ash using seawater: A comparison with standard leaching tests. *Waste Management*, *62*, 139–146. https://doi.org/10.1016/j.wasman.2016.12.003

Long, Y., Hu, L. fang, & Shen, D. S. (2009). Nitrogen transformation in the hybrid bioreactor landfill. *Bioresource Technology*, *100*(9), 2527–2533. https://doi.org/10.1016/j.biortech.2008.11.036

López Meza, S., Kalbe, U., Berger, W., & Simon, F. G. (2010). Effect of contact time on the release of contaminants from granular waste materials during column leaching experiments. *Waste Management*, *30*(4), 565–571. https://doi.org/10.1016/j.wasman.2009.11.022

Lü, F., He, P., Guo, M., Yang, N., & Shao, L. (2012). Ammonium-dependent regulation of aerobic methane-consuming bacteria in landfill cover soil by leachate irrigation. *Journal of Environmental Sciences*, *24*(4), 711–719. https://doi.org/10.1016/S1001-0742(11)60813-9

Luo, H., Cheng, Y., He, D., & Yang, E. (2019a). Science of the Total Environment Review of leaching behavior of municipal solid waste incineration. *Science of the Total Environment*, *668*, 90–103. https://doi.org/10.1016/j.scitotenv.2019.03.004

Luo, H., Cheng, Y., He, D., & Yang, E. H. (2019b). Review of leaching behavior of municipal solid waste incineration (MSWI) ash. In *Science of the Total Environment* (Vol. 668, pp. 90–103). Elsevier B.V. https://doi.org/10.1016/j.scitotenv.2019.03.004

Luo, H., Wu, Y., Zhao, A., Kumar, A., Liu, Y., Cao, B., & Yang, E. H. (2017). Hydrothermally synthesized porous materials from municipal solid waste incineration bottom ash and their interfacial interactions with chloroaromatic compounds. *Journal of Cleaner Production*, *162*, 411–419. https://doi.org/10.1016/j.jclepro.2017.06.082

Luo, H., Zeng, Y., Cheng, Y., He, D., & Pan, X. (2020). Recent advances in municipal landfill leachate: A review focusing on its characteristics, treatment, and toxicity assessment. In *Science of the Total Environment* (Vol. 703). Elsevier B.V. https://doi.org/10.1016/j.scitotenv.2019.135468

Ma, H., Guo, Y., Qin, Y., & Li, Y. Y. (2018). Nutrient recovery technologies integrated with energy recovery by waste biomass anaerobic digestion. In *Bioresource Technology* (Vol. 269, pp. 520–531). Elsevier Ltd. https://doi.org/10.1016/j.biortech.2018.08.114

Mali Sandip, T., Khare Kanchan, C., & Biradar Ashok, H. (2012). Enhancement of methane production and bio-stabilisation of municipal solid waste in anaerobic bioreactor landfill. *Bioresource Technology*, *110*, 10–17. https://doi.org/10.1016/j.biortech.2011.12.027

Mangesh, G., Richard, C., Callum, C., Zulfiqur, A., & Graham, H. (2015). Analysis of waste hierarchy in the European waste directive 2008/98/EC. *Waste Management*, *39*, 305–313.

Mangialardi, T. (2004). Effects of a washing pre-treatment of municipal solid waste incineration fly ash on the hydration behaviour and properties of ash-Portland cement mixtures. *Advances in Cement Research*, *16*(2), 45–54. https://doi.org/10.1680/adcr.2004.16.2.45

Mantis, I., Voutsa, D., & Samara, C. (2005). Assessment of the environmental hazard from municipal and industrial wastewater treatment sludge by employing chemical and biological methods.

Ecotoxicology and Environmental Safety, *62*(3), 397–407. https://doi.org/10.1016/j.ecoenv.2004.12.010

Massarutto, A. (2015). Economic aspects of thermal treatment of solid waste in a sustainable WM system. *Waste Management*, *37*, 45–57. https://doi.org/10.1016/j.wasman.2014.08.024

Maszkowska, J., Kołodziejska, M., Białk-Bielińska, A., Mrozik, W., Kumirska, J., Stepnowski, P., Palavinskas, R., Krüger, O., & Kalbe, U. (2013). Column and batch tests of sulfonamide leaching from different types of soil. *Journal of Hazardous Materials*, *260*, 468–474. https://doi.org/10.1016/j.jhazmat.2013.05.053

Mattila, T. J., Pakarinen, S., & Sokka, L. (2010). Quantifying the total environmental impacts of an industrial symbiosis-a comparison of process-, hybrid and input-output life cycle assessment. *Environmental Science and Technology*, *44*(11), 4309–4314. https://doi.org/10.1021/es902673m

Mitrano, D. M., Mehrabi, K., Dasilva, Y. A. R., & Nowack, B. (2017). Mobility of metallic (nano)particles in leachates from landfills containing waste incineration residues. *Environmental Science: Nano*, *4*(2), 480–492. https://doi.org/10.1039/c6en00565a

Montejo, C., Tonini, D., Márquez, M. del C., & Fruergaard Astrup, T. (2013). Mechanical-biological treatment: Performance and potentials. An LCA of 8 MBT plants including waste characterization. *Journal of Environmental Management*, *128*, 661–673. https://doi.org/10.1016/j.jenvman.2013.05.063

Morello, L., Cossu, R., Raga, R., Pivato, A., & Lavagnolo, M. C. (2016). Recirculation of reverse osmosis concentrate in lab-scale anaerobic and aerobic landfill simulation reactors. *Waste Management*, *56*, 262–270. https://doi.org/10.1016/j.wasman.2016.07.030

Nanda, S., & Berruti, F. (2021). Municipal solid waste management and landfilling technologies: a review. In *Environmental Chemistry Letters* (Vol. 19, Issue 2, pp. 1433–1456). Springer Science and Business Media Deutschland GmbH. https://doi.org/10.1007/s10311-020-01100-y

Nestor, P., Djonga, D., Atchana, J., Nankap, A., Mbakop, F. K., Djackba, R., & Tame, A. (2020). *Biogas purification by Alkali-based column washing extracted from wood ash and banana stalks*. *8*(1), 6–14. https://doi.org/10.11648/j.sjee.20200801.12

Norbu, T., Visvanathan, C., & Basnayake, B. (2005). Pretreatment of municipal solid waste prior to landfilling. *Waste Management*, *25*(10), 997–1003. https://doi.org/10.1016/j.wasman.2005.06.006

Olgun, A., Atar, N., & Wang, S. (2013). Batch and column studies of phosphate and nitrate adsorption on waste solids containing boron impurity. *Chemical Engineering Journal*, *222*, 108–119. https://doi.org/10.1016/j.cej.2013.02.029

Pantini, S., Verginelli, I., & Lombardi, F. (2015). Analysis and modeling of metals release from MBT wastes through batch and up-flow column tests. *Waste Management*, *38*(1), 22–32. https://doi.org/10.1016/j.wasman.2014.12.002

Parajuly, K., Fitzpatrick, C., Muldoon, O., & Kuehr, R. (2020). Behavioral change for the circular economy: A review with focus on electronic waste management in the EU. In *Resources, Conservation and Recycling: X* (Vol. 6). Elsevier B.V. https://doi.org/10.1016/j.rcrx.2020.100035

Paritosh, K., Kushwaha, S. K., Yadav, M., Pareek, N., Chawade, A., & Vivekanand, V. (2017). Food Waste to Energy: An Overview of Sustainable Approaches for Food Waste Management and Nutrient Recycling. In *BioMed Research International* (Vol. 2017). Hindawi Limited. https://doi.org/10.1155/2017/2370927

Parodi, A., Feuillade-Cathalifaud, G., Pallier, V., & Mansour, A. A. (2011). Optimization of municipal solid waste leaching test procedure: Assessment of the part of hydrosoluble organic compounds. *Journal of Hazardous Materials*, *186*(2–3), 991–998. https://doi.org/10.1016/j.jhazmat.2010.11.090

Pecorini, I., Baldi, F., Bacchi, D., Carnevale, E. A., & Corti, A. (2017). Leaching behaviour of hazardous waste under the impact of different ambient conditions. *Waste Management*, *63*, 96–106. https://doi.org/10.1016/j.wasman.2016.10.037

Pires, A., & Martinho, G. (2019). Waste hierarchy index for circular economy in waste management. *Waste Management*, *95*, 298–305. https://doi.org/10.1016/j.wasman.2019.06.014

Pivnenko, K., & Astrup, T. F. (2016). The challenge of chemicals in material lifecycles. In *Waste Management* (Vol. 56, pp. 1–2). Elsevier Ltd. https://doi.org/10.1016/j.wasman.2016.08.016

Pivnenko, K., Eriksen, M. K., Martín-Fernández, J. A., Eriksson, E., & Astrup, T. F. (2016a). Recycling of plastic waste: Presence of phthalates in plastics from households and industry. *Waste Management*, *54*, 44–52. https://doi.org/10.1016/j.wasman.2016.05.014

Pivnenko, K., Laner, D., & Astrup, T. F. (2016b). Material cycles and chemicals: Dynamic material flow analysis of contaminants in paper recycling. *Environmental Science and Technology*, *50*(22), 12302–12311. https://doi.org/10.1021/acs.est.6b01791

Ponsá, S., Gea, T., & Sánchez, A. (2010). The effect of storage and mechanical pretreatment on the biological stability of municipal solid wastes. *Waste Management*, *30*(3), 441–445. https://doi.org/10.1016/j.wasman.2009.10.020

Prateep Na Talang, R., & Sirivithayapakorn, S. (2021). Environmental and financial assessments of open burning, open dumping and integrated municipal solid waste disposal schemes among different income groups. *Journal of Cleaner Production*, *312*. https://doi.org/10.1016/j.jclepro.2021.127761

Raga, R., & Cossu, R. (2013). Bioreactor tests preliminary to landfill in situ aeration: A case study. *Waste Management*, *33*(4), 871–880. https://doi.org/10.1016/j.wasman.2012.11.014

Read, A. D., Hudgins, M., & Phillips, P. (2001). Aerobic landfill test cells and their implications for sustainable waste disposal. In *The Geographical Journal* (Vol. 167, Issue 3).

Ritzkowski, M., & Stegmann, R. (2013). Landfill aeration within the scope of post-closure care and its completion. *Waste Management*, *33*(10), 2074–2082. https://doi.org/10.1016/j.wasman.2013.02.004

Romano, G., Rapposelli, A., & Marrucci, L. (2019). Improving waste production and recycling through zero-waste strategy and privatization: An empirical investigation. *Resources, Conservation and Recycling*, *146*, 256–263. https://doi.org/10.1016/j.resconrec.2019.03.030

Rowe, R. K. (2005). *Long-term performance of contaminant barrier systems* (Vol. 55, Issue 9).

Rubinos, D. A., & Spagnoli, G. (2018). Utilization of waste products as alternative landfill liner and cover materials–A critical review. *Critical Reviews in Environmental Science and Technology*, *48*(4), 376–438. https://doi.org/10.1080/10643389.2018.1461495

Saddawi, A., Jones, J. M., Williams, A., & Le Coeur, C. (2012). Commodity fuels from biomass through pretreatment and torrefaction: Effects of mineral content on torrefied fuel characteristics and quality. *Energy and Fuels*, *26*(11), 6466–6474. https://doi.org/10.1021/ef2016649

Sazali, N., Harun, Z., & Sazali, N. (2020). A review on batch and column adsorption of various adsorbent towards the removal of heavy metal. *Journal of Advanced Research in Fluid Mechanics and Thermal Sciences*, *67*(2), 66–88.

Sekhohola-Dlamini, L., & Tekere, M. (2020). Microbiology of municipal solid waste landfills: a review of microbial dynamics and ecological influences in waste bioprocessing. In *Biodegradation* (Vol. 31, Issues 1–2). Springer. https://doi.org/10.1007/s10532-019-09890-x

Soyez, K & Plickert, S (2002). Mechanical-Biological pre-treatment of waste: State of the Art and Potentials of Biotechnology. *Acta Biotechnol*. 22, 3-4.

Sun, X., & Yi, Y. (2020). pH evolution during water washing of incineration bottom ash and its effect on removal of heavy metals. *Waste Management*, *104*, 213–219. https://doi.org/10.1016/j.wasman.2020.01.023

Tang, Q., Gu, F., Zhang, Y., Zhang, Y., & Mo, J. (2018). Impact of biological clogging on the barrier performance of landfill liners. *Journal of Environmental Management*, *222*, 44–53. https://doi.org/10.1016/j.jenvman.2018.05.039

Tisserant, A., Pauliuk, S., Merciai, S., Schmidt, J., Fry, J., Wood, R., & Tukker, A. (2017). Solid Waste and the Circular Economy: A Global Analysis of Waste Treatment and Waste Footprints. *Journal of Industrial Ecology*, *21*(3), 628–640. https://doi.org/10.1111/jiec.12562

Tiwari, M. K., Bajpai, S., Dewangan, U. K., & Tamrakar, R. K. (2015). Suitability of leaching test methods for fly ash and slag: A review. *Journal of Radiation Research and Applied Sciences*, *8*(4), 523–537. https://doi.org/10.1016/j.jrras.2015.06.003

Torrente-Velásquez, J. M., Giampietro, M., Ripa, M., & Chifari, R. (2020). Landfill reactions to society actions: The case of local and global air pollutants of Cerro Patacón in Panama. *Science of the Total Environment*, *706*. https://doi.org/10.1016/j.scitotenv.2019.135988

Townsend, T. G., Powell, J., & Jain, P. (2015). Sustainable practices for landfill design and operation. *Springer.*

Trebouet, D., Schlumpf, J. P., Jaouen, P., & Quemeneur, F. (2001). Stabilized landfill leachate treatment by combined physico-chemical nanofiltration processes. *Water Research. 35*(12), 2935–2942.

Tsang, D. C. W., & Yip, A. C. K. (2014). Comparing chemical-enhanced washing and waste-based stabilisation approach for soil remediation. *Journal of Soils and Sediments*, *14*(5), 936–947. https://doi.org/10.1007/s11368-013-0831-y

Vallini, G., Pera, A., Frassinetti, S., Cecchi, F., & Cecchi, F. (1991). Co-composting for managing effluent from thermophilic anaerobic digestion of municipal solid waste. *Environmental Technology (United Kingdom)*, *12*(12), 1137–1145. https://doi.org/10.1080/09593339109385114

Vaverková, M. D. (2019). Landfill impacts on the environment— review. In *Geosciences (Switzerland)* (Vol. 9, Issue 10). MDPI AG. https://doi.org/10.3390/geosciences9100431

Velts, O., Hautaniemi, M., Kallas, J., Kuosa, M., & Kuusik, R. (2010). Modeling calcium dissolution from oil shale ash: Part 2. Continuous washing of the ash layer. *Fuel Processing Technology*, *91*(5), 491–495. https://doi.org/10.1016/j.fuproc.2009.12.009

Voegelin, A., Barmettler, K., & Kretzschmar, R. (2003). Heavy Metal Release from Contaminated Soils. *Journal of Environmental Quality*, *32*(3), 865–875. https://doi.org/10.2134/jeq2003.8650

Wang, K. S., Sun, C. J., & Yeh, C. C. (2002). The thermotreatment of MSW incinerator fly ash for use as an aggregate: A study of the characteristics of size-fractioning. *Resources, Conservation and Recycling*, *35*(3), 177–190. https://doi.org/10.1016/S0921-3449(01)00121-5

Wang, Q., Yang, J., Wang, Q., & Wu, T. (2009). Effects of water-washing pretreatment on bioleaching of heavy metals from municipal solid waste incinerator fly ash. *Journal of Hazardous Materials*, *162*(2–3), 812–818. https://doi.org/10.1016/j.jhazmat.2008.05.125

Wang, X., Wang, M., Zou, D., Wu, C., Li, T., Gao, M., Liu, S., Wang, Q., & Shimaoka, T. (2020). Comparative study on inorganic Cl removal of municipal solid waste fly ash using different types and concentrations of organic acids. *Chemosphere*, *261*, 127754. https://doi.org/10.1016/j.chemosphere.2020.127754

Weiss, M., Haufe, J., Carus, M., Brandão, M., Bringezu, S., Hermann, B., & Patel, M. K. (2012). A Review of the Environmental Impacts of Biobased Materials. *Journal of Industrial Ecology*, *16*(SUPPL.1). https://doi.org/10.1111/j.1530-9290.2012.00468.x

Westlake, K. (1997). Sustainable Landfill—Possibility or Pipe-Dream? *Waste Management & Research: The Journal for a Sustainable Circular Economy*, *15*(5), 453–461. https://doi.org/10.1177/0734242x9701500502

Widomski, M. K., Gleń, P., & Łagód, G. (2017). *Sustainable Landfilling as Final Step of Municipal Waste Management System Zrównoważone składowiska jako końcowy etap systemu gospodarki odpadami komunalnymi* (Vol. 12). https://ssrn.com/abstract=2947628

Xia, Y., He, P., Shao, L., & Zhang, H. (2017). Metal distribution characteristic of MSWI bottom ash in view of metal recovery. *Journal of Environmental Sciences (China)*, *52*, 178–189. https://doi.org/10.1016/j.jes.2016.04.016

Xu, Q., Jin, X., Ma, Z., Tao, H., & Ko, J. H. (2014). Methane production in simulated hybrid bioreactor landfill. *Bioresource Technology*, *168*, 92–96. https://doi.org/10.1016/j.biortech.2014.03.036

Yaman, C., Anil, I., & Alagha, O. (2020). Potential for greenhouse gas reduction and energy recovery from MSW through different waste management technologies. *Journal of Cleaner Production*, *264*. https://doi.org/10.1016/j.jclepro.2020.121432

Yan, D., Peng, Z., Yu, L., Sun, Y., Yong, R., & Helge Karstensen, K. (2018). Characterization of heavy metals and PCDD/Fs from water-washing pretreatment and a cement kiln co-processing municipal solid waste incinerator fly ash. *Waste Management*, *76*, 106–116. https://doi.org/10.1016/j.wasman.2018.03.006

Yang, R., Liao, W., & Wu, P. (2012). *Basic characteristics of leachate produced by various washing processes for MSWI ashes in Taiwan*. *104*. https://doi.org/10.1016/j.jenvman.2012.03.008

Yang, Y. L., Wu, Y., Lu, Y. X., Cai, Y., He, Z., Yang, X. L., & Song, H. L. (2021). A comprehensive review of nutrient-energy-water-solute recovery by hybrid osmotic membrane bioreactors. In *Bioresource Technology* (Vol. 320). Elsevier Ltd. https://doi.org/10.1016/j.biortech.2020.124300

Yang, Y., Liew, R. K., Tamothran, A. M., Foong, S. Y., Yek, P. N. Y., Chia, P. W., van Tran, T., Peng, W., & Lam, S. S. (2021). Gasification of refuse-derived fuel from municipal solid waste for energy production: a review. In *Environmental Chemistry Letters* (Vol. 19, Issue 3, pp. 2127–2140). Springer Science and Business Media Deutschland GmbH. https://doi.org/10.1007/s10311-020-01177-5

Yang, Z., Tian, S., Ji, R., Liu, L., Wang, X., & Zhang, Z. (2017). Effect of water-washing on the co-removal of chlorine and heavy metals in air pollution control residue from MSW incineration. *Waste Management*, *68*, 221–231. https://doi.org/10.1016/j.wasman.2017.06.039

Yao, J., Kong, Q., Zhu, H., Long, Y., & Shen, D. (2013). Content and fractionation of Cu, Zn and Cd in size fractionated municipal solid waste incineration bottom ash. *Ecotoxicology and Environmental Safety*, *94*, 131–137. https://doi.org/10.1016/j.ecoenv.2013.05.014

Yu, J., & Klarup, D. O. U. G. (1994). Extraction kinetics of copper, zinc, iron, and manganese from contaminated sediment using disodium ethylenediaminetetraacetate. *Water, Air and Soil Pollution* 75: 205-225.

Zeiss, R., Ixmeier, A., Recker, J., & Kranz, J. (2021). Mobilising information systems scholarship for a circular economy: Review, synthesis, and directions for future research. *Information Systems Journal*, *31*(1), 148–183. https://doi.org/10.1111/isj.12305

Zhang, F. S., & Itoh, H. (2006). Extraction of metals from municipal solid waste incinerator fly ash by hydrothermal process. *Journal of Hazardous Materials*, *136*(3), 663–670. https://doi.org/10.1016/j.jhazmat.2005.12.052

Zhang, W., Huang, H., Tan, F., Wang, H., & Qiu, R. (2010). Influence of EDTA washing on the species and mobility of heavy metals residual in soils. *Journal of Hazardous Materials*, *173*(1–3), 369–376. https://doi.org/10.1016/j.jhazmat.2009.08.087

Zhang, W., & Lo, I. M. (2006). EDTA-Enhanced Washing for Remediation of Pb- and/or Zn-Contaminated Soils. *Journal of Environmental Engineering*, *132*(10), 1282–1288. https://doi.org/10.1061/(asce)0733-9372(2006)132:10(1282)

Zohoori, M., & Ghani, A. (2017). Municipal Solid Waste Management Challenges and Problems for Cities in Low-Income and Developing Countries. In *International Journal of Science and Engineering Applications* (Vol. 6). Online. www.ijsea.com

2. PhD Project Outline

2.1 Research Program Scheme

This thesis originated from a concern about the final storage of waste: Back to the Earth Sites (BES), which explores a desire to achieve a sustainable way of addressing the final storage of municipal waste issues. Sustainable waste management not only meets the need for seeking appropriate solutions in terms of technical and economic levels, to solve emergent contemporary issues but also is in accord with the principles of the Circular Economy (CE) and long-term sustainable strategy. The concept of CE highlights saving non-renewable resources, limiting pollution and decreasing the production of waste and consists of all the stages of the life of a product: extraction of raw materials, production, utilization and discard, responding to the sustainability of waste management.

The work had a particular focus on the last step in the circular approach: the final storage of the discarded residues from the circular approach of CE, concerning two of the most critical aspects of the sustainable landfilling concept: pre-treating the landfilling waste and long-term emissions. Although landfill is regarded as "a hazardous system, obsolete and polluting" ranking the last place of a top-down triangle in waste hierarchy (the ranking from top to bottom are: Prevention, Preparation to Reuse, Recycling and Final disposal), landfill so far continues to play a fundamental role in providing a final sink for non-recyclable residues of CE. Additionally, considering the financial and technical issues, landfill will continue to represent a viable and economic way in waste management. Thus, applying sustainability to the final sink is a feasible and effective approach to managing landfilling waste on both technical and economical levels. The aim of this program was to enhance the quality of landfilling residues and evaluate the final storage quality of long-term emissions, which was accomplished by conducting two main laboratory-scale landfill simulation tests.

The main work in this thesis was briefly schemed in **Figure 2. 1**, and all the research activities were developed in detail following this scheme over a three-year period.

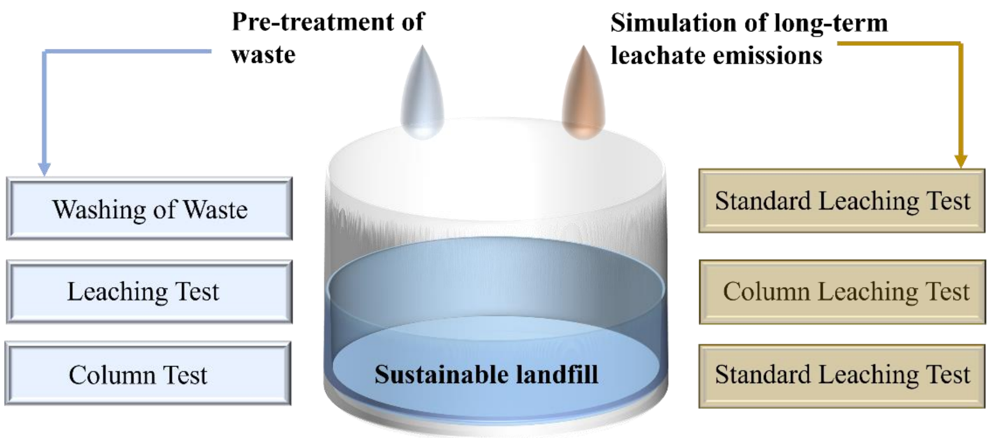

Figure 2. 1. Scheme of the research program.

2.2 Research Activities

On a basis of the scheme indicated in **Figure 2. 1**, the holistic program was completed by a combination of theoretical and practical research parts according to the following phases.

1. Phase one:

In phase one, all the activities mainly focused on this study proceeded according to an initial clue of pursuing sustainable approaches to municipal solid waste management (MSWM). Gaining a better understanding of sustainable waste management by exploring academic background knowledge about globally appropriate and sustainable technical solutions, not only in environmental terms but also in economic and technical terms. Further extensive literature reviews carried out built a logical strategy for the development of the program and profound the background of the first lab-scale job. International conferences provided a profound and brain-storm discussion for the development of this project. A clear and scheduled research plan of high concerns on sustainable landfill was built on a basis of theoretical preparation in this first step. Subsequently, hypotheses about applying sustainability concepts to landfilling waste were materialized by a series of laboratory-scale works.

2. Phase two:

In the second phase, washing of waste was investigated as pre-treatment tool to control long-term emissions and immobilizing contaminants by enhancing the quality of landfilling waste. Laboratory-scale experiments were designed based on theoretical support from data collection and analysis. Washing tests were carried out on three different types of CE residues in order to remove the leachable contaminants before landfilling. The efficiency of washing pre-treatment was evaluated by standard batch leaching tests (UNI EN 12547-2). After pre-treatment, column landfill simulation tests were performed to predict and compare the landfill long-term emissions of both washed and raw residues. Rainfalls in real landfills were simulated by means of daily water irrigation, and leachates generated from columns were analysed to ascertain the accomplishment of the final storage quality (FSQ) limits set by the Italian Regional Guideline. In sustainable landfills, FSQ is an important value to evaluate the sustainability of landfills, and it is defined as a set of values of different parameters to be achieved within the span of one generation, representing an acceptable equilibrium between the landfill and the environment. In this work, washing pre-treatment was proved to remove the leachable contaminants shortening the time required to reach sustainability of landfill.

3. Phase three:

In the third step, research activities focused on the investigation appropriate tools for long-term emissions simulation in view of proper ascertain the accomplishment of FSQ targets within one generation time. In particular, the research activities were carried out with the following objectives:

• Investigate the significance of both column flow-percolations and standard batch leaching tests (UNI EN 12547-2) in view of assessing the landfill sustainability.

• Investigate the effect of different L/S ratios (corresponding to one generation time under different rainfall/landfill conditions) and irrigation frequencies on the estimation of long-term emissions. FSQ targets set by Italian Regional Guideline were used to ascertain the accomplishment of sustainability.

Standard batch leaching tests were performed at the beginning of the experiment on raw waste to evaluate the waste emission potential. The results were compared with landfill admissibility criteria set by Italian legislation.

Column leaching tests were conducted on raw waste samples to simulate different landfilling conditions by adding different cumulative water quantities: 1.5 L/kgTS and 3 L/kgTS, defined as low (L) and high (H) water input. Both water input quantities were irrigated at different frequencies: 10, 20 and 40 times, distributed over a period of 40 days. Leachates gained from column tests were analyzed and final leachate quality was compared to FSQ target fixed by Italian Regional Guideline.

Waste samples after column tests were testes by standard batch leaching test, to estimate the waste emission potential after 30 years landfilling. The results revealed that, although waste accomplish the limits for non-hazardous landfills according to standard batch leaching tests, long term emission not always fulfilled the FSQ limits. Compared with the statistic batch leaching tests, column flow-percolations leaching tests demonstrated to achieve a more realistic simulation of the natural landfill conditions. Irrigation frequencies had a significant impact on the reduction of waste emission potential in column flow-percolations tests, and it could be taken into account to improve the in-situ treatments.

3. Part One: Washing Pre-treatment

3.1 Literature Review

It is an acknowledged fact that creating a zero-waste society in a rapid-growth world is an absolute Utopia. Building a sustainable, green and environmental-friendly society is a generally acceptable and available concept by the world. Although recycling and reusing waste materials have been a more and more encouraging way of reducing waste generation and environmental burden, with respect to economics and technology, landfilling disposal of waste still and will continue to be a predominant disposal way in the future, in particular in some low-income countries (Jha et al., 2011).

Landfill as the final sink for disposal of non-recyclable remains is the step most closely to our environment as well as the active participant of a natural eco-cycling. Although the multibarrier system has been a developed technique to reduce contaminants released from landfills, the multibarrier is never able to isolate pollutants from earth sites. It is more likely that landfill plays a role of temporary storage and intermediate transition. The oversaturation effect of landfills is always an obvious risk. A good example is a discharge of accumulated leachates with high concentrations of heavy metals (HMs) and dissolved organic matters (DOM) cause severe pollution of groundwater and soil (Kjeldsen et al., 2002). In addition, greenhouse gases, ordors and newly-generated contaminants by biological and geochemical reactions impose a heavy burden on the environment. In order to minimize problems associated with landfilling, different pre-treatment techniques should be used before landfilling (Bramryd & Binder, 2007).

Pre-treatment has been used as an effective approach to gain the optimum level of sorting and waste stabilization. It will also open up for the recovery of resources from waste, like bio-energy and nutrients. Besides, pre-treatment techniques can be able to affect both waste reactions after disposal and landfill behaviour in the final storage step. In this sense, pre-treatment technologies can also be taken use of with the ultimate objective of adjusting landfill behavior that is the biological and physicochemical reaction (Norbu et al., 2005). Mechanical shredding is important processing of reducing the volume of residual waste, as well as improving the efficiency of biological treatment. Biological pre-treatment plays an important role in reducing long-term setting and gas-emissions (Bramryd & Binder, 2007; Montejo et al., 2013). Thermal pre-treatment is normally regarded as a significantly effective technique to reduce waste mass and decrease the biological activities of buried waste (Massarutto, 2015). Although each technique has been applied to reach the same aim of reducing environmental burden, a specific technique is normally used to achieve a specific treatment purpose or dispose of a certain spectrum of wastes in a region.

Waste washing treatment has been demonstrated as an efficient pre-treatment technique to remove harmful elements and keep waste stabilised in numerous studies (Cossu et al., 2012; Nestor et al., 2020; Yang et al., 2017). Heavy metals, organic matters, inorganic components and Xenobiotic organic compounds can be reduced or removed during the processing of waste washing. Non-recyclable remains (e.g. automotive shredder residue) pre-treated by a waste washing process before landfilling would produce less emission and show a within a short time frame (Cossu & Lai, 2015). Meanwhile, waste washing remarkably enhances the rate of resource recovery. Co-processing of washed air pollution control residues as an alternative material in cement manufacture not only recycles waste materials but also stabilizes leachable polluted elements (Bogush et al., 2020; Yan et al., 2018). In order to achieve the goal of enhancing washing efficiency, washing techniques have been improved by changing the washing parameters, conditions and methods. Mathematic modelling and simulated condition are also applied to predict and improve washing results (Cossu & Lai, 2012; Velts et al., 2010). However, the characteristics of waste washing quite depend on various factors, including pH, liquid/solid ratio (L/S), washing temperature, washing solvents, as well as waste properties (Beiyuan et al., 2017; X. Chen et al., 2016; Gudka et al., 2016). Although numerous studies have demonstrated that waste washing results can be considerably different due to different influence factors, few review articles on waste washing make a comprehensive review to understand washing behaviours and the stabilization function in different periods.

In order to set up a reliable technique line for accomplishing the first target of the holistic program: washing pre-treatment of waste, in this step, data collections from previous research summarize pollutants generated from washing processing, the factors affecting waste washing, methodologies to study washing from different wastes, potential mechanisms behind washing and the impact of waste washing on the environment. Those data also offer an optional strategy of waste disposal and contribute to future sustainable waste management.

3.1.1 Washing Methods

In the case of people's common sense, washing is an operation to remove unwanted substances to achieve the aim of cleaning. Washing waste with the application of removing hazardous components, degradable organics and high-toxic heavy metals from residues to gain a similar purpose: to clean residues resulting in waste stabilization and emission reduction. Rainwater flushing, groundwater socking as well as leachate recharging are normally able to be categorized as a sort of washing. However, application-oriented artificial washing techniques with proper methods and equipment can achieve fixed washing purposes (such as reducing HMs or waste stabilization) in a targeted manner by adjusting washing conditions (e.g. pH-vale, washing solvents and washing frequency) (X. Chen et al., 2016; Cossu & Lai, 2012, 2013). Besides, systematic washing process with remarkable research significance in understanding washing mechanism and washing behavior, further improving and developing washing techniques.

Waste washing process can be classified into two main methods: batch washing and column washing with respect to waste leaching mechanism (Voegelin et al., 2003). Though both two washing techniques have demonstrated respectively huge advantages, as shown in Table 3.1. 1, the specific application of methods in corresponding studies mainly depends on different research purposes. In comparison to batch washing, continuous column washing is more advantageous as it is more suitable for large-scale study and more thorough removal effect. However, the advantages of time-saving and solvent-saving make batch washing popular in laboratory-scale research (Sazali et al., 2020). Based on the initial mechanism of those two washing methods, extensively modified washing methods have been established to improve the washing efficiency and analyze the applied mechanism. For example, a semi-batch washing column is designed by Velts, et al. to investigate the changes of Ca contents in water flowing and the stagnant layer of ash (Velts et al., 2010). Chimenos et al. developed a multi-step batch washing facility to improve washing efficiency. The results show that the removal efficiency of chlorides and heavy metals in multi-step washing is much higher than that in regular batch washing with optimal conditions (Chimenos & Ferna, 2005). In addition, washing process can also be categorized into acid washing, alkaline washing and water according to different washing solvents (Nestor et al., 2020; Wang et al., 2020; Yang et al., 2017). Furthermore, the temperature during the washing process significantly affects washing efficiency (Bandara et al., 2020; W. S. Chen et al., 2012). Thus, low-temperature and high-temperature washing can be also used as a classification of washing methods.

In short, although washing techniques can be classified in detail with respect to research conditions, washing efficiency is always the dominant parameter to assess the process.

Table 3.1. 1. Comparison of the advantages and disadvantages in soil washing between column and batch washing.

Washing method	waste	Washing solvents	Removal efficiency (%)	Advantage	Disadvantage	Reference
Column	Artificially contaminated Soil	$Na_2S_2O_5$	Pb 61, Zn 94	(a) Higher removal efficiency (b) Easy-operated and continual operation (c) More suitable for simulating real-scale application (d) To achieve different liquid/solid ratios affecting removal efficiency from only one column washing process (e) Higher accuracy of studying results	(a) Time-consuming (b) Full washing process needs a large number of washing solvents (c) No adjustment of pH value during the procedure	(Abumaizar & Smith, 1999; Davis & Singh, 1995; Evangelista & Zownir, 1989; Khan & Abumaizar, 1996; Pantini et al., 2015; Sazali et al., 2020)
	Artificially contaminated Soil	EDTA HCl $CaCl_2$	Pb 85 Pb 100 Pb 78			
	Artificially contaminated Soil	NaOCl	Zn 38-81			
Batch	Artificially contaminated Soil	HCl EDTA CH_3COOH $CaCl_2$	Pb 65-100 60-100	(a) Short washing time (b) Less amount of washing solvent (c) Easy to adjust pH values during washing process (d) Simple facility to operate	(a) The relatively lower removal efficiency (b) Small-scale study (c) Different L/S ratios experimental designs should be conducted in different batches.	
	Contaminated Soil	EDTA	Pb 95			
	Contaminated Soil	EDTA	Pb 85-97			

3.1.2 Factors Affecting Waste Washing Behaviour and Efficiency

1. Liquid to Solid Ratio (L/S)

The effect of liquid and solid (L/S) ratio on washing behavior has been widely regarded as one of the most important factors. The key to the impact of L/S on washing behavior is closely related to the waste leaching behavior during the washing process. This is because washing efficiency is normally assessed by the extraction amount of elements affected by leachability. L/S ratio contributes to leaching and washing behavior of soluble inorganic constituents (e.g. HMs) (Lin et al., 2017), detailed data are graphically shown in **Figure 3.1. 1**. The washing efficiency of HMs, including lead (Pb), zinc (Zn) and copper (Cu), is higher with the increasing L/S ratio. Because a higher L/S ratio can generally promote the dissolution of minerals and accelerates the extraction of HMs (Luo et al., 2019a). Although the higher L/S ratio normally contributes to higher washing efficacy, optimal L/S conditions should be designed with respect to economic and environmental factors (Yang et al., 2017). Because the highest removal efficiency of the specific element usually appears at the optimal L/S ratio level, instead of simply being increasing or declining with ratios. Ju et al. demonstrated that the reduced amount of complex-constituents MSWI increases with L/S firstly during washing process, then drops into a low value. The largest reduction appears at L/S=2.5 instead of other higher L/S (Ju et al., 2020). In addition, the effect of L/S ratio on washing is also element-specific, as shown in **Figure 3.1. 2**. Under the same washing conditions (same L/S ratios and duration time), the removal efficiency of chlorine (Cl) is considerably higher than the efficiency of sulfate (S). Besides, heavy metals are discovered with higher removal rate than that of inorganic salts in the same L/S, although the real concentration of heavy metals is at trace level. The figures also show that the optimal L/S ratio changes as different elements.

$$Removal\ efficiency\ equation\ of\ washing\ residues:\quad \eta_{X.i} = (e_{r.i} - e_{w.i})/e_{r.i} \quad (3.1\text{-}1)$$

Removal efficiency equation of washing residues from reference (Cossu & Lai, 2013), where, $e_{r,i}$ =concentration of the i-substance in the eluate of washing test for raw waste; $e_{w,i}$ =concentration of the i-substance in the eluate of washing test for washed waste.

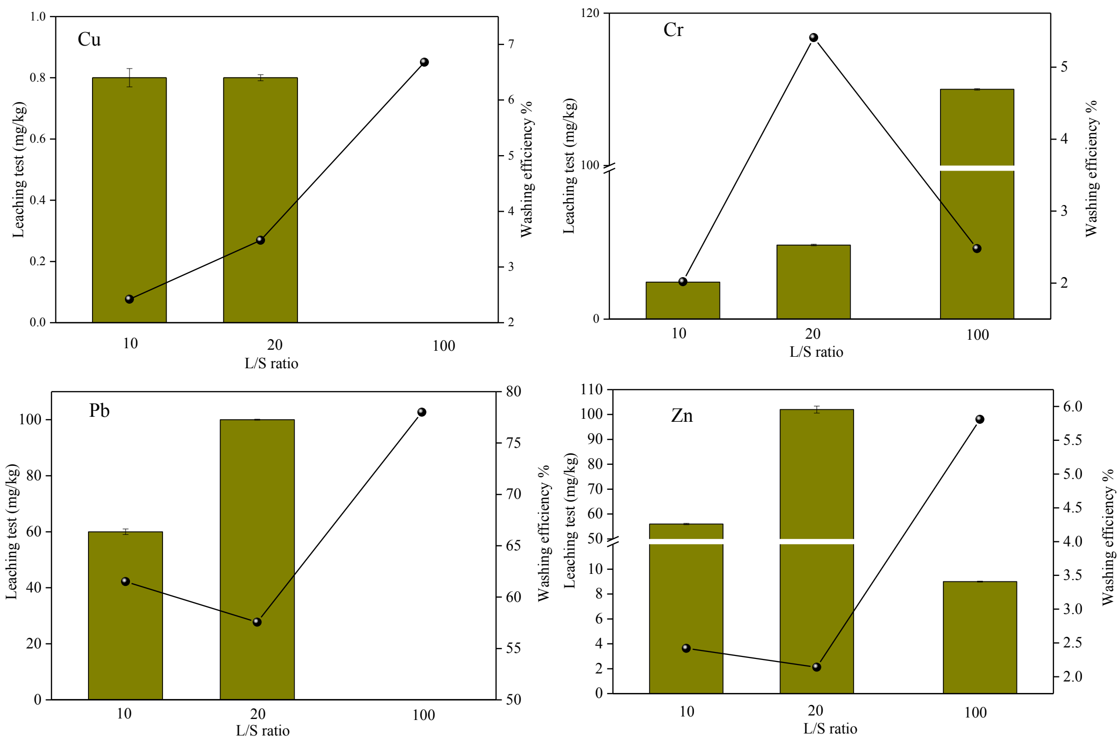

Figure 3.1. 1. Leaching concentrations and washing efficiency of HMs with different L/S ratios. Data was modified from reference (Chiang & Hu, 2010).

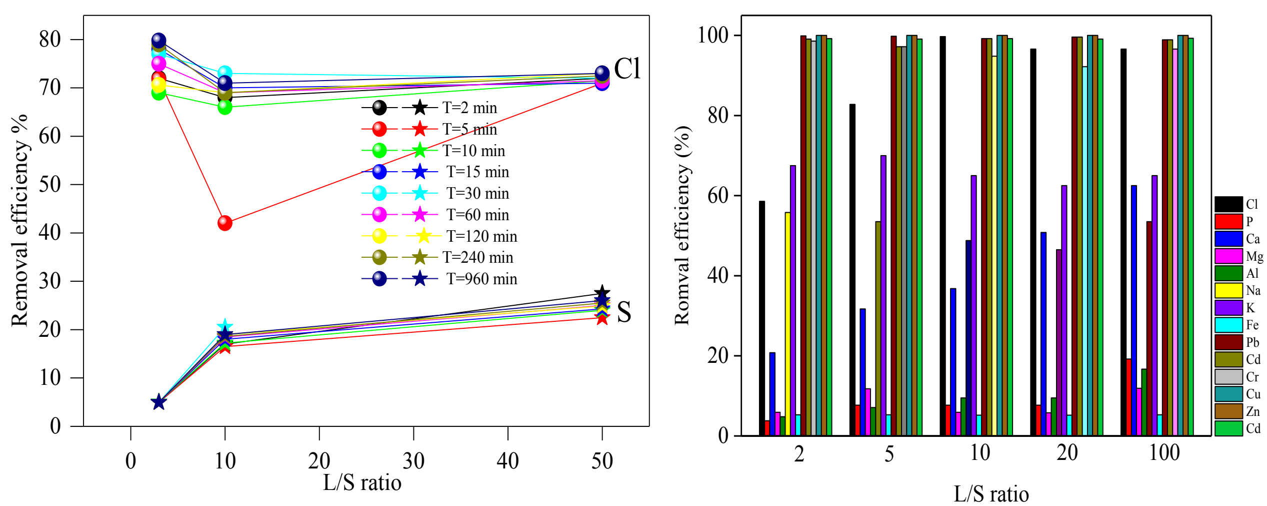

Figure 3.1. 2. Different L/S ratios affect component removal efficiency. Data was modified from reference (Yang et al., 2017).

2. Impact of pH Value

Of the variables considered, the pH of the washing and extraction process proved to be of paramount importance. Due to the heterogeneous characteristics of buried solid residues, residue-removal by washing process is highly pH-dependent. For example, MSW fresh incineration ash is usually at an alkaline (pH=10-12) (Dijkstra et al., 2006), the application of a low-pH solution to removing residues in the MSW incineration ash can be significantly effective (Sun & Yi, 2020). As can be seen in **Figure 3.1. 3**, utilization of different chemical acid to adjust the pH of washing solution can promote washing efficiency, especially in alkaline MSWI ash washing process. The results demonstrate a high dechlorination rate can be gained at a low pH, however, the removal tendency lines go down with increasing pH. The similar tendency can be found in the figures of removing MSWI fly ash amount (Chen et al., 2012). Moreover, washing HMs is also highly pH-dependent as HMs' extraction behavior and solubility are linked with pH-value (Komonweeraket et al., 2015). At the same L/S level, all the heavy metals (Cr, Zn, Cd and Pb shown in **Figure 3.1. 4**) removal efficiencies decrease with pH increases.

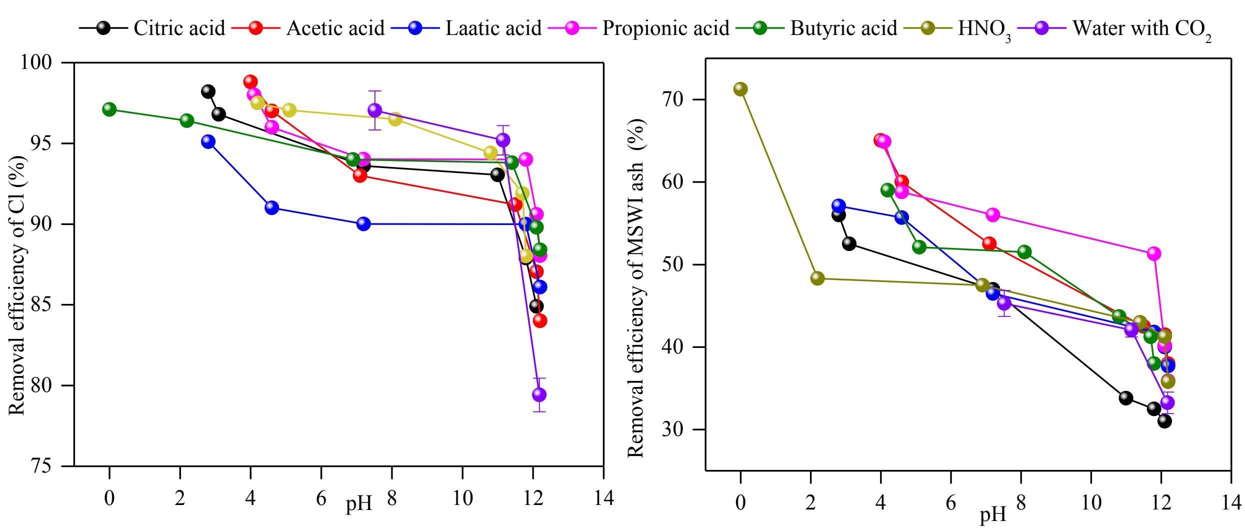

Figure 3.1. 3. Effect of solution pH on removal efficiency of Cl and MSWI fly ash. Data modified from reference (Chen et al., 2012).

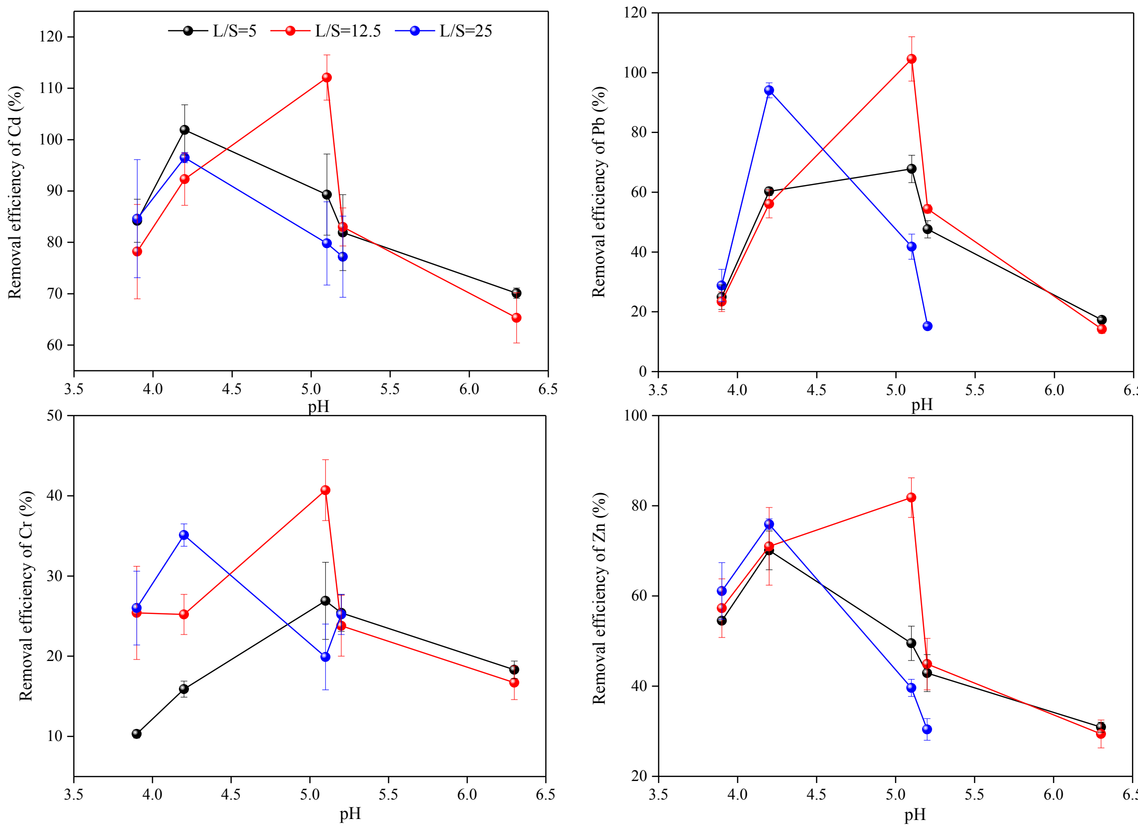

Figure 3.1. 4. Effect of solution pH on removal efficiency of heavy metals. Data modified from reference (Abumaizar & Smith, 1999).

3. The Influence of Washing Temperature

Temperature as an important function has a great impact on substance solubility. Changes in temperature during waste washing process enable washing efficiency and removal rate of residues to reach the target aim (Deng et al., 2013). Chen et al. found that Cl removal rate increased into 93.27 ±1.375% from 79.42 ±1.04% as the temperature up to 90 °C from 25 °C. In addition, the total MWSI weight-loss efficiency also shows an increasing tendency as temperature increases (W. S. Chen et al., 2012). However, the superposition of multiple variables has a fluctuant influence on washing effect that might likely be linear with a single temperature variable. Chen et al. designed an orthogonal washing experiment to investigate the relationship between waste removal efficiency and different variables. The study demonstrates the removal efficiency of Chlorides (e.g. 30 84.55% to 71.33%, 85.24%) is not able to maintain a stably linear tendency when the temperature and stirring speed simultaneously (X. Chen et al., 2016). However, temperature continues to becomes a key function affecting the washing being taken into account in the experimental design.

4. Different Washing Solvents

Washing solvent is one of the most important parts of washing process. Thus, extensive research is dedicated to seeking high-efficiency washing solvents to achieve the target aim of removing waste, as shown in **Table 3.1. 2**. Chemical-enhanced waste washing has been proven to be a promising technology for the remediation of environmental pollution (e.g. contaminated sand and soil) with potential applicability and economic feasibility (W. Zhang & Lo, 2006). For example, chemical chelating agents such as ethylenediaminetetraacetic acid (EDTA) can form stable and soluble complexes with heavy metals, greatly improve their solubility and mobility in aqueous phase, and thus become a good washing solvent to assist heavy-metal removal from soils (F. S. Zhang & Itoh, 2006). However, chemical-enhanced washing solvents have potential risk of polluting the environment after being used as the remover. Thus, environmentally friendly washing solvents are called for being applied to experiments and researches. Deionized water without any original chemicals become a top option. Numerous studies have gained good results by using deionized water during waste washing process (Cossu & Lai, 2013; Yan et al., 2018). Tap water and seawater also have been proved to be good extraction solvent with respect to both economic factors and practicable operation, although DOM in the seawater may be able to affect the leaching behavior of HMs (Lin et al., 2017; R. Yang et al., 2012).

Table 3.1. 2 The effect of different washing solvents

Waste	Untreated Compositions	Washing Solvents	L/S	Removal Efficacy %	Reference
Soil	(mg/kg) Cu 550, As 930	Chelating agents	20	Cu 58~68 %, As 9~21 %	(Tsang & Yip, 2014)
Soil	(mg/kg) Cu 550, As 930	Humic substances	20	Cu 0~6 %, As 2.5~4 %	(Tsang & Yip, 2014)
Soil	(mg/kg) Cu 550, As 930	Inorganic acids	20	Cu 37~50 %, As 4~53 %	(Tsang & Yip, 2014)
Biomass	Cl, Na, K	Water+ HCl + CH_3COONH_4	-	Cl 100 %, Na 92 %, K 62%	(Saddawi et al., 2012)
Marine sediment	TOC 4.85 ± 0.02%, TP 2453 ± 16 mg/kg	HCl	5	TOC~ 33 %, TP~ 47 %	(K. Kim et al., 2020)
Marine sediment	TOC 4.85 ± 0.02%, TP 2453 ± 16 mg/kg	HNO_3	5	TOC~ 20 %, TP~ 50 %	(K. Kim et al., 2020)
Marine sediment	TOC 4.85 ± 0.02%, TP 2453 ± 16 mg/kg	H_2SO_4	5	TOC~ 21 %, TP~ 51 %	(K. Kim et al., 2020)s
Soil	(mg/kg) Cu 89 ±77, Ni 1933± 130, Zn 13565 ±975, Cr 3912 ± 170, Pb 976 ±68	EDTA	20	Cu~9 %, Ni~ 1.7 %, Zn~11%, Cr~10.5 %, Pb ~ 3%	(W. Zhang et al., 2010)
Soil	Cu, Zn, Pb	EDTA+EDDS	-	Cu ~80 %, Zn ~80%, Pb 100 %	(Beiyuan et al., 2018)
MSWI fly ash	Cl	Citric acid	1-20	Cl~ 96%	(X. Wang et al., 2020)
MSWI fly ash	Cl	Acetic acid	1-20	Cl~ 96%	(X. Wang et al., 2020)
MSWI fly ash	Cl	Lactic acid	1-20	Cl~ 91%	(X. Wang et al., 2020)
MSWI fly ash	Cl	Propionic acid	1-20	Cl~ 94%	(X. Wang et al., 2020)
MSWI fly ash	Cl	Butyric acid	1-20	Cl~ 92%	(X. Wang et al., 2020)
ASR	(mg/ml) DOC 83, COD 200, TKN 19.6	Distilled water	3-5	DOC~60%, TKN~ 60%, COD~ 60 %	(Cossu & Lai, 2013)
Sediment	(mg/kg) Cu 970, Zn 2500	Na_2EDTA	40	Cu 55 %, Zn 32 %	(Yu & Klarup, 1994)
MSWI fly ash	(mg/kg) Cd 60, Mn 750, Pb 2100, Zn 5220, Cr 180, Fe 22510	Deionized water	2	Cd 96%, Mn 91%, Pb 73%, Zn 68%, Cr 35%, Fe 30%	(Q. Wang et al., 2009)
Soil	(mg/kg) Cu 1280, Zn 706, Pb 520	EDDS	10	Cu 34%, Zn 23%, Pb 31%	(Beiyuan et al., 2016)
MSW	DOC, COD, TKN, Cr, Zn, Cl^-, F^-	Deionized water	5-10	DOC~85%, COD 85 %, TKN~ 76 %, Cr~ 89 %, Zn 91%, Cl^- 92%, F^- 33%	(Cossu & Lai, 2012a)

5. Particle Size of Waste

The size distribution of waste residues is also an important parameter which is able to affect the washing behavior. On the one hand, previous studies have demonstrated that many toxic contaminants are preferentially concentrated in smaller particles, especially heavy metals (Xia et al., 2017; Yao et al., 2013). Wang et al. and Bogush et al. found that toxic elements (e.g., Cr, Pb and Cd) with a smaller particle size (< 4 mm) in the MSWI bottom ash could cause more contaminates (Bogush et al., 2019; K. S. Wang et al., 2002). On the other hand, particle size contributes to the mobility of polluted elements in landfilling emissions (Mitrano et al., 2017). Meanwhile, fine fractions are also relevant to washing and leaching kinetics as well as washing efficiency due to the bigger surfaces (Bandara et al., 2020; Luo et al., 2017). Cossu and Tai compared the washing efficacies by washing ground ASR (< 4 mm) and not-ground ASR and found that the removal efficiency of Cu is much higher in grounded samples in conditions of L/S 3 (Cossu & Lai, 2013). In addition, small particles are also advocated to enhance the removal efficiency of unwanted elements in biomass washing (Gudka et al., 2016). In addition, the particle size can be the critical parameter affecting hydraulic conductivity, resulting in a difference in flow rate of waste washing and washing effectivity (Arya et al., 1999).

6. Duration Time of Washing Process

To an extent, the duration time of washing solvent contacted with washed waste can be a determining factor affecting the extraction results. During a certain time range, the longer the washing duration time, the higher the removal efficiency of elements (Faravash & Ashtiani, 2007). Because the concentration of soluble wastes is at an unsaturated level in the initial step of washing process. During the unsaturated period, waste can be uptaken by washing solvent (Garrido et al., 2019). However, the duration can not be a decisive factor during a long-term washing process, as shown in **Figure 3.1. 5**. The removal rate both of HMs and inorganic salts would keep at a stable level after reaching a certain rate. Besides, washing solvents can reach a saturated level after uptaking enough. For example, the optimal duration of bubbling CO_2 into the washing solvent to adjust the pH-value removing the most total amount of HMs is 5 min (Z. Yang et al., 2017). Though washing duration time is not always a dominant role in impacting washing behavior, it can improve washing process by connecting other factors as a synergy (e.g. pH-value and L/S).

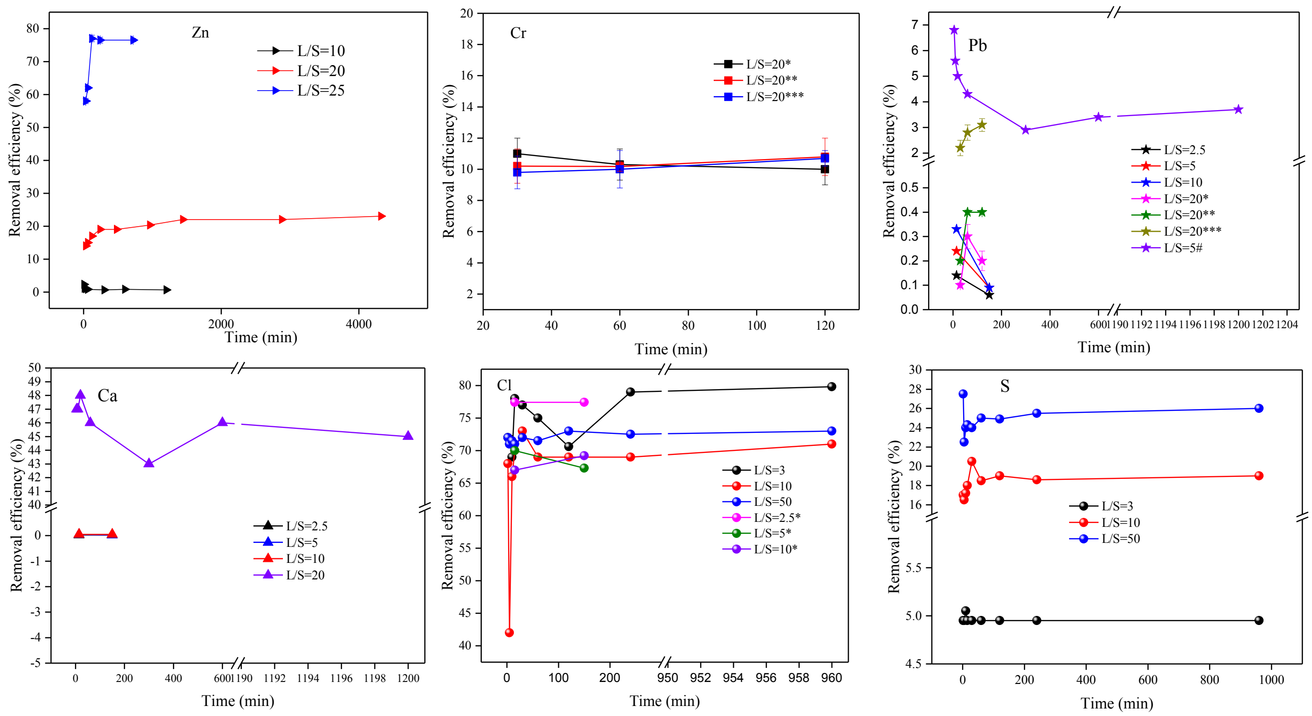

Figure 3.1. 5. The relationship between duration time and removal rate during waste washing process. Figure data modified from references (Cheng et al., 2020; S. Y. Kim et al., 2003; Z. Yang et al., 2017; F. S. Zhang & Itoh, 2006; W. Zhang et al., 2010; W. Zhang & Lo, 2006).

7. Washing Frequency Affecting Washing Process

During the washing process, the pH value of soaking solvents is variable as a consequence of the accumulation and dissolution of soluble salts in the eluates, resulting in a change of the washing efficiency (Sun & Yi, 2020b; K. S. Wang et al., 2002). Thus, the times and cycles of washing can affect the solubility and concentration of soluble elements in washing eluates and lead to the evolution of washing efficiency. Multi-washing steps and cyclic washing procedures have been developed in numerous studies to reach the aim of economical washing and improving efficacy. Chen et al. designed an efficient multi-step washing process to select the optimal washing condition and enhanced the removal efficiency of Cl (up to 99%) in a cyclic washing process (X. Chen et al., 2016). With the increasing number of washing times, washed waste tends to more stable as a consequence of more contaminants have been taken away in the eluates. More washing steps result in lower extraction of polluted elements (e.g., soluble salts and heavy metals) and higher removal efficiency during the washing process (Z. Abbas et al., 2003). Moreover, cyclic washing steps not only increase washing times but also enhance the economical utilization of washing solvents. Organic matters dissolved in eluates can not only change the pH value of leachate, but also enhance the affinity of heavy metals with water facilitating their leachability (Gu et al., 2019; Liaw & Wu, 2013; Luo et al., 2019). The application of multi-step and cyclic washing procedure select optimal washing condition in a condition of economically feasible amount of washing solvents.

8. Other Factors

Apart from the above factors, many other factors, including waste characteristics, quality of waste to be washed, geographic locations, as well as waste pretreated or unpretreated, also have direct and indirect influence on washing behavior and washing efficiency (Beiyuan et al., 2018; Cossu & Lai, 2012, 2013). For example, Cossu and Lai have demonstrated that the washing efficiency of removing DOC and HMs from different municipal solid wastes (under-sieve residues from plastics sorting process, end residues from plastics sorting process, mechanical–biological treated waste, and automotive shredder residues) under the same washing conditions (L/S=5, washing time =6 h) could be totally different (Cossu & Lai, 2012). In addition, different lifestyles and cultures, related decrees and limits to track with environmental problems can also lead to great differences in raw waste constituents, resulting in different further washing pre-treatments (Grossule et al., 2018; Mangialardi, 2004; Yan et al., 2018).

3.1.3 Conclusion

Waste washing treatment applied before landfilling provides a possible strategy to reduce, within a short time frame, leachate environmental impacts in groundwater and soil (Cossu & Lai, 2013). This review summarizes extensive research on waste washing, specifically, factors governing washing, contaminants generated through washing, recycling clean waste materials and treatments to wastewater generated from waste washing. It was proved that L/S ratio, pH-value and washing frequency are the major factors affecting washing efficiency and pollutants removal. Waste particle size, washing duration time and washing solvents, to a certain extent, also become the imperative factors governing washing behaviors. Pollutants generated from the washing process are strongly linked with washing conditions as well as waste properties, which become good research object for investigating waste washing behavior. The complex reactions and element-affinity among the different polluted components (heavy metals, dissolved organic matters, inorganic salts) also play an important role in ultimate washing results. Although waste washing is not the most efficient method to reduce emissions due to its element-specificity, wastewater production and strongly environmental-factor dependence, it still can be able to become a potential pre-treatment. Many pre-treatment methods have been proposed. However, not all approaches are effective and some methods are associated with high energy and high cost, which makes them less economically feasible and attractive (Luo et al., 2019). Besides, whatever pre-treatment applied to waste, the problem of landfilling waste and emissions always exist. Therefore, waste washing becomes a feasible solution that has the quick-respond and effective reduction of emissions in a short time. Furthermore, a combination of washing pre-treatment and specific landfill management techniques contribute towards achieving an equilibrium with the environment, resulting in reducing environmental burden and enhancing resource recovery as much as possible.

References

Abbas, Z., Partovi, A., & Steenari, B. (2003). Release of salts from municipal solid waste combustion residues. *Waste Management. 23*, 291–305.

Abumaizar, R. J., & Smith, E. H. (1999). Heavy metal contaminants removal by soil washing. *Journal of Hazardous Materials*, *70*(1–2), 71–86. https://doi.org/10.1016/S0304-3894(99)00149-1

Arya, L. M., Leij, F. J., Shouse, P. J., & van Genuchten, M. Th. (1999). Relationship between the Hydraulic Conductivity Function and the Particle-Size Distribution. *Soil Science Society of America Journal*, *63*(5), 1063–1070. https://doi.org/10.2136/sssaj1999.6351063x

Bandara, Y. W., Gamage, P., & Gunarathne, D. S. (2020). Hot water washing of rice husk for ash removal: The effect of washing temperature, washing time and particle size. *Renewable Energy*, *153*, 646–652. https://doi.org/10.1016/j.renene.2020.02.038

Beiyuan, J., Tsang, D. C. W., Ok, Y. S., Zhang, W., Yang, X., Baek, K., & Li, X. D. (2016). Integrating EDDS-enhanced washing with low-cost stabilization of metal-contaminated soil from an e-waste recycling site. *Chemosphere*, *159*, 426–432. https://doi.org/10.1016/j.chemosphere.2016.06.030

Beiyuan, J., Tsang, D. C. W., Valix, M., Baek, K., Ok, Y. S., Zhang, W., Bolan, N. S., Rinklebe, J., & Li, X. D. (2018). Combined application of EDDS and EDTA for removal of potentially toxic elements under multiple soil washing schemes. *Chemosphere*, *205*, 178–187. https://doi.org/10.1016/j.chemosphere.2018.04.081

Beiyuan, J., Tsang, D. C. W., Valix, M., Zhang, W., Yang, X., Ok, Y. S., & Li, X. D. (2017). Selective dissolution followed by EDDS washing of an e-waste contaminated soil: Extraction efficiency, fate of residual metals, and impact on soil environment. *Chemosphere*, *166*, 489–496. https://doi.org/10.1016/j.chemosphere.2016.09.110

Bogush, A. A., Stegemann, J. A., & Roy, A. (2019). Changes in composition and lead speciation due to water washing of air pollution control residue from municipal waste incineration. *Journal of Hazardous Materials*, *361*(February 2018), 187–199. https://doi.org/10.1016/j.jhazmat.2018.08.051

Bogush, A. A., Stegemann, J. A., Zhou, Q., Wang, Z., Zhang, B., Zhang, T., Zhang, W., & Wei, J. (2020). Co-processing of raw and washed air pollution control residues from energy-from-waste facilities in the cement kiln. *Journal of Cleaner Production*, *254*, 119924. https://doi.org/10.1016/j.jclepro.2019.119924

Bramryd, T., & Binder, M. (2007). Environmental and Efficiency Evaluation of Different Pre-Stabilization Techniques for Solid Wastes. *Sustainable Land Filling*.

Chen, W. S., Chang, F. C., Shen, Y. H., Tsai, M. S., & Ko, C. H. (2012). Removal of chloride from MSWI fly ash. *Journal of Hazardous Materials*, *237–238*, 116–120. https://doi.org/10.1016/j.jhazmat.2012.08.010

Chen, X., Bi, Y., Zhang, H., & Wang, J. (2016). Chlorides Removal and Control through Water-washing Process on MSWI Fly Ash. *Procedia Environmental Sciences*, *31*, 560–566. https://doi.org/10.1016/j.proenv.2016.02.086

Cheng, S., Lin, Q., Wang, Y., Luo, H., Huang, Z., Fu, H., Chen, H., & Xiao, R. (2020). The removal of Cu, Ni, and Zn in industrial soil by washing with EDTA-organic acids. *Arabian Journal of Chemistry*, *13*(4), 5160–5170. https://doi.org/10.1016/j.arabjc.2020.02.015

Chiang, K. Y., & Hu, Y. H. (2010). Water washing effects on metals emission reduction during municipal solid waste incinerator (MSWI) fly ash melting process. *Waste Management*, *30*(5), 831–838. https://doi.org/10.1016/j.wasman.2009.12.009

Chimenos, J. M., & Ferna, A. I. (2005). *Optimizing the APC residue washing process to minimize the release of chloride and heavy metals*. *25*, 686–693. https://doi.org/10.1016/j.wasman.2004.12.014

Cossu, R., & Lai, T. (2012). Washing of waste prior to landfilling. *Waste Management*, *32*(5), 869–878. https://doi.org/10.1016/j.wasman.2011.12.005

Cossu, R., & Lai, T. (2013). Washing treatment of automotive shredder residue (ASR). *Waste Management*, *33*(8), 1770–1775. https://doi.org/10.1016/j.wasman.2013.04.007

Cossu, R., & Lai, T. (2015). Automotive shredder residue (ASR) management: An overview. *Waste Management*, *45*, 143–151. https://doi.org/10.1016/j.wasman.2015.07.042

Cossu, R., Lai, T., & Pivnenko, K. (2012). Waste washing pre-treatment of municipal and special waste. *Journal of Hazardous Materials*, *207–208*, 65–72. https://doi.org/10.1016/J.JHAZMAT.2011.07.121

Davis, B. A. P., & Singh, I. (1995). Washing of Zinc (II) from contaminated soil column. *ASCE, 121*(2), 174–185.

Deng, L., Zhang, T., & Che, D. (2013). Effect of water washing on fuel properties, pyrolysis and combustion characteristics, and ash fusibility of biomass. *Fuel Processing Technology*, *106*, 712–720. https://doi.org/10.1016/j.fuproc.2012.10.006

Dijkstra, J. J., Van Der Sloot, H. A., & Comans, R. N. J. (2006). The leaching of major and trace elements from MSWI bottom ash as a function of pH and time. *Applied Geochemistry*, *21*(2), 335–351. https://doi.org/10.1016/j.apgeochem.2005.11.003

Evangelista, R. A., & Zownir, A. P. (1989). Lead contamination removal by soil washing. In *Proceedings - A&WMA Annual Meeting* (Vol. 6).

Faravash, R. S., & Ashtiani, F. Z. (2007). *Original article The effect of pH, ethanol volume and acid washing time on the yield of pectin extraction from peach pomace*. *2001*, 1177–1187. https://doi.org/10.1111/j.1365-2621.2006.01324.x

Garrido, Y., Marín, A., Tudela, J. A., Allende, A., & Gil, M. I. (2019). Chlorate uptake during washing is influenced by product type and cut piece size, as well as washing time and wash water content. *Postharvest Biology and Technology*, *151*(February), 45–52. https://doi.org/10.1016/j.postharvbio.2019.01.014

Grossule, V., Morello, L., Cossu, R., & Lavagnolo, M. C. (2018). Bioreactor landfills: Comparison and kinetics of the different systems. *Detritus*, *3*(September), 100–113. https://doi.org/10.31025/2611-4135/2018.13703

Gu, N., Liu, J., Ye, J., Chang, N., & Li, Y.-Y. (2019). Bioenergy, ammonia and humic substances recovery from municipal solid waste leachate: A review and process integration. *Bioresource Technology*, *293*, 122159. https://doi.org/10.1016/J.BIORTECH.2019.122159

Gudka, B., Jones, J. M., Lea-Langton, A. R., Williams, A., & Saddawi, A. (2016). A review of the mitigation of deposition and emission problems during biomass combustion through washing pre-treatment. *Journal of the Energy Institute*, *89*(2), 159–171. https://doi.org/10.1016/j.joei.2015.02.007

Jha, A. K., Singh, S. K., Singh, G. P., & Gupta, P. K. (2011). Sustainable municipal solid waste management in low income group of cities: a review. *Tropical Ecology*, *52*(1), 123–131. www.tropecol.com

Ju, Y., Zhang, K., Yang, T., Deng, D., Qiao, J., Wang, P., You, Y., Du, L., Chen, G., Kołodyńska, D., & Dionysiou, D. D. (2020). The influence of a washing pretreatment containing phosphate anions on single-mode microwave-based detoxification of fly ash from municipal solid waste incinerators. *Chemical Engineering Journal*, *387*(January). https://doi.org/10.1016/j.cej.2020.124053

Khan, L. I., & Abumaizar, R. (1996). Laboratory Investigation of Heavy Metal Removal by Soil Washing. *Journal of the Air and Waste Management Association*, *46*(8), 765–768. https://doi.org/10.1080/10473289.1996.10467512

Kim, K., Yoon, S., Kwon, H. ah, & Choi, Y. (2020). Effects of treatment agents during acid washing and pH neutralization on the fertility of heavy metal-impacted dredged marine sediment as plant-growing soil. *Environmental Pollution*, *267*, 115466. https://doi.org/10.1016/j.envpol.2020.115466

Kim, S. Y., Matsuto, T., & Tanaka, N. (2003). Evaluation of pre-treatment methods for landfill disposal of residues from municipal solid waste incineration. *Waste Management and Research*, *21*(5), 416–423. https://doi.org/10.1177/0734242X0302100504

Kjeldsen, P., Barlaz, M. A., Rooker, A. P., Baun, A., Ledin, A., & Christensen, T. H. (2002). Present and long-term composition of MSW landfill leachate: A review. In *Critical Reviews in Environmental Science and Technology* (Vol. 32, Issue 4, pp. 297–336). https://doi.org/10.1080/10643380290813462

Komonweeraket, K., Cetin, B., Aydilek, A. H., Benson, C. H., & Edil, T. B. (2015). Effects of pH on the leaching mechanisms of elements from fly ash mixed soils. *Fuel*, *140*, 788–802. https://doi.org/10.1016/j.fuel.2014.09.068

Liaw, S. B., & Wu, H. (2013). Leaching characteristics of organic and inorganic matter from biomass by water: Differences between batch and semi-continuous operations. *Industrial and Engineering Chemistry Research*, *52*(11), 4280–4289. https://doi.org/10.1021/ie3031168

Lin, W. Y., Heng, K. S., Nguyen, M. Q., Ho, J. R. I., Mohamed Noh, O. A. Bin, Zhou, X. D., Liu, A., Ren, F., & Wang, J. Y. (2017). Evaluation of the leaching behavior of incineration bottom ash using seawater: A comparison with standard leaching tests. *Waste Management*, *62*, 139–146. https://doi.org/10.1016/j.wasman.2016.12.003

Luo, H., Cheng, Y., He, D., & Yang, E. (2019). Science of the Total environment review of leaching behavior of municipal solid waste incineration. *Science of the Total Environment*, *668*, 90–103. https://doi.org/10.1016/j.scitotenv.2019.03.004

Luo, H., Wu, Y., Zhao, A., Kumar, A., Liu, Y., Cao, B., & Yang, E. H. (2017). Hydrothermally synthesized porous materials from municipal solid waste incineration bottom ash and their interfacial interactions with chloroaromatic compounds. *Journal of Cleaner Production*, *162*, 411–419. https://doi.org/10.1016/j.jclepro.2017.06.082

Mangialardi, T. (2004). Effects of a washing pre-treatment of municipal solid waste incineration fly ash on the hydration behaviour and properties of ash-Portland cement mixtures. *Advances in Cement Research*, *16*(2), 45–54. https://doi.org/10.1680/adcr.2004.16.2.45

Massarutto, A. (2015). Economic aspects of thermal treatment of solid waste in a sustainable WM system. *Waste Management*, *37*, 45–57. https://doi.org/10.1016/j.wasman.2014.08.024

Mitrano, D. M., Mehrabi, K., Dasilva, Y. A. R., & Nowack, B. (2017). Mobility of metallic (nano)particles in leachates from landfills containing waste incineration residues. *Environmental Science: Nano*, *4*(2), 480–492. https://doi.org/10.1039/c6en00565a

Montejo, C., Tonini, D., Márquez, M. del C., & Fruergaard Astrup, T. (2013). Mechanical-biological treatment: Performance and potentials. An LCA of 8 MBT plants including waste characterization. *Journal of Environmental Management*, *128*, 661–673. https://doi.org/10.1016/j.jenvman.2013.05.063

Nestor, P., Djonga, D., Atchana, J., Nankap, A., Mbakop, F. K., Djackba, R., & Tame, A. (2020). Biogas purification by Alkali-based column washing extracted from wood ash and banana stalks. *Science Journal of Energy Engineering. 8*(1), 6–14. https://doi.org/10.11648/j.sjee.20200801.12

Norbu, T., Visvanathan, C., & Basnayake, B. (2005). Pretreatment of municipal solid waste prior to landfilling. *Waste Management*, *25*(10), 997–1003. https://doi.org/10.1016/j.wasman.2005.06.006

Pantini, S., Verginelli, I., & Lombardi, F. (2015). Analysis and modeling of metals release from MBT wastes through batch and up-flow column tests. *Waste Management*, *38*(1), 22–32. https://doi.org/10.1016/j.wasman.2014.12.002

Saddawi, A., Jones, J. M., Williams, A., & Le Coeur, C. (2012). Commodity fuels from biomass through pretreatment and torrefaction: Effects of mineral content on torrefied fuel characteristics and quality. *Energy and Fuels*, *26*(11), 6466–6474. https://doi.org/10.1021/ef2016649

Sazali, N., Harun, Z., & Sazali, N. (2020). A review on batch and column adsorption of various adsorbent towards the removal of heavy metal. *Journal of Advanced Research in Fluid Mechanics and Thermal Sciences*, *67*(2), 66–88.

Sun, X., & Yi, Y. (2020). pH evolution during water washing of incineration bottom ash and its effect on removal of heavy metals. *Waste Management*, *104*, 213–219. https://doi.org/10.1016/j.wasman.2020.01.023

Tsang, D. C. W., & Yip, A. C. K. (2014). Comparing chemical-enhanced washing and waste-based stabilisation approach for soil remediation. *Journal of Soils and Sediments*, *14*(5), 936–947. https://doi.org/10.1007/s11368-013-0831-y

Velts, O., Hautaniemi, M., Kallas, J., Kuosa, M., & Kuusik, R. (2010). Modeling calcium dissolution from oil shale ash: Part 2. Continuous washing of the ash layer. *Fuel Processing Technology*, *91*(5), 491–495. https://doi.org/10.1016/j.fuproc.2009.12.009

Voegelin, A., Barmettler, K., & Kretzschmar, R. (2003). Heavy Metal Release from Contaminated Soils. *Journal of Environmental Quality*, *32*(3), 865–875. https://doi.org/10.2134/jeq2003.8650

Wang, K. S., Sun, C. J., & Yeh, C. C. (2002). The thermo-treatment of MSW incinerator fly ash for use as an aggregate: A study of the characteristics of size-fractioning. *Resources, Conservation and Recycling*, *35*(3), 177–190. https://doi.org/10.1016/S0921-3449(01)00121-5

Wang, Q., Yang, J., Wang, Q., & Wu, T. (2009). Effects of water-washing pretreatment on bioleaching of heavy metals from municipal solid waste incinerator fly ash. *Journal of Hazardous Materials*, *162*(2–3), 812–818. https://doi.org/10.1016/j.jhazmat.2008.05.125

Wang, X., Wang, M., Zou, D., Wu, C., Li, T., Gao, M., Liu, S., Wang, Q., & Shimaoka, T. (2020). Comparative study on inorganic Cl removal of municipal solid waste fly ash using different types and concentrations of organic acids. *Chemosphere*, *261*, 127754. https://doi.org/10.1016/j.chemosphere.2020.127754

Xia, Y., He, P., Shao, L., & Zhang, H. (2017). Metal distribution characteristic of MSWI bottom ash in view of metal recovery. *Journal of Environmental Sciences (China)*, *52*, 178–189. https://doi.org/10.1016/j.jes.2016.04.016

Yan, D., Peng, Z., Yu, L., Sun, Y., Yong, R., & Helge Karstensen, K. (2018). Characterization of heavy metals and PCDD/Fs from water-washing pretreatment and a cement kiln co-processing municipal solid waste incinerator fly ash. *Waste Management*, *76*, 106–116. https://doi.org/10.1016/j.wasman.2018.03.006

Yang, R., Liao, W., & Wu, P. (2012). *Basic characteristics of leachate produced by various washing processes for MSWI ashes in Taiwan*. *104*. https://doi.org/10.1016/j.jenvman.2012.03.008

Yang, Z., Tian, S., Ji, R., Liu, L., Wang, X., & Zhang, Z. (2017). Effect of water-washing on the co-removal of chlorine and heavy metals in air pollution control residue from MSW incineration. *Waste Management*, *68*, 221–231. https://doi.org/10.1016/j.wasman.2017.06.039

Yao, J., Kong, Q., Zhu, H., Long, Y., & Shen, D. (2013). Content and fractionation of Cu, Zn and Cd in size fractionated municipal solid waste incineration bottom ash. *Ecotoxicology and Environmental Safety*, *94*, 131–137. https://doi.org/10.1016/j.ecoenv.2013.05.014

Yu, J., & Klarup, D. O. U. G. (1994). *Extraction kinetics of copper, zinc, iron, and manganese from contaminated sediment using disodium ethylenediaminetetraacetate*. 205–225.

Zhang, F. S., & Itoh, H. (2006). Extraction of metals from municipal solid waste incinerator fly ash by hydrothermal process. *Journal of Hazardous Materials*, *136*(3), 663–670. https://doi.org/10.1016/j.jhazmat.2005.12.052

Zhang, W., Huang, H., Tan, F., Wang, H., & Qiu, R. (2010). Influence of EDTA washing on the species and mobility of heavy metals residual in soils. *Journal of Hazardous Materials*, *173*(1–3), 369–376. https://doi.org/10.1016/j.jhazmat.2009.08.087

Zhang, W., & Lo, I. M. (2006). EDTA-Enhanced Washing for Remediation of Pb- and/or Zn-Contaminated Soils. *Journal of Environmental Engineering*, *132*(10), 1282–1288. https://doi.org/10.1061/(asce)0733-9372(2006)132:10(1282)

3.2 Lab-scale Washing Test

Based on: Q Luo., V Grossule., MC Lavagnolo. Washing of residues from the circular economy prior to sustainable landfill: effects on long-term impacts.

Published in: *Waste Management and Research*, Accepted: 19 August 2022

Abstract

Sustainable landfill continues to play a fundamental role in closing the loop of residual materials of the Circular Economy. The sustainable landfill relies on both pre-treatments and in situ treatments to stabilise the residual waste and immobilise the contaminants, achieving the Final Storage Quality (FSQ) within one generation (typically 30 years). The aim of the study was to investigate the efficiency of the waste washing pre-treatment in reducing the waste leaching fraction prior to landfilling, and in decreasing the time needed to reach the FSQ. A laboratory scale washing test was performed on three different kinds of residues from municipal solid waste treatment, usually landfilled: residues sieved from separately collected bio-waste (RB); residues sieved from compost (RC); and residues sieved from mixed waste treatment-plastic line (RP). Column landfill simulation tests were performed to predict and compare the landfill long term emissions of both washed and raw residues. The results revealed that the washing pre-treatment significantly reduced the leachable fraction of contaminants, decreasing the time needed to reach the Chemical Oxygen Demand (COD) and ammonia FSQ limits. However, RP residue was the only one respecting both FSQ limits within 30 years.

Keywords: Waste washing, leachate, circular economy residues, Final Storage Quality (FSQ)

3.2.1. Introduction

The current approach of the European Union to the waste management consists in the principle of the circular economy, which is aimed at saving resources and minimising waste generation through the application of a series of actions, defined by the European Commission Communication (2015) as the “waste hierarchy”. Although waste prevention should be the most preferrable action, the focus of the European legislation in the last years has been the recycling of the waste, with increasingly ambitious recycling targets being set for the next years (EU Directives 851, 852/2018). Consequently, recycling is often perceived as a fundamental tool to reach the Zero Waste target whereas, landfilling and other disposal activities are perceived as obsolete and hazardous. However, recycling has several limits, among which, the production of unavoidable residues in which contaminants have accumulated (Pivnenko et al., 2016) and which need a secure final sink in order to avoid diffuse pollution (i.a. Muola et al., 2021). Sustainable landfill plays the fundamental role of closing the loop of the materials which cannot be further reintroduced in the Circular Economy, and they are given Back to the Earth (Cossu, 2016) in a stable, not contaminating form. The sustainable landfill relies not only on physical barriers, which have a limited lifespan (Sun et al., 2019), but also on waste pre-treatments (e.g. mechanical-biological, thermal treatment) and in-situ treatments, (e.g. semi aerobic-landfill, forced aeration, flushing) (Grossule and Stegmann, 2020), properly selected according to the specific context (Lavagnolo and Grossule, 2018)

Both pre-treatments and in-situ treatments should be purposely designed to stabilise the residual waste and immobilise the contaminants, with the aim of achieving, within the time frame of one-generation (30 years), a Final Storage Quality (FSQ) of the landfill in equilibrium with the environment (Cossu et al., 2020a). When the FSQ is achieved, the emission potential of the landfilled waste and the uncontrolled emissions will not necessarily correspond to zero, but they should be compatible with the surrounding environment, without any non-acceptable impacts, whilst exploiting the environmental self-depuration ability. The FSQ is typically defined by limit values for landfill emissions in terms of leachate quality and load, gas production and waste stability (Grossule, 2020).

Among the pre-treatment options, the washing of waste is a physical/chemical process, suitable for the removal of leachable contaminants and particularly indicated for pre-treatment of mainly inorganic or well stabilised waste.

Being leachate the main cause for long-term environmental impact of landfills, the washing of residues prior to landfilling may represent an interesting option to remove the leachable fractions of the contaminants, reducing the leachable emission potential and improving the quality of landfill emissions.

Extensive studies demonstrated the efficiency of waste washing treatment in reducing the leachable fraction of chlorides, metals, Chemical Oxygen Demand (COD) and nitrogen (Total Khjeldal Nitrogen – TKN) from different kind of wastes (e.g. incineration ashes, automotive shredded residues, etc.; Sun and Yi, 2021; Cossu and Lai, 2013). Furthermore, when co-processing with other techniques (e.g. concrete manufacturing and cement solidification) (Bogush et al., 2020; Keulen et al., 2016; Yan et al., 2018), the waste washing treatment can significantly enhance the quality of waste and the possibility of recycling. Some studies highlighted the different factors affecting the washing effect, including liquid/solid (L/S) ratio, washing agents, pH value and washing frequency (Chen et al., 2016; Jiang et al., 2009; Kim et al., 2020; Wang et al., 2020).

Incineration ashes was the main waste typology tested for waste washing studies (i.a. Sun and Yi, 2021; Wei et al., 2021). Only few studies investigated the washing of plastic waste (Chaudhuri et al., 2001 and Cossu et al., 2012), while mechanical-biological treated waste and automotive shredded residues were tested for washing treatment only by Cossu et al., 2012; Cossu and Lai, 2013; Cossu et al., 2012. Besides the already studied waste types, many other different kinds of residues from Circular Economy continue to be landfilled, in many cases without proper waste pre-treatments in view of sustainable landfill.

The present study aimed to further evaluate the effect of waste washing prior to landfilling on reducing the emission potential of different selected Circular Economy residues and on long-term impacts.

A laboratory scale washing test was performed on three different kinds of residues from municipal solid waste treatment, usually landfilled: residues (impurities such as plastic bags) sieved from separately collected bio-waste (RB); residues (inert or well stabilised impurities) sieved from compost (RC); and residues sieved from mixed waste treatment-plastic line (RP). Washing treatment, was set using the suggested parameters from literature (L/S=5 l/kg, t=6 h; Cossu and Lai, 2012), as these operating conditions proved to achieve the best trade-off between the washing efficiency, duration and water consumption of the test in view of the upscaling. Column landfill simulation tests were performed to predict and compare the landfill long term emissions of both washed and raw residues, in view of accomplishing the FSQ limits set by Italian Regional Guideline (Cossu et al., 2020b). Accordingly, being all selected fraction affected by organics contamination, suitability of washing pre-treatment was evaluated.

3.2.2. Materials and Methods

3.2.2.1 Research Program Scheme

The scheme of the research program is shown in **Figure 3.2. 1**. In the first step, a laboratory-scale washing was applied to three different types of waste residues. The efficiency of washing pre-treatment was evaluated by standard batch leaching tests (UNI EN 12547-2). In the second step, column landfill simulation tests were performed to predict and compare the landfill long term emissions of both washed and raw residues. Collected leachates were analysed to ascertain the accomplishment of the FSQ limits set by Italian Regional Guideline (Cossu et al., 2020b).

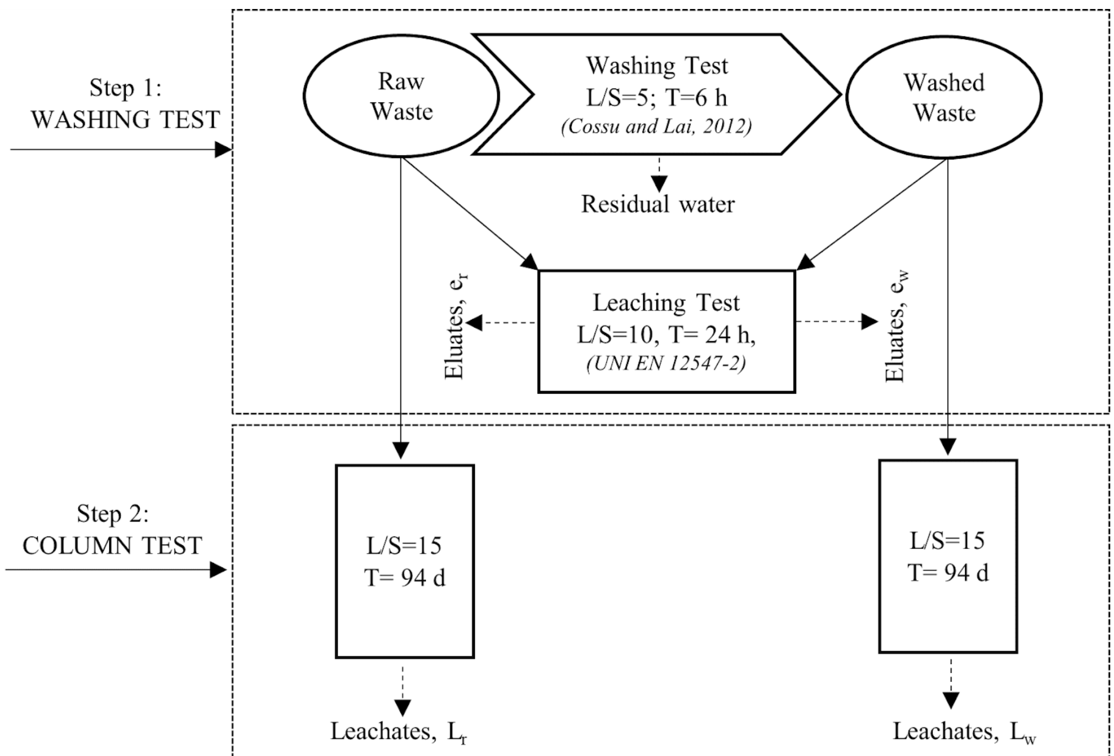

Figure 3.2. 1. Scheme of the research programme (L/S = Liquid/Solid ratio, hence the ratio between the amount of washed residue and amount of washing water; T=testing time; e_r, e_w=eluate of batch leaching test of raw and washed waste, respectively; L_r = leachate of column test of raw waste; L_w = leachate of column test of washed waste).

3.2.2.2 Residues Characterization

Three types of residues (**Figure 3.2. 2**) have been used in this study:

- Oversieved residues from separately collected bio-waste (RB)
- Oversieved residues from compost (RC)
- Oversieved residues from mixed waste treatment-plastic line (RP)

Figure 3.2. 2. The tested raw residues (RB: oversieved residues from separately collected bio-waste; RC: oversieved organic waste after composting; RP: oversieved plastic materials from mixed waste treatment).

Residues samples were collected in municipal solid waste treatment plants located in North of Italy. Originating process of the residues and the European Waste Code (EWC) are reported in **Figure 3.2. 5**.

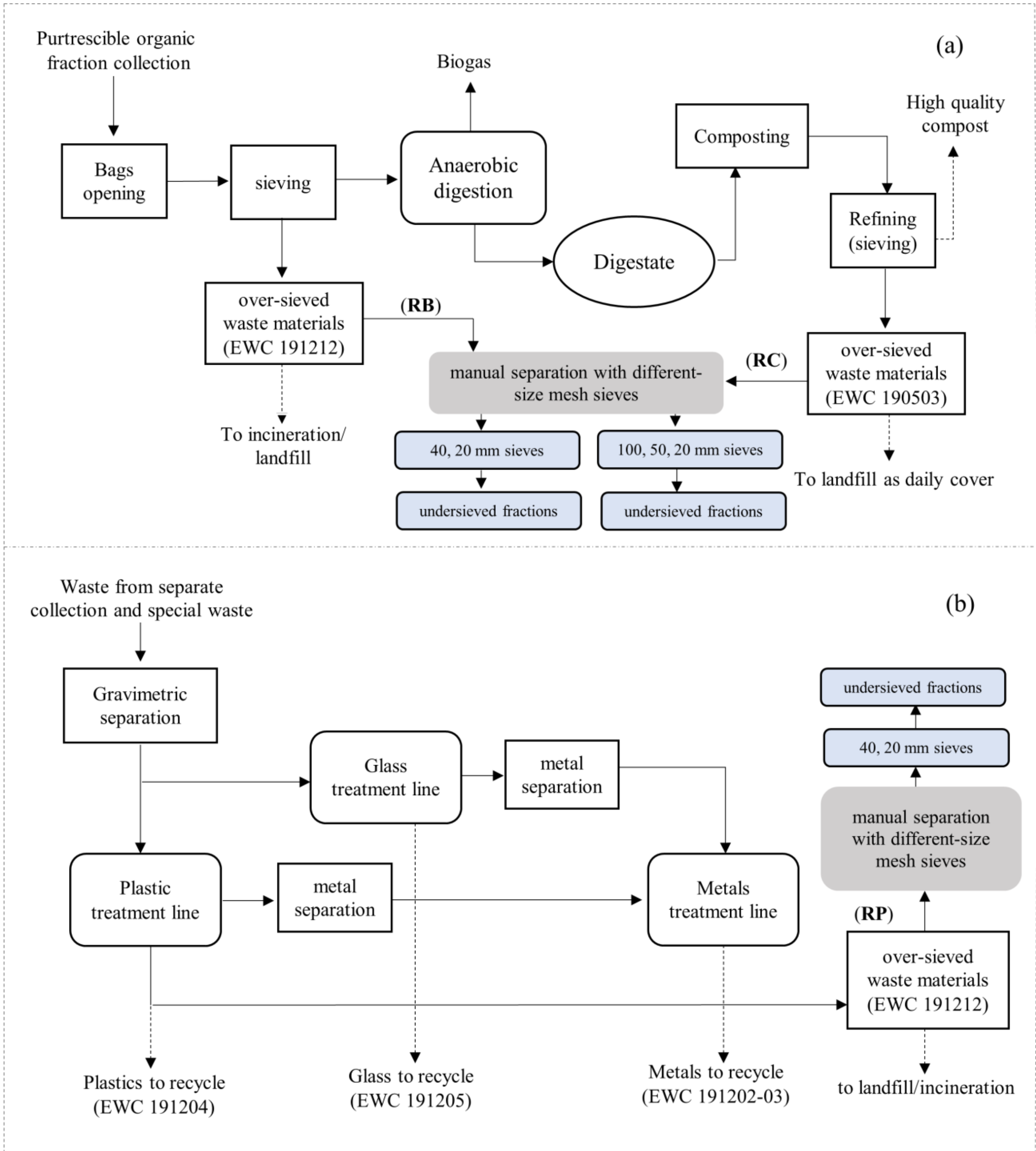

Figure 3.2. 3. Originating process of residues. The European Waste Code (EWC) is reported in bracket. (RB: oversieved residues from separately collected bio-waste; RC: oversieved organic waste after composting; RP: oversieved plastic materials from mixed waste treatment).

100 kg sample of each residue was classified by means of a trade analysis (UNI 10802:2013), as shown in **Figure 3.2. 4**. Prior to washing test, all samples were shredded to a size <20 mm.

Total Solids (TS), Volatile Solids (VS), TKN, Total Organic Carbon (TOC), seven days Respirometric Index (RI_7) and metals (Cd, Cr, Cu, Fe, Mn, Ni, Pb, Zn) were analysed on each residue.

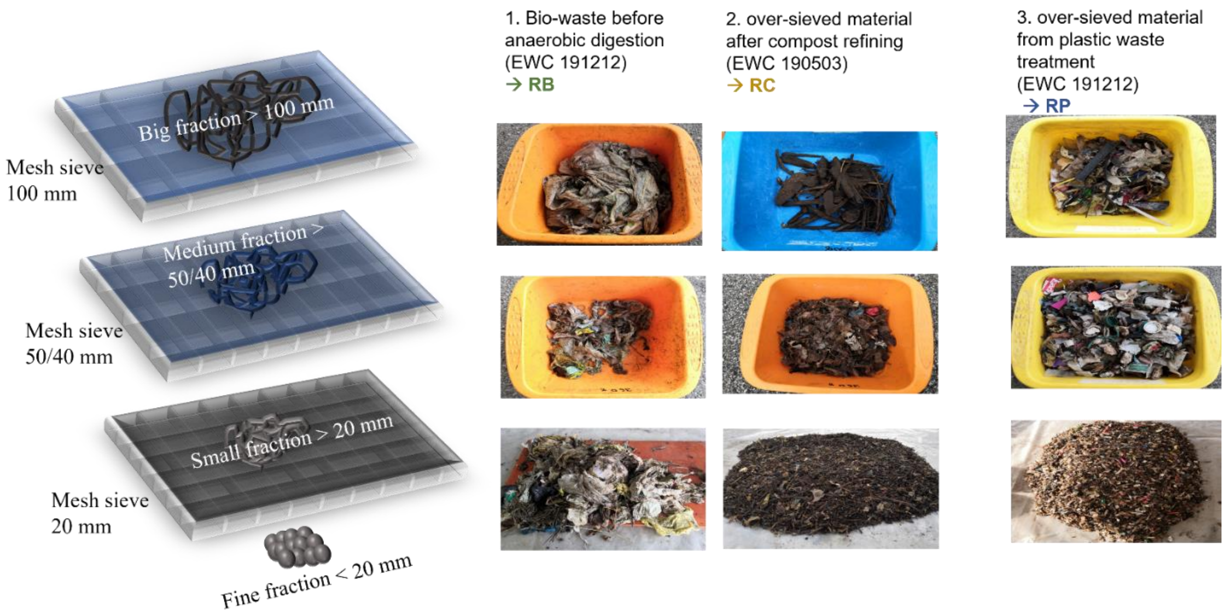

Figure 3.2. 4. Pictures of trade analysis.

3.2.3 Washing Pre-treatment Tests

Washing pre-treatment tests were conducted following the suggestions of previous studies (Cossu & Lai, 2012, 2013) considering a liquid to solid ratio equal to 5 L/kg_{TS} (L/S=5) and a test duration of 6 hours. Waste washing was performed by placing 1.5 kg_{TS} of sample and 7.5 L of deionised water in a HDPE container, then continuously mixed at a speed of 22 rpm. Residual waters from washing tests were analysed for Dissolved Organic Carbon (DOC) during the washing at interval times of 0.5, 2, 4, 6 hours; while COD, Biological Oxygen Demand after 5 days (BOD_5), TKN, ammonia nitrogen (NH_3-N), pH at the end of the test (after 6 hours).

Standard batch leaching tests (UNI EN 12547-2: L/S=10, duration 24 h) were performed both on the raw and washed residues. Leachable fraction of contaminants was analysed in eluates (*e*) in terms of DOC, COD, BOD_5, NH_3-N, TKN, pH, chlorides and metals. Analysis were all performed in triplicate, results are given as average value.

The maximum potential leaching rates from residues were calculated, in terms of TKN and DOC heavy metals (Cd, Cr, Cu, Fe, Mn, Ni, Pb, Zn), by comparing the contaminants concentrations in eluates of raw residues (e_r) and those in the solid raw residues (x), using the following formula:

$$p_i = (e_{ri} \cdot L)/(x_i \cdot S) \qquad (3.2\text{-}1)$$

where i=i-contaminant, L= quantity of eluate (L), S= mass of tested waste (kg_{TS}).

The leaching rates achieved with washing (w_i) were calculated by substituting in Eq. 3.2-1 the contaminants concentrations in eluates (e_{ri}) with those detected in water from washing (w_{ri}):

$$w_i = (w_{ri} \cdot L)/(x_i \cdot S) \qquad (3.2\text{-}2)$$

p_i and w_i were compared to evaluate the effectiveness of washing in removing leachable fraction of contaminats.

Washing efficiencies, defined as the percentage reduction of the maximum potential leaching rates obtained by the washing treatment, were calculated according to the following equation (Cossu & Lai, 2012b):

$$\eta_{x,i} = \frac{e_{r,i} - e_{w,i}}{e_{r,i}} \qquad (3.2\text{-}3)$$

where $e_{r,i}$ and $e_{w,i}$ represent the concentration of the i-substance in the eluate of the batch leaching test of the raw and washed waste residues, respectively.

3.2.4 Column Landfill Simulation Tests

Long term landfill leaching emissions were simulated by column tests using polymethylmethacrylate columns (internal diameter 10 cm, height 104 cm, **Figure 3.2. 5**)

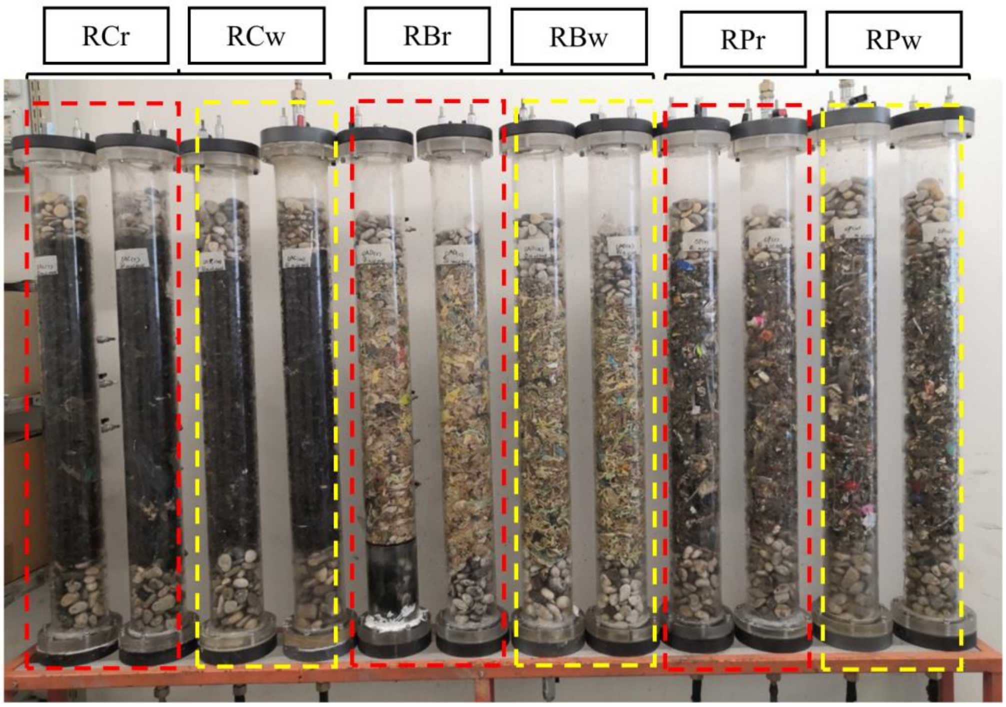

Figure 3.2. 5. Column landfill simulation test apparatus (RB: organic waste before anaerobic digestion; RC: over-sieved organic waste after composting; RP: over-sieved plastic materials from mixed waste treatment; r: raw residues; w: washed residues)

Both raw and washed residues were tested in duplicate. Each column was filled with 1.25 kg_{TS} residues samples. 200 mL of deionized water, simulating rainwater, was added daily to each column for 94 days. Leachate samples were collected and analysed at different L/S ratios (0.5, 1, 1.5, 2, 5, 10, 15; calculated as produced leachate over mass of dry residues in the column) for the following parameters: TOC, COD, NH_3-N, Chlorides.

Ammonia nitrogen and recalcitrant organic matter are the main responsible for the longest-lasting environmental impacts of a landfill (Kjeldsen et al., 2002; Luo et al., 2020). Long-term emissions were, thus, predicted in terms of COD and ammonia, considering a hypothetical landfill and performing a mass balance of water influx and leachate produced, as described by Cossu et al. (2012). Accordingly, the final L/S=15 L/kg_{TS} achieved in the column tests corresponds to 280 years, which is the time required by the hypothetical landfill to achieve the same L/S. Landfill sustainability reference time of 30 years corresponds to a L/S=2. The time trend of COD and ammonia concentrations was determined by fitting the concentrations measured in column tests by using the following first order kinetic formula:

$$C_t = C_0 * e^{-k(t-t_0)} \qquad (3.2\ -4)$$

where, C_t is the concentration of substance at time t (mg/L), C_0 is the concentration of substance at the start time of the model simulation at time t_0 (mg/L); k is the first order kinetic constant (y^{-1}) (Cossu & Lai, 2012b); t0 is time taken at the start of model simulation.

The results were discussed, considering most relevant parameters (COD and ammonia), in view of accomplishing the FSQ limits set by Italian Regional Guideline (Cossu et al., 2020b) considering the landfill sustainability reference time of 30 years.

3.2.5 Analytical Methods

The parameters analysed on liquid samples were pH, TOC, DOC, COD, BOD_5, TKN, N-NH_3,metals (Cd, Cr, Cu, Fe, Mn, Ni, Pb, Zn) and Chlorides. The parameters analysed on solid samples were: RI_7, TS, VS, TKN, TOC. TOC and DOC were determined using a TOC–VCSN Shimadzu Analyzer, in case of DOC after filtration at 0.45µm. COD was measured using a photometric method and BOD was determined using a dissolved O_2 probe. NH_3-N and TKN were measured by means of a distillation-titration procedure. Metals were measured using an ICP-OES analyser. The RI_4 was performed using Sapromat E.

3.2.6 Results and Discussion

3.2.5.1 Raw Waste Characteristics

Figure 3.2. 6 illustrates the trade composition of each tested residue.

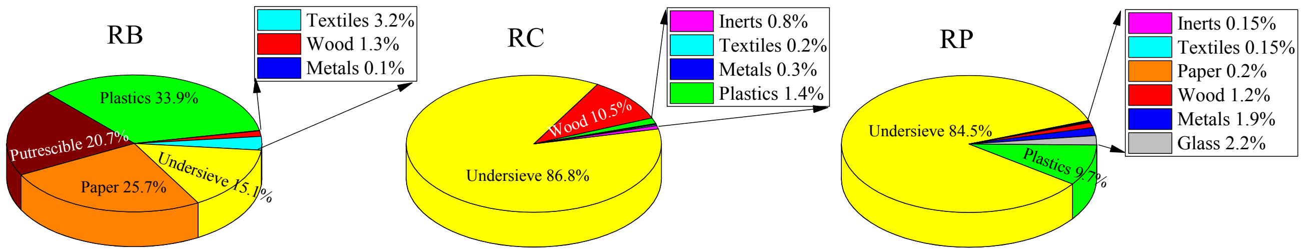

Figure 3.2. 6. Trade composition of raw residues (% on weight). (RB: oversieved residues from separately collected biowaste; RC: oversieved organic waste after composting; RP: oversieved plastic materials from mixed waste treatment).

RB is mainly characterized by coarse fractions, including plastics, paper as well as putrescible in decreasing percentages. Conversely, undersieve (fine fraction, <20 mm) represents the greatest fraction in both RC and RP residues (more than 84%), mainly represented by stabilized biowaste in RC and inert material in RP. The composition reflects the origin of residues and affects their biological stability, and carbon and nitrogen content.

Table 3.2. 1 illustrates the chemical-physical characterization of the raw residues in terms of biological stability (RI_7) and contaminants concentrations (x_i) in the dry mass. The maximum potential leaching rates (p_i) are also provided for Carbon (DOC), Nitrogen (TKN) and metals (Cd, Cr, Cu, Fe, Mn, Ni, Pb, Zn) calculated using the Eq. 3.2-1. The higher RI_7, VS, TOC and TKN values were detected, as expected, on RB residue, characterized by a relevant not stabilized putrescible waste fraction, while the lowest values were detected on RP residue. Conversely, the highest concentrations of metals were detected in RP for Fe and Mn.

When considering the maximum potential leachable rates, RB residues are characterised by the highest amounts of leachable Carbon (9%), due to the presence of the high putrescible fraction, that could be hydrolysed and transferred to the liquid matrix effectively.

Conversely, the highest amount of leachable TKN was detected in RP, which could be justified by a prevalent form of ammonia nitrogen respect to organic nitrogen compared to the other residues. When considering heavy metals, the leachable fractions didn't significantly differ among the different residues.

Table 3.2. 1. Characterization of the raw residues (RW) in terms of contaminants concentrations in the residues dry matter (x_i, mg/kg_{TS}) and maximum potential leaching rates calculated for the i-contaminant (p_i, %). p_i valueswere calculated for Carbon (DOC), Nitrogen (TKN) and metals (Cd, Cr, Cu, Fe, Mn, Ni, Pb, Zn) using the following formula (Eq. 3.2-1): $pi = (e_{ri} \bullet L)/(x_i \bullet S)$; where i = i-contaminant; e_r = contaminant concentration in eluates of raw residues (mg/L); w_r= contaminant concentration in water from washing; L= quantity of eluate/water (L); S =mass of tested/washed waste (kg_{TS}). (RB: oversieved residues from separately collected biowaste; RC: over-sieved organic waste after composting; RP: over-sieved plastic materials from mixed waste treatment).

	x_i Units	RB		RC		RP	
		x_i	p_i	x_i	p_i	x_i	p_i
RI_7	g_{O2}/kg_{TS}	164.5		24.8		7.3	
TS	g_{TS}/kg_{RW}	380		670		770	
VS	g_{VS}/kg_{TS}	860		500		500	
DOC	g_C/kg_{TS}	600	9%	338	3%	290	1.2%
TKN	g_N/kg_{TS}	19.1	23%	16.6	25%	2.2	32%
Cd	mg/kg_{TS}	1.8	5.7%	1.35	7.4%	8.8	1.1%
Cr	mg/kg_{TS}	7.7	1.3%	27.8	0.4%	19.2	0.5%
Cu	mg/kg_{TS}	37.4	0.3%	55.1	0.2%	120	0.1%
Fe	mg/kg_{TS}	2107	-	6125	-	984	-
Mn	mg/kg_{TS}	377	-	545	-	284	-
Ni	mg/kg_{TS}	9.9	1.0%	10.3	1.0%	23.7	0.4%
Pb	mg/kg_{TS}	11.3	0.9%	24.3	0.4%	143	0.1%
Zn	mg/kg_{TS}	219.5	0.1%	387	-	931	-

3.2.6.1 Washing pre-treatment tests: characterisation of water from washing tests and washing efficiency

Residual waters were sampled and analysed for DOC during the washing treatment with the aim of evaluate whether the selected washing procedure achieved the best trade-off between the washing efficiency, duration, and water consumption. DOC concentrations in the washing waters collected over time, demonstrated that washing duration does not affect DOC release (**Table 3.2. 2**), and 0.5 h pre-treatment would be enough, with a significant consequence, in case, on the design of the reactor volume and on the energy needs.

Table 3.2. 2. Dissolved organic carbon (DOC) concentrations analysed in washing water at different sampling times during the washing process. (RB: oversieved residues from separately collected bio-waste; RC: oversieved organic waste after composting; RP: oversieved plastic materials from mixed waste treatment).

Time (h)	**RB (mg/)**	**RC (mg/)**	**RP (mg/)**
0.5	10900	1475	525
2	10500	1510	576
4	11050	1490	588
6	10900	2020	630

Residual waters resulting from the washing tests (six hours duration, L/S = 5 L/kg_{TS}) were analysed in terms of DOC, COD, BOD_5, TKN, NH_3-N, Chlorides and pH. The results, expressed in terms of concentrations are reported in **Table 3.2. 3**.

The highest contaminant concentrations were detected in water from RB washing, confirming the results obtained for the maximum potential leachable rates (p_i). In general, all residual waters from washing treatment are characterised by a high BOD/COD ratio, suggesting that a biological treatment would be required for their treatment.

Washing leaching rates (w_i) were calculated according to the Eq. 3.2-2 for most relevant parameters (DOC, Ammonia) and reported in brackets in Table 2. When comparing the maximum potential leaching rates (p_i, **Table 3.2. 1**) with the washing leaching rates (w_i, **Table 3.2. 3**), the results demonstrated how the washing test allowed the removal of most of the leachable fraction.

Table 3.2. 3. Characterisation of the residual water resulting from the washing tests in terms of DOC, COD, BOD_5, TKN, NH_3-N, Chlorides concentrations (which represented the w_{ri} term in Eq. 3.2-2) and pH. The washing leaching rates (w_i), calculated according to the Eq. 3.3- 2, are reported in brackets for most relevant parameters (DOC, Ammonia). (RB: oversieved residues from separately collected biowaste; RC: over-sieved organic waste after composting; RP: over-sieved plastic materials from mixed waste treatment)

	Units	RB	RC	RP
DOC	mg_C/L	10900 (9%)	2020 (3%)	630 (1.1%)
COD	mg_{O2}/L	42800	10240	2470
BOD_5	mg_{O2}/L	19588	799	657
TKN	mg_N/L	802 (21%)	770 (23.2%)	113 (25.7%)
NH_3-N	mg/L	209	239	39
Chlorides	mg/L	1214	797	233
pH	-	5.2	7.2	7

Washing efficiencies ($\eta_{x,i}$, Eq. 3.3-3), defined as the percentage reduction of the maximum potential leaching rates obtained by the washing treatment, are reported in **Table 3.2. 4**, jointly with the contaminants concentrations in eluates from batch tests.

As expected, highest washing efficiencies have been detected for inorganic contaminants such as chlorides (between 75 and 94%) and ammonia nitrogen (between 63 and 85%), confirming the results of p_i. Conversely, organic substances washing efficiencies were lower, particularly in case of RP residues (17% and 16% for DOC and BOD respectively). This result is in line with previous studies, where comparable washing efficiencies were achieved (Cossu et al., 2012).

Table 3.2. 4. Batch leaching test results: i-contaminant concentration in eluates of both raw ($e_{r,i}$) and washed ($e_{w,i}$) residues; washing efficiency of the i-contaminant ($\eta_{x,i}$), defined as the percentage reduction of the maximum potential leaching rates obtained by the washing treatment and calculated according to the following equation: $\eta_{x,i} = \frac{e_{r,i} - e_{w,i}}{e_{r,i}}$. (RB: oversieved residues from separately collected biowaste; RC: oversieved organic waste after composting; RP: oversieved plastic materials from mixed waste treatment).

	RB			RC			RP		
	$e_{r,i}$ (mg/L)	$e_{w,i}$ (mg/L)	$\eta_{x,i}$ (%)	$e_{r,i}$ (mg/L)	$e_{w,i}$ (mg/L)	$\eta_{x,i}$ (%)	$e_{r,i}$ (mg/L)	$e_{w,i}$ (mg/L)	$\eta_{x,i}$ (%)
DOC	5400	3620	33	1014	378	63	338	280	17
COD	22800	13350	41	5505	1800	67	1532	832	46
BOD_5	10604	3652	66	500	207	59	350	295	16
TKN	439	236	46	415	154	62	70	21	70
NH_3-N	145	54	63	125	27	78	26	4	85
Chlorides	607	< 35	> 94	412	< 35	> 92	140	< 35	> 75
Cd	< 0.01	<0.01	0	< 0.01	<0.01	0	< 0.01	< 0.01	-
Cr	0.03	0.04	-	< 0.01	<0.02	-	< 0.02	< 0.01	> 50
Cu	0.11	0.13	-	0.32	0.15	55	0.45	0.246	45
Fe	1.27	1.72	-	1.78	1.5	16	2.36	1.2	49
Mn	5.73	2.84	50	0.1	0.11	-	0.41	0.48	-
Ni	0.06	0.06	-	< 0.01	< 0.01	-	0.17	0.0777	55
Pb	< 0.01	< 0.01	0	0.04	0.04	6	0.08	0.0373	54
Zn	0.74	1.48	-	0.1	0.12	-	0.77	0.627	19

3.2.6.2 Long term landfill leaching emissions: column tests

Long term landfill leaching emissions were simulated by using column tests, filled with both raw and washed residues. Leachate extracted was analysed for the most relevant parameters (COD, ammonia and Chlorides) at different L/S (**Figure 3.2. 7**).

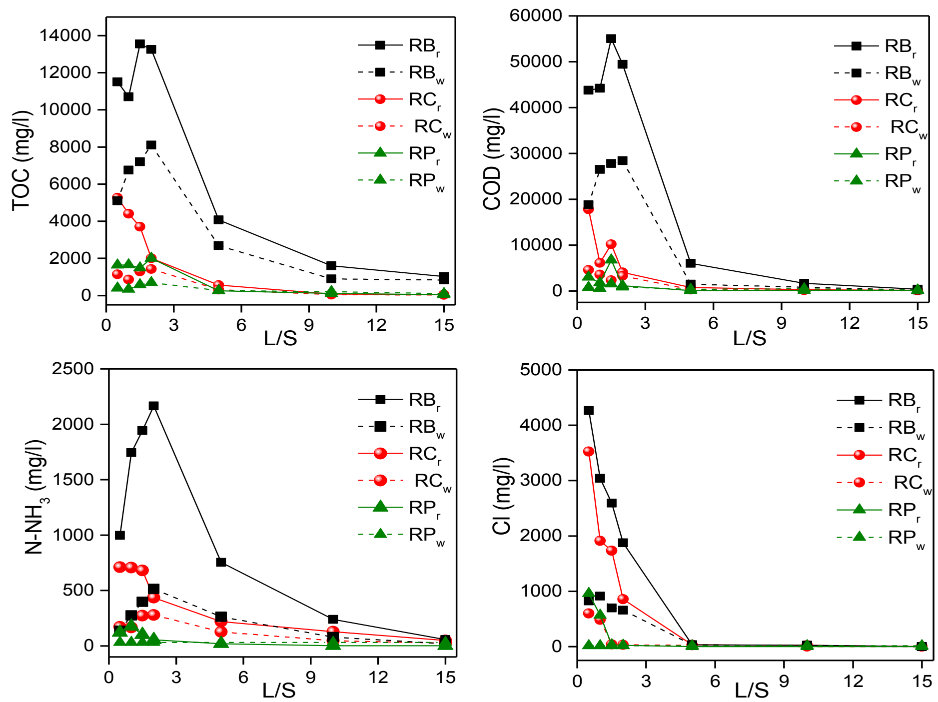

Figure 3.2. 7. Leachate characterisation for the target L/S ratios. (RB: oversieved residues from separately collected biowaste; RC: over-sieved organic waste after composting; RP: over-sieved plastic materials from mixed waste treatment; r: raw residues; w: washed residues).

COD, ammonia and Chloride concentrations in leachate from washed residues were always lower than leachate from raw waste. Washing was particularly effective on RB and RC residues, for which the maximum leachate contaminants concentrations were reduced respectively by approx. 40% and 70% in terms of COD, by approx. 75% and 60% in terms of ammonia and by approx. 80% in terms of chloride. Moreover, between L/S=0.5 and L/S=2 (corresponding to the first 30 years lifespan after landfill closure), a higher increase in COD and ammonia concentrations occurred in leachate from raw RB residues compared to the washed ones, due to organic substances hydrolysis and ammonification processes, demonstrating the effectiveness of washing pre-treatment in reducing the leachable potential emissions of residues, reducing the long term impact.

The positive effect of washing pre-treatment on inorganic compounds, is attested by the trend of Chlorides which concentration in short time reached very low values in all samples and particularly RP reached very soon the undetectable concentration.

The variation of the contaminants concentrations in leachate over the time was predicted considering an hypothetical landfill. **Figure 3.2. 8** illustrated the predicted concentrations of COD and ammonia over the time, of both raw and pre-washed residues. The results were compared to COD and ammonia FSQ limits set by Italian Regional Guideline, (1500 mgO_2/L and 50 mgN/L respectively), which should be achieved within 30 years after landfill closure in accordance with the sustainability concept.

Time required to reach COD and ammonia FSQ limits was always shorter in case of washed residues compared to the raw ones. However, washed RP residues was the only one respecting both COD and ammonia FSQ limits within 30 years after landfill closure.

When considering COD, the time to reach the FSQ limit was significantly shortened in washed RB residues, which reached the target limit in 100 years, 40 years earlier than the time reached by raw sample (140 years after closure). Conversely, RC residues showed a narrow duration gap (15 years) in reaching COD limits between raw and washed samples (76 and 61 years, respectively). With respect to RP samples, time required to reach COD limits is approx. 40 and 61 years respectively for washed and raw samples.

When considering ammonia, RC washed samples achieved the FSQ limit 177 years after landfill closure, significantly earlier compared to raw residues (around 266 years later). Raw and washed RB samples reached the limit respectively in 212 and 240 years. Washing was particularly effective in case of RP residues, accomplishing with FSQ limits since the beginning in case of washed samples. The results suggested that, although washing pre-treatment reduced the long term impact of all tested residues, additional combined in situ treatment, such as in situ aeration, would be required in case of RB and RC residues in order to improve the stabilisation of organic substances and removal of nitrogen.

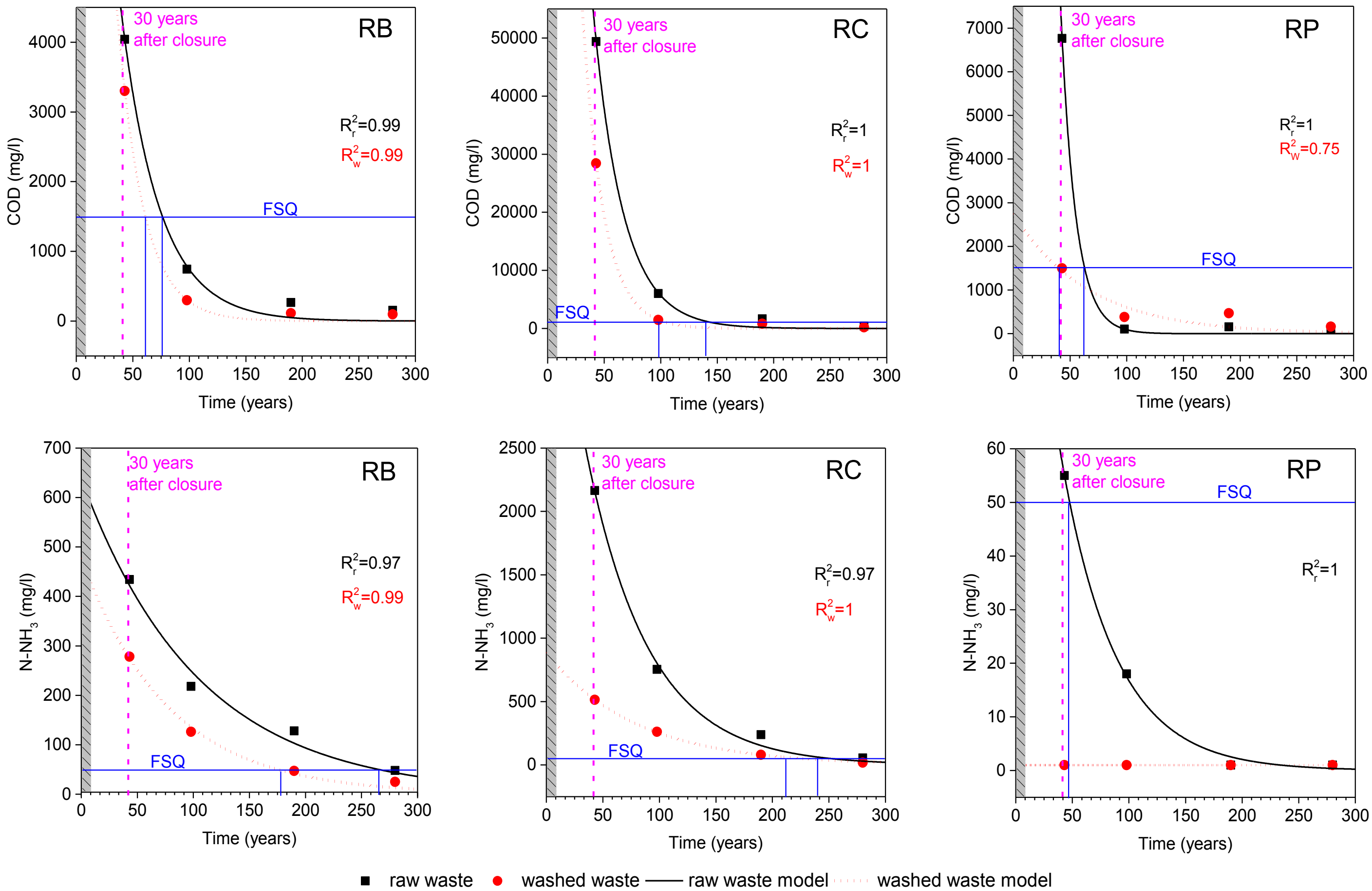

Figure 3.2. 8. Predicted concentrations of COD and ammonia over the time of an hypothetical landfill, filled with the different kind of residues, both raw and pre-washed. The reported FSQ limits, fixed by Italian Regional Guideline, are1500 mgO_2/L and 50 mgN/L respectively for COD and ammonia. The grey part of the graphs corresponds with the opening period of the landfill. The reference landfill sustainability time is 30 years after landfill closure, indicated in the graphs with the broken line. (RB: oversieved residues from separately collected bio-waste; RC: over-sieved organic waste after composting; RP: over-sieved plastic materials from mixed waste treatment; r: raw residues; w: washed residues).

3.2.7 Conclusions

The study evaluated the effectiveness of waste washing prior to landfilling on reducing the emission potential and long-term impacts of three different kinds of residues from municipal solid waste treatment, usually landfilled: residues sieved from separately collected bio-waste (RB); residues sieved from compost (RC); and residues sieved from mixed waste treatment-plastic line (RP).

The results obtained allowed the following conclusions to be drawn:

- The highest washing efficiencies (defined as the percentage reduction of the maximum potential leaching rates obtained by the washing treatment) were achieved in case of inorganic contaminants such as chlorides and ammonia, confirming the effectiveness of the washing pre-treatment particularly in case of residues with high leachable inorganic contaminants.
- The highest washing efficiencies of organic substances occurred in RB and RC residues, due to the high putrescible fraction, easily transferred to water. However, they resulted to be not sufficient for accomplishing the FSQ limits, due to the high organic substances content in the original residues.
- Time required to reach COD and ammonia FSQ limits was always shorter in case of washed residues compared to the raw ones, significantly reducing the long term emissions. However, washed RP residues was the only one respecting both COD and ammonia FSQ limits within 30 years. Due to the high contamination of not stabilised organics, in case of RB and RC, biological pre-treatments or in situ treatment, would be preferred in order to stabilise the organic substances and remove nitrogen.
- Other different washing procedures could be tested in order to improve the washing performance, such as by using multiple short washing runs (Colangelo et al., 2012).
- Although the washing of waste might produce significant quantities of wastewater to be treated, it may be compensated by the benefit of reducing the long term impacts of wastes, reducing the cost for the landfill aftercare.
- Further investigation should focus on the Life Cycle Assessment of the washing of different residues, to evaluate the short- and long-term impacts in both scenarios (with and without pre-treatment) including the impacts associated to the wastewater treatment. Moreover, the

stabilisation of contaminants, separated from wastewater through sludge should be properly addressed to assure their sustainable disposal.

- Under lab scale testing conditions, both washing efficacy and landfill operation are optimised by the shredding of the waste material and by the small, controlled landfill simulation. When the process is scaled up, both washing efficacy and long-term impacts should be evaluated under pilot scale conditions of by considering a proper safety factor.

References

Bogush, A.A., Stegemann, J.A., Zhou, Q., Wang, Z., Zhang, B., Zhang, T., Zhang, W., Wei, J., 2020. Co-processing of raw and washed air pollution control residues from energy-from-waste facilities in the cement kiln. Journal of Cleaner Production 254, 119924. https://doi.org/10.1016/j.jclepro.2019.119924

Chaudhuri, B., Ghosh, B., Datta, B.K., Bhattacharjee, S. 2001. Washing of waste plastics and characteristics of wash water. Journal of the Institution of Engineers (India): Environmental Engineering Division, 82(1), pp. 27-30.

Chen, X., Bi, Y., Zhang, H., Wang, J., 2016. Chlorides Removal and Control through Water-washing Process on MSWI Fly Ash. Procedia Environmental Sciences 31, 560–566. https://doi.org/10.1016/j.proenv.2016.02.086

Colangelo, F., Cioffi, R., Montagnaro, F., Santoro, L., 2012. Soluble salt removal from MSWI fly ash and its stabilization for safer disposal and recovery as road basement material. Waste Management 32, 1179–1185. https://doi.org/10.1016/j.wasman.2011.12.013

Cossu, R., 2016. Back to Earth Sites: From "nasty and unsightly" landfilling to final sink and geological repository. Waste Management, Volume 55, pages 1-2. https://doi.org/ 10.1016/j.wasman.2016.07.028

Cossu, R., Grossule, V., Lavagnolo, M.C., 2020a. What about residues from circular economy and role of landfilling? Detritus. https://doi.org/10.31025/2611-4135/2020.13920

Cossu, R., Lai, T., 2013. Washing treatment of automotive shredder residue (ASR). Waste Management 33, 1770–1775. https://doi.org/10.1016/j.wasman.2013.04.007

Cossu, R., & Lai, T., 2012. Washing of waste prior to landfilling. Waste Management 32, 869–878. https://doi.org/10.1016/j.wasman.2011.12.005

Cossu, R., Lai, T., Pivnenko, K., 2012. Waste washing pre-treatment of municipal and special waste. Journal of Hazardous Materials 207–208, 65–72. https://doi.org/10.1016/J.JHAZMAT.2011.07.121

Cossu, R., Sciunnach, D., Cappa, S., Gallina, G., Grossule, V., Raga, R., 2020b. First worldwide regulation on sustainable landfilling: guidelines of the Lombardy Region (Italy). Detritus. Volume 12, 114-124. DOI 10.31025/2611-4135/2020.14001.

Grossule, V., 2020. Final quality of a sustainable landfill and postclosure management. Detritus 13, 148–159. https://doi.org/10.31025/2611-4135/2020.13999

Grossule, V., Stegmann, R., 2020. Problems in traditional landfilling and proposals for solutions based on sustainability. Detritus 12, 78–91. https://doi.org/10.31025/2611-4135/2020.14000

Jiang, Y., Xi, B., Li, X., Zhang, L., Wei, Z., 2009. Effect of water-extraction on characteristics of melting and solidification of fly ash from municipal solid waste incinerator. Journal of Hazardous Materials 161, 871–877. https://doi.org/10.1016/j.jhazmat.2008.04.033

Keulen, A., van Zomeren, A., Harpe, P., Aarnink, W., Simons, H.A.E., Brouwers, H.J.H., 2016. High performance of treated and washed MSWI bottom ash granulates as natural aggregate replacement within earth-moist concrete. Waste Management 49, 83–95. https://doi.org/10.1016/j.wasman.2016.01.010

Kim, K., Yoon, S., Kwon, H. ah, Choi, Y., 2020. Effects of treatment agents during acid washing and pH neutralization on the fertility of heavy metal-impacted dredged marine sediment as plant-growing soil. Environmental Pollution 267, 115466. https://doi.org/10.1016/j.envpol.2020.115466

Kjeldsen, P., Barlaz, M.A., Rooker, A.P., Baun, A., Ledin, A., Christensen, T.H., 2002. Present and long-term composition of MSW landfill leachate: A review. Critical Reviews in Environmental Science and Technology 32, 297–336. https://doi.org/10.1080/10643380290813462

Lavagnolo, M.C., Grossule, V., 2018. From 3R to 3S: An appropriate strategy for Developing Countries. Detritus, Volume 04, 1-3. DOI:10.31025/2611-4135/2018.13749Luo, H., Zeng, Y., Cheng, Y., He, D., Pan, X., 2020. Recent advances in municipal landfill leachate: A review focusing on its characteristics, treatment, and toxicity assessment. Science of the Total Environment. https://doi.org/10.1016/j.scitotenv.2019.135468

Muola, A., Fuchs, B., Laihonen, M., Rainio, K., Heikkonen, L., Ruuskanen, S., Saikkonen, K., Helander, M., 2021. Risk in the circular food economy: Glyphosate-based herbicide residues in manure fertilizers decrease crop yield. Science of The Total Environment, Volume 750, 141422, https://doi.org/10.1016/j.scitotenv.2020.141422.

Pivnenko, K., Laner, D., Astrup, T.F., 2016. Material Cycles and Chemicals: Dynamic Material Flow Analysis of Contaminants in Paper Recycling. Environ. Sci. Technol. 2016, 50, 12302−12311. https://doi.org/10.1021/acs.est.6b01791.

Sun, X., Xu, Y., Liu, Y.Q., Nai, C.X., Dong, L., Liu, J.C., Huang, Q.F., 2019. Evolution of geomembrane degradation and defects in a landfill: Impacts on long-term leachate leakage and groundwater quality. Journal of Cleaner Production Volume 224, 1 July 2019, Pages 335-345. https://doi.org/10.1016/j.jclepro.2019.03.200

Sun, X., Yi, Y., 2021. Acid washing of incineration bottom ash of municipal solid waste: Effects of pH on removal and leaching of heavy metals. Waste Management, Volume 120, Pages 183 – 192. , https://doi.org/10.1016/j.wasman.2020.11.030

Wei, Y. M., Yao R. X, Chen S., Zhou H.L., Liu S.J., 2021. Removal of chloride from MSWI fly ash: a comparison of accelerated carbonation and water flushing. Zhongguo Huanjing Kexue/China Environmental Science. 41(9), pp. 4184-4192

Wang, X., Wang, M., Zou, D., Wu, C., Li, T., Gao, M., Liu, S., Wang, Q., Shimaoka, T., 2020. Comparative study on inorganic Cl removal of municipal solid waste fly ash using different types and concentrations of organic acids. Chemosphere 261, 127754. https://doi.org/10.1016/j.chemosphere.2020.127754

Yan, D., Peng, Z., Yu, L., Sun, Y., Yong, R., Helge Karstensen, K., 2018. Characterization of heavy metals and PCDD/Fs from water-washing pretreatment and a cement kiln co-processing municipal solid waste incinerator fly ash. Waste Management 76, 106–116. https://doi.org/10.1016/j.wasman.2018.03.006

4. Part Two: Column Leaching Test

Based on: Q Luo., V Grossule., MC Lavagnolo. Significance of self-designed column and standard batch leaching tests in view of the sustainability of landfills.

Submitted: *Waste Management*

Abstract

Long-term emissions generated from a landfill achieve a Final Storage Quality (FSQ) in equilibrium with the surrounding environment within a generation (typically 30 years) and pose no potential risks are the critical indicator defining the sustainability of a landfill. The aim of the study was to investigate the significance of both column flow-percolations and standard batch leaching tests (UNI EN 12547-2) in view of assessing sustainability of a landfill, as well as evaluating the FSQ. Both self-designed column leaching tests and standard batch leaching tests were performed on residues from an old landfill (over 18 years). Different liquid/solid ratio (L/S) irrigation and frequencies were applied in column leaching tests simulating different rainfalls, in order to investigate the effect of different conditions on long-term emissions. The results revealed that values obtained from standard batch leaching tests were not enough to assess the sustainability of a landfill, and overestimated the real emissions occurring under landfill conditions. Irrigation frequency in column leaching tests influenced both organics and inorganics variation only under low water input conditions (L tests): higher the irrigation frequency, higher the flushing effects and faster the concentration variation. Conversely, no significant influence was detected under high water input conditions (H tests). Furthermore, a significant impact caused by irrigation frequencies is reducing waste emission potential, implying possible improvements in in-situ treatments. Meanwhile, values of ammonia nitrogen revealed that achieving FSQ limits requires a longer period would over 30 years.

Keywords: Colum leaching test, standard batch leaching test, Final Storage Quality (FSQ)

4.1. Introduction

Sustainable landfill plays the fundamental role of closing the loop of the materials which cannot be further reintroduced in the Circular Economy (CE), giving them Back to the Earth (BEA, Cossu, 2016) in a stable, not contaminating form. The concept of BEA implies that the CE residues, after proper treatment, should be returned to their non-mobile state, as they were before they were extracted from the ground to be used as raw materials. A landfill is considered sustainable and the BEA properly fulfilled when, the emission quality achieved in a generation time (30 years) does not have any significant environmental impact, achieving a Final Storage Quality (FSQ) in equilibrium with the surrounding environment, so an active control is no longer required.

Accordingly, the landfill should be designed and developed in line with the principle of environmental sustainability, adopting all measures required to control the waste stability and immobilisation of contaminants (Grossule & Stegmann, 2020), such as (Cossu et al., 2020)

- minimization of the quantities of contaminants to be landfilled and of their potential emissions by means of specific pre-treatments;
- maximisation of the controlled removal of mobile contaminants, by means of biogas and leachate generation and extraction;
- maximisation of waste stabilisation by means of in situ treatments;
- use of physical barriers to control and remove mobile contaminants (bottom liner, drainage, emissions collection, etc.,).

The effectiveness of the designed measures adopted for the sustainable landfill and the capability of achieving the FSQ within one generation time should be properly assessed a priori. Being leachate the main cause for long-term environmental impact of landfills, the leachate quality forecast is an effective way of predicting the sustainability of the landfill.

Leaching tests represent the typical tool performed to assess the contaminants leachability potential from waste, in which a liquid solvent is put in contact with the solid material. Leaching tests allow to achieve several main goals: (1) predicting the total quantity of potential contaminants can be released; (2) the time of waste samples achieves the equilibrium with environment; (3) physicochemical and geochemical changes of samples undergo with time; (4) the difference of time-dependent release before and after the proper treatment; (5) effects of leaching solvents and waste properties (Freyssinet et al., 2002.; Grathwohl & Susset, 2009; Luo et al., 2019; Pecorini et al., 2017; Tiwari et al., 2015)

In general, leaching tests can be categorised in two macrocategories:

- Batch tests: single-addition leachant and shaking (constant and high Liquid/Solid ratio (L/S))
- Column tests: renewed leachant and flow-percolating (accumulative L/S).

In addition to the leaching method, leaching behavior and results are also greatly dependent on following factors: pH of solvents, the time of water in contact with the waste solid matrix, the L/S ratio, temperature, and the intrinsic characteristics of the sample itself (particle size distribution, chemical composition and the solubility of the mineral phases constituting the residues, etc.) (Luo et al., 2019b; Parodi et al., 2011). Numerous studies investigated leaching mechanisms (Grathwohl, 2014; Maszkowska et al., 2013; Olgun et al., 2013; Sazali et al., 2020), and leaching results presented differences of orders of magnitude, especially as a function of pH, but also in terms of the L/S ratios.

Examples of standard leaching tests are collected and briefly described in **Table 4. 1**.

Table 4. 1. Standard leaching tests: column and batch leaching tests.

Category	Filling process	L/S (l/kg)	Time	pH	Leachants	Particle size	Standards
Column	Saturated for 16 h up to three days, then vertical up flow irrigation at a rate of 10-20 ml/h	0-10		5 - 7.5	Distilled water/ nitric acid/acetone	< 4 mm	EN 14405 (2017)
	up-flow at a rate of 12 ml/h	0-10			Distilled water/ seawater	< 50 mm	EN 14405 (2017)
	saturated for three days, then the pump was started again setting a flow rate of around 48 ± 5 ml/h	0-10			Distilled water	< 12 mm	EN 14405 (2017)
	saturated for three days, then the upwards flow rate was set at 72.1 ml/h	0-10			Distilled water	< 4 mm	EN 14405 (2017)
	saturated for three days, then the up flow rate was set at 10-15 ml/h	0-10			Distilled water		EN 14405 (2017)
	saturated for 16 h up to 72 h, then percolated for 10 -24 h	0-10	10-24		Distilled water	4-10 mm	EN 14405 (2017)
	upwards flow rate of 15 cm/day	0-10			Distilled water	< 4 mm	EN 14405 (2017)
	saturated for three days, then the up flow rate was set at 13 ml/h	0-10			Deionised water	< 4 mm	EN 14405 (2017)
Batch	First step: shaken for 6 h; then collected leachates; second step: renown regents, shaken for 18 h	First step: 2; second step: 8	First 6 h; second 8 h		Distilled water	2-10 mm	EN 12457-3
	Agitation	2	24 ± 0.5 h		Water	< 4 mm	EN 12457-1 (2002)
	Agitation	10	24 ± 0.5 h		Water	< 4 mm	EN 12457-2 (2002)
	Agitation	First step 2; second step 8	6 ± 0.5 h; 8 ± 0.5 h		Water	< 4 mm	EN 12457-3 (2002)
	Agitation	10	24 ± 0.5 h		Water	< 10 mm	EN 12457-4 (2002)
	Rotated at speed of 30 ± 2 rpm	5 or 10 or 20 or 50	48 h	3	deionized water	< 2 mm	TCLP 1311 (US 1999)
	Rotated at speed of 10 rpm	10	24 h		Distilled water or seawater	< 2 mm and 2 – 50 mm	EN 12457-2 (2002)
	Shaked	10	24 h		Distilled water	< 4 mm	EN 12547-2 (2002)

Continued Table 4. 2. Standard leaching tests: column and batch leaching tests.

Category	Filling process	L/S (l/kg)	Time	pH	Leachants	Particle size	Standards
	Agitation at 30 ± 2 rpm	20	18 h	2.88 ± 0.05	Glacial acetic acid+ water	< 4 mm	TCLP 1311 (US 1999)
	Agitation at 10 ± 2 rpm	10	24 ± 0.5 h		Deionized water	< 4 mm	EN 12457-2 (2002)
	Agitation	10	24 h		Demineralized water	< 4 mm	EN 12457-2 (2002)
	Stirred at 10 rpm	10	24 h		Deionized water	0.1 - 4 mm	EN 12457-2 (2002)
	Tumbled at a speed of 30 rpm	20	18 h		Mixed leaching fluids (pH=4.93 and pH=2.88)	< 4.75 mm	TCLP 1311 (US 1999)
	Tumbled at a speed of 30 rpm	20	18 h		Mixed leaching fluids (pH=4.20 and pH=5.00)	< 4.75 mm	SPLP 1312 (US 1994)
	Rotated at 10 rpm	10	24 h		Distilled water	< 4 mm	EN 12457-2 (2002)
	Rotated at 10 rpm	First step 2, second step 8	First 6 h, second 8 h		Distilled water	< 4 mm	EN 12457-3 (2002)
Batch	Rotated at 15 rpm	20	2 h	7.3	Buffered solution	< 4 mm	ISO 14870
	Rotated at 15 rpm	20	18 ± 2 h	2.88 ± 0.05	Glacial acetic acid+ deionized water	< 4 mm	TCLP 1311 (US 1999)
	Rotated at 15 rpm	200	18 ± 2 h	5 ± 0.05	Sulfuric and nitric acid+ deionized water	< 4 mm	SPLP 1312 (US 1994)
	Rotated at 15 rpm	2.5	2		Ammonia acid	< 4 mm	ISO 19730
	Rotated 5-10 rpm	10	24 h		Distilled water		EN 12457
	Rotated 1000 rpm	10	48 h	10	Distilled water		prEN 14997
	Stirred at 250 rpm	2	24 h		Deionized water	< 4 mm	EN 12457-1
	Stirred at 250 rpm	10	24 h		Deionized water	< 4 mm	EN 12457-2

Continued Table 4. 3. Standard leaching tests: column and batch leaching tests.

Category	Filling process	L/S (l/kg)	Time	pH	Leachants	Particle size	Standards
	Stirred at 250 rpm	20	18 h		NaOH + Acetic acid	< 1 mm	TCLP 1311 (US 1999)
	Rotated at 13 rpm	20	18 ± 2 h	4.93 ± 0.05	Acetic acid and sodium hydroxide	< 9.5 mm	TCLP 1311 (US 1999)
	Rotated at 13 rpm	20	18 ± 2 h	4.20 ± 0.05	Sulfuric and nitric acids	< 9.5 mm	SPLP 1312 (US 1994)
	Four sequential extraction steps		18 h; 42 h; 66 h; 90 h	4.20 ± 0.05	Acid solution	< 6.4 mm	SPLP 1312 (US 1994)
	Shaked	16 - 20	24 h	5.0 ± 0.2	Acetic acid	< 9.5 mm	TCLP 1311 (US 1999)
Batch	Shaked	20	18 ± 0.3 h	5.0	Sulfuric/nitric acid	< 4.75 mm	SPLP 1312 (US 1994)
	Shaked	20	18 ± 0.3 h	3.0	Acetate buffer	< 4.75 mm	TCLP 1311 (US 1999)
	Shaked	10	24 h		Demineralized water	< 4 mm	EN 12457-2
	Two steps and rotated	First 6 and second 10	10 h		Demineralized water	5 mm	EN 12457-3
	Shaked	10	24 h		Deionised water		EN 12457-2
	Shaked	10	24 h		Deionised water + 1 mM CaCl		ISO/TS 21268-2:2010

As the feature of simple and fast operation, batch leaching test is extensively applied to leaching investigation (di Gianfilippo et al., 2016). Standard batch leaching test (UNI EN 12547-2) (L/S =10, T=24 h) is the official method used in many European countries to determine potential risk of waste materials (Cossu et al., 2012; Cossu & Lai, 2013; Ettler & Johan, 2014; Mantis et al., 2005), and it is used to establish the admissibility criteria to landfill by Italian legislations.

Batch test is typically established by the contact between the solid phase and the chemistry of the liquid phase rather than by contact time. As a consequence, biological phenomena are not taken into account during the fast contact leaching process, leading to imprecise estimating of organic pollutants that evolve during the phases of degradation (di Gianfilippo et al., 2016). Moreover, the typically high L/S ratio, applied in batch test, assures the determination of a potential contaminants release but in most of the cases unrealistic and with no information of the concentration evolution till the achievement of a pseudo-equilibrium conditions (Galvín et al., 2012; Jiao et al., 2016), missing the goal of ascertain the FSQ achievement and thus the landfill sustainability.

Compared with the statistic batch leaching tests, dynamic leaching tests with flow-through system and low-L/S ratio demonstrate to achieve a more realistic simulation of the natural conditions (López Meza et al., 2010; Maszkowska et al., 2013). Slow flow-percolations prolong the contact of solid and liquid phase, not only simulating a closer natural condition, but also accumulating possibly released both inorganic and organic pollutants, which improve the accuracy of conclusions (Gupta et al., 2019). However, uncertainty still exists, related to the test duration (particularly in case of biodegradable waste), appropriate choice of L/S ratio and irrigation frequencies, which influencing the flow velocities would lead to a wrong estimation of the released concentration that may be reached under field conditions (Grathwohl, 2014).

With the purpose of properly estimate the long-term emission from landfill, in view of assessing the landfill sustainability by achieving the FSQ target, L/S ratio corresponding to one generation time can be defined according to an estimation model of leachate production based on mass balance of water influx and water released from the landfill, considering realistic rainfall data, waste density value and effect of final top cover (Cossu et al., 2012). Regarding the irrigation frequencies, their influence on results and the need of properly adjust them is still unknown.

Consequently to the discussed limitations of leaching tests, the objectives of the present study were the following:

- Investigate the significance of both column flow-percolations and standard batch leaching tests (UNI EN 12547-2) in view of assessing the landfill sustainability.
- Investigate the effect of different L/S ratios (corresponding to one generation time under different rainfall/landfill conditions) and irrigation frequencies on the estimation of long-

term emissions. FSQ targets set by Italian Regional Guideline (Grossule, 2020) were used to ascertain the accomplishment of sustainability.

4.2. Materials and Methods

4.2.1 Research Program Scheme

The research program is graphically represented in **Figure 4. 1**.

Standard batch leaching tests (UNI EN 12547-2) (L/S = 10 l/kg_{TS}, TS = Total solids; T = 24 h) were performed at the beginning of the experiment on raw waste to evaluate the waste emission potential. The results were compared with landfill admissibility criteria set by Italian legislation.

Column leaching tests were conducted on raw waste samples to simulate different landfilling conditions by adding different cumulative water quantities: 1.5 L/kg_{TS} and 3 L/kg_{TS}, defined as low (L) and high (H) water input. Both water input quantities were irrigated at different frequencies: 10, 20 and 40 times, distributed over a period of 40 days. Leachates gained from column tests were analyzed and final leachate quality was compared to FSQ target fixed by Italian Regional Guideline (Grossule, 2020).

Waste samples after column tests were testes by standard batch leaching test, to estimate the waste emission potential after 30 years landfilling.

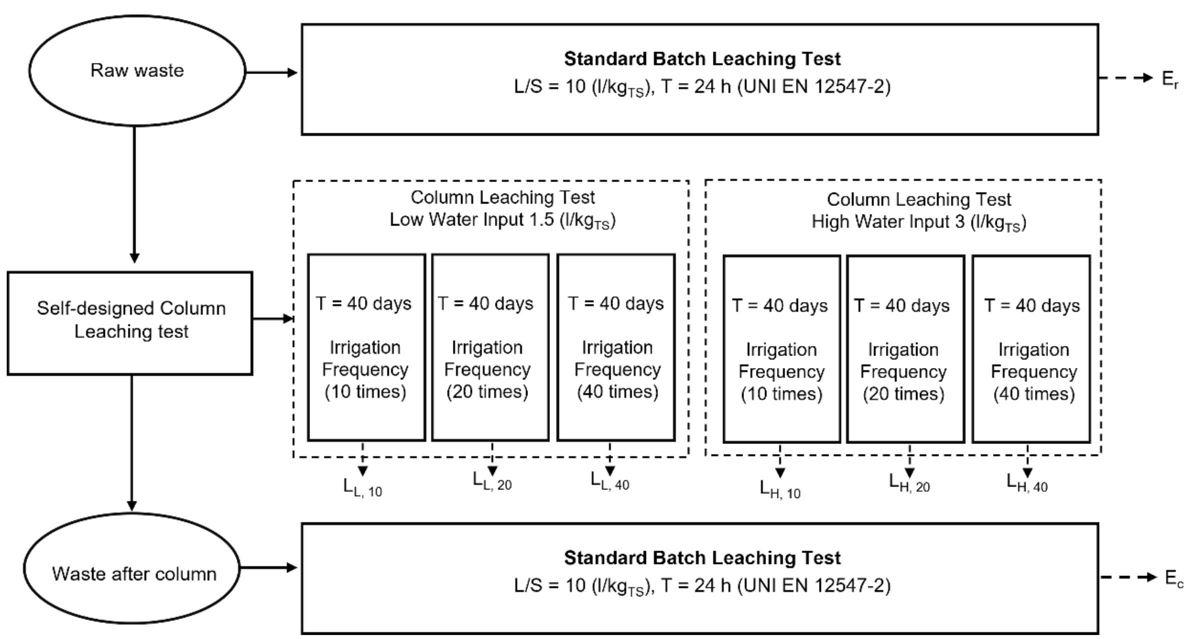

Figure 4. 1. Scheme of the research program (L/S = Liquid/Solid ratio; T= testing time; E_r = Eluates from batch leaching test of raw waste, and E_c = Eluates from batch leaching test of waste

gained after column test; L_L, L_H = leachates of column test with low and high quantity of input water, respectively, and numbers represent irrigation frequency).

4.2.2 Waste Material

Waste material was collected from an old landfill (> 18 years) located in central Italy, and both municipal solid waste and non-hazardous industrial waste were buried in this landfill. Considering the heterogeneity of buried residues, quartering method (**Figure 4. 2**) was applied to collect the representative sample of about 230 kg. Compositions of the waste material were determined by means of a trade analysis (UNI 10802:2013). Large fractions of sample were shredded to a size < 20 mm for the column leaching tests and to a size of 4 mm for standard batch leaching test. Total Solids (TS), Volatile Solids (VS), seven days Respirometric Index (RI_7) and metals were analyzed on the sample.

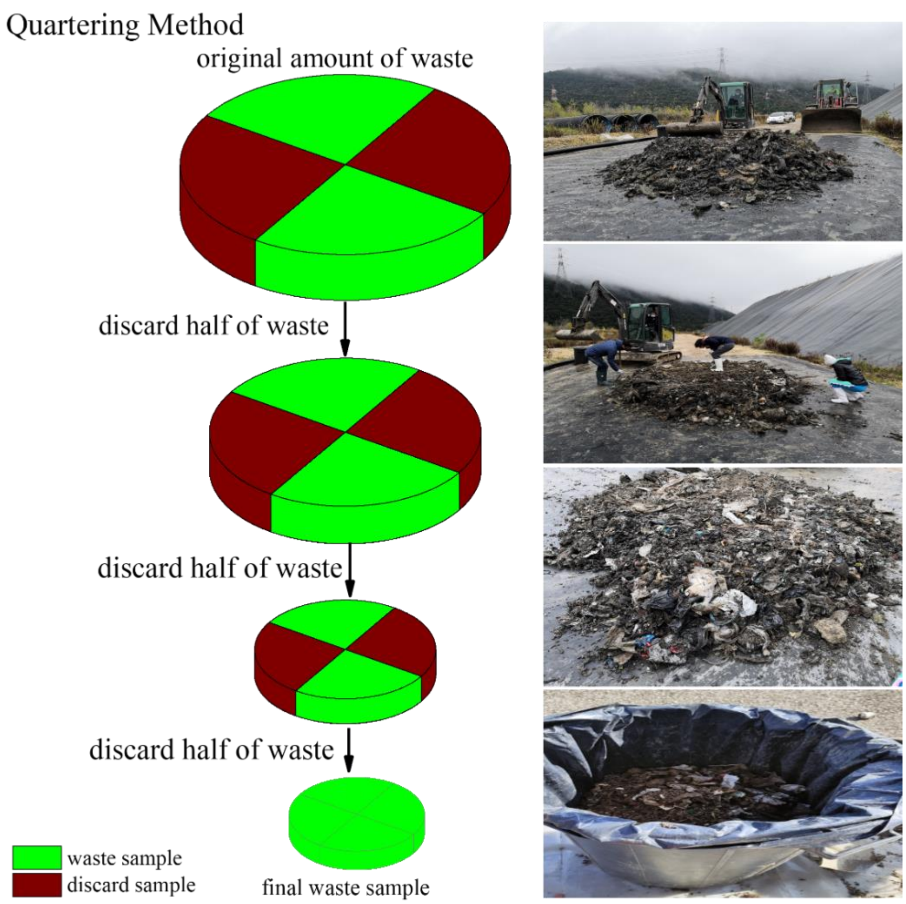

Figure 4. 2. Quartering methods for collecting the waste sample**.**

4.2.3 Analytical Methods

The parameters analyzed on both solid waste and liquid samples gained from both standard batch leaching tests and self-designed column leaching tests were TS, VS, RI_7, COD, NH_4-N, TOC, Chlorides, Sulfates, pH, and metals, respectively. Detailed methods and equipment regarding analyses of all tested solid and liquid samples are reported in

.

Table 4.2 Standard methods used in analysing solid and liquid samples

Parameter	Sample	Method
pH	liquid	IRSA-CNR 29/2003 vol.1 n°2060
RI_7	Solid	Sapromat and VoithSulzer Respiromat
TS, VS	solid	IRSA-CNR Q.64/84 vol.2 n°2
NH_4-N	liquid	IRSA-CNR 29/2003 vol.1 n°4030$_C$
BOD_5	liquid	IRSA-CNR 29/2003 vol.2 n°5120$_{A, B, B2}$
TOC	liquid	Shimadzu TOC-VCSN analyzer
COD	liquid	IRSA-CNR 29/2003 vol.2 n°5130
Metals (Cd, Co, Cr, Cu, Fe, Mn, Mo, Ni, Pb, V, Zn)	liquid	IRSA-CNR 29/2003 vol.1 n°3010
Chlorides	liquid	IRSA-CNR 29/2003 vol.2 n°4090$_B$
Sulphates	liquid	IRSA-CNR 29/2003 vol.2 n°4140$_B$

4.3. Results and Discussion

4.3.1 Raw Waste Analysis

Trade composition and granulometry of raw solid waste are illustrated in **Figure 4. 3**. Almost half amount (49.0%) of the raw waste consists of undersieve (fine fractions < 20 mm). The coarse fractions are mainly characterized by inerts (e.g. stones and glass) and plastics (rubber and mixed plastics), accounting for 22.2% and 16.8%, respectively. Textile and wood contributed 8% of the total, followed by metals (2.2%). Paper was in the lowest proportion (1.0%) due to the long-term biodegradation (**Figure 4.3a**). The composition reflects the origin of residues and affects their biological stability, and carbon and nitrogen content. The characteristics of raw solid waste samples are illustrated in **Table 4.3** and **Table 4.4**. The low values of RI_7 (1.3 mg/g_{TS} and BOD/COD ratio (approx. 0.17) indicated an high biological stability of waste sample (Cossu and Stegmann, 2018; Kjeldsen et al., 2002), which is corresponding to the old landfilling residues. Relatively high heavy metals (e.g. Fe 360 µg/l, Mo 88.7 µg/l, Cu 79.1 µg/l, etc.) concentrations are due to the presence of industrial waste.

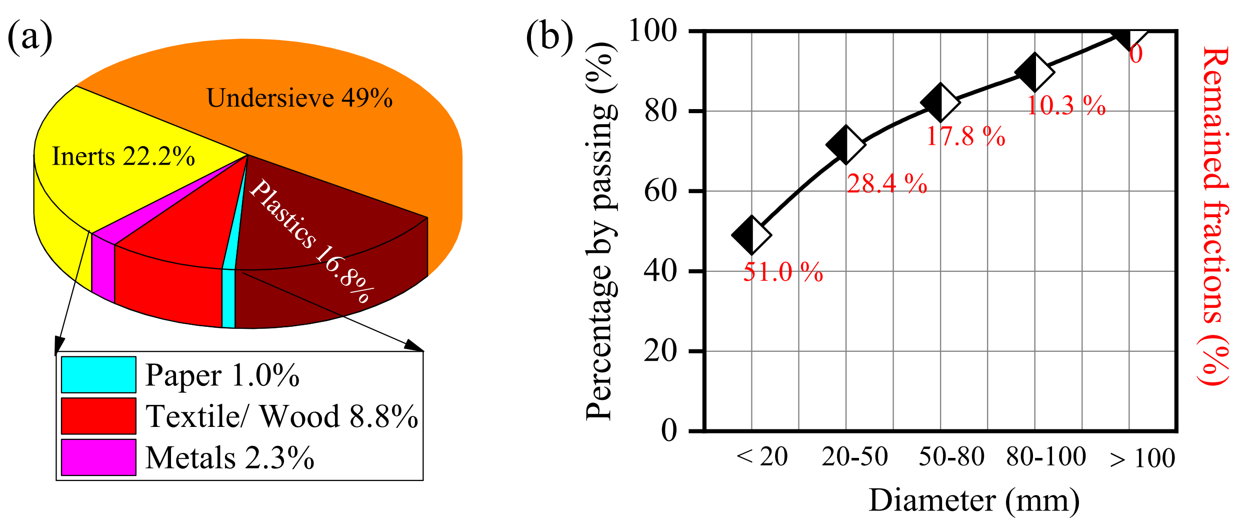

Figure 4. 3. Trade composition of raw residues (% on weight) and granulometric distribution for the waste sample (% on weight).

Regarding the granulometry of waste, the majority of the material had a dimension smaller than 20 mm, accounting for the same proportion (49.0%) as undersieve fraction (**Figure 4.3b**). The second large proportion was (28.4%) fraction's dimension was between 20 and 50 mm, followed by two relatively large sizes (50-80, 80-100 mm), accounting for 17.8% and 10.3%, respectively. Considering granulometric distribution of residues sample influenced by the composition of the waste, the coarse fractions were shredded for further analyses.

Table 4.3 Characterization of the raw residues sample.

Parameters	RI_7	TS	VS	Cd	Co	Cr	Cu	Fe	Mn	Mo	Ni	Pb	V	Zn
Units	mg_{O_2}/g_{TS}	g_{TS}/kg_{RW}	g_{VS}/kg_{TS}	µg/l	µg/l	µg/l	µg/l	µg/l	µg/l	µg/l	µg/l	µg/l	µg/l	µg/l
Raw waste	1.3	710	210	< 10	< 10	< 10	79.1	360	12.5	88.7	40	< 10	19.2	36.4

4.3.2 Standard Batch Leaching Test on Raw Waste

Eluates from standard batch leaching test of raw waste were analyzed for TOC, COD, ammonia nitrogen, chlorides and sulphates and the results are reported in **Table 4. 4.** The same table provides the limits fixed by the Italian legislation (D.M. 27/09/2010) for TOC, COD, chlorides and sulphates, for the acceptability of waste in non-hazardous landfill. All parameters (both organic and inorganic contaminants) complied with limits established for disposal in non-hazardous waste landfills.

Table 4. 4. Quality of eluate from standard batch leaching test on raw waste.

Standard batch Leaching test	TOC (mg/l)	COD (mg/l)	BOD_5 (mg/l)	BOD/COD	NH_4-N (mg/l)	Cl^- (mg/l)	SO_4^{2-} (mg/l)
Raw waste	60	250	42.5	0.17	69	63	235
Limits for non-hazardous landfill	100	-	-	-	-	2500	5000

4.3.3 Column Leaching Test of Raw Waste

Long-term landfill leaching emissions were simulated by using column leaching tests (**Figure 4. 4**), in order to investigate whether the emission quality achieved in a generation time does have any significant environmental impact and does achieve the FSQ in equilibrium with the surrounding environment.

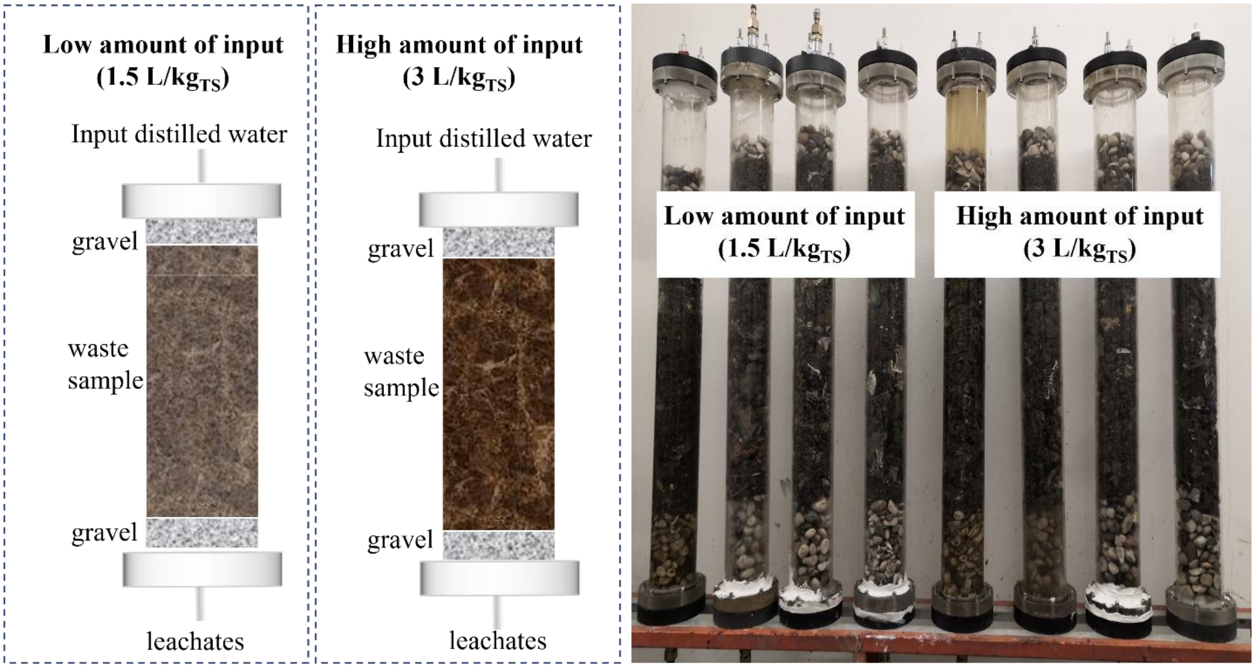

Figure 4. 4. Self-designed Column leaching tests apparatus.

According to the landfill hydrological model applied by Cossu and Lai, 2012, two water input (high-H and low-H) were chosen and tested to simulate two L/S corresponding to 30 years after the closure of a landfill under different landfill conditions, respectively of 1.5 and 3 L/kg$_{TS}$. Particularly, the L/S achieved in one generation time might differ due to different rainfall conditions and landfill top cover typologies.

Both water inputs were irrigated with different frequencies over a period of 40 days.

The variation of inorganic parameters, such as ammonia nitrogen, chlorides and sulfates, under different irrigation conditions are illustrated in Figure 3. In the same figure, FSQ limits are also illustrated.

The presence of ammonia nitrogen in leachate is one of the main factors causing the longest-lasting environmental impacts (Kjeldsen et al., 2002). It therefore becomes the critical parameter for evaluating the sustainability of a landfill.

With respect to the quantity of irrigation, the fastest decrease of NH_4-N occurred in L tests, (**Figure 4. 5**) probably due to the combination of both biological degradation and flushing effect. Presence of biological degradation was also suggested by the initial increase of ammonia concentration, that might be justified by ammonification processes, particularly evident under high frequency irrigation conditions (L_{40}). Conversely, the solely flushing effect occurring in H tests may justify the slowest variation of ammonia concentrations. As a result, all L tests (with the exception of L_{40}) achieved the FSQ limit within the 30 years L/S (1.5 L/kg_{TS}), while none of the H tests achieved the FSQ target.

Irrigation frequency influenced ammonia variation only under low water input conditions (L tests): higher the irrigation frequency, higher the flushing effects and faster the ammonia variation. The only exception was test L_{40}, where ammonification increased the leachable nitrogen, and ammonia concentrations remained higher compared to the other L tests. Conversely, no influence occurred under high water input conditions (H tests), implying the rainfall frequency might not have the same significant impact on NH_4-N emission as rainfall quantity under high rainfall conditions.

With regard to Chlorides and Sulfates, flushing effect was discovered as the main factor contributing to decreasing the concentrations both in high- and low-quantity irrigation tests. Due to the highly readily leachability of chlorides and sulfates, those two inorganic compounds are easy to be extracted by water (Alam et al., 2017). Therefore, various irrigation frequencies didn't affect significantly the concentration variations. Furthermore, concentrations of chlorides and sulfates in all groups have been found to reach the FSQ within 30 years, demonstrating leachable inorganic compounds are easy to achieve equilibrium with the environment.

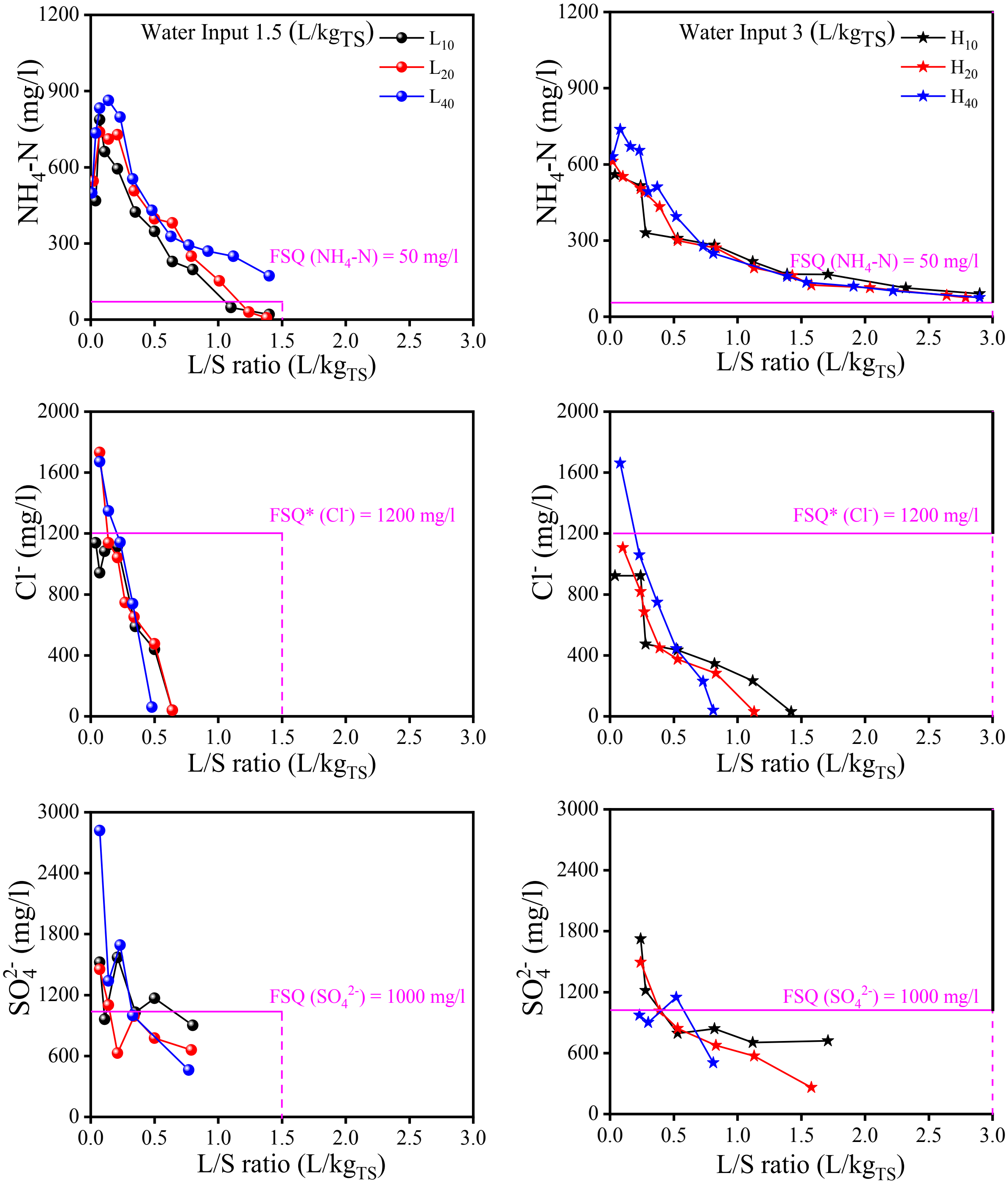

Figure 4. 5. Inorganic values of leachates generated from low and high groups of column leaching tests. L represents a low-quantity of water irrigation (1.5 L/kg$_{TS}$), H represents a high-quantity of water irrigation (3 L/kg$_{TS}$). Numbers 10, 20 and 40 represent 10-time, 20-time and 40-time input frequencies during 40 days, respectively.

Variation of organic contaminants concentration, such as COD and TOC, are illustrated in **Figure 4.6**. Being quite low the BOD_5 value and BOD/COD ratio measured in eluates from batch leaching test of raw waste (**Table 4.4**), most of organics can be considered as not biodegradable, and the variation of both COD and TOC can be ascribed in all tests to the only flushing effect.

COD concentrations were below FSQ limit in all tests, however both COD and TOC variation were influenced by the input water: fastest concentration decrease occurred in L tests, in particular in high frequency irrigation test (L_{40}), but lowest concentrations were achieved in H tests.

Irrigation frequency influenced organics variation only under low water input conditions (L tests): higher the irrigation frequency, higher the flushing effects and faster the concentration variation. Conversely, no significant influence was detected under high water input conditions (H tests), similarly to ammonia variation.

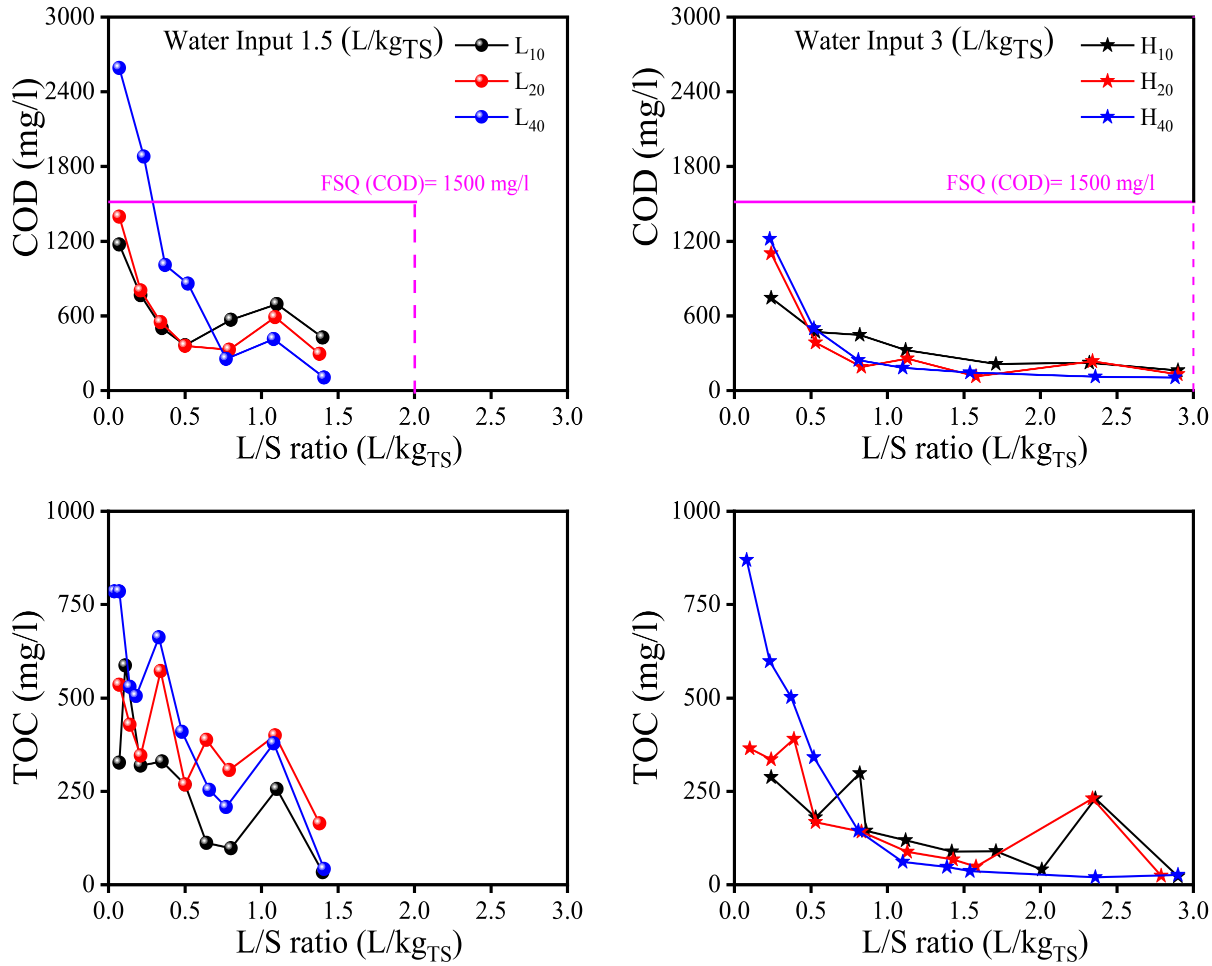

Figure 4. 6. Organic values of leachates generated from low and high groups of column leaching tests. L represents a low-quantity of water irrigation (1.5 L/kg$_{TS}$), H represents a high-quantity of water irrigation (3 L/kg$_{TS}$). Numbers 10, 20 and 40 represent 10-time, 20-time and 40-time input frequencies during 40 days, respectively.

4.3.4 Cumulative Contaminants Extraction from Column Leaching Tests

The cumulative contaminants extraction from column leaching tests normalized to the maximum potential emissions (provided by the standard batch leaching test on raw waste) is illustrated in **Figures 4.7** and **4.8.**

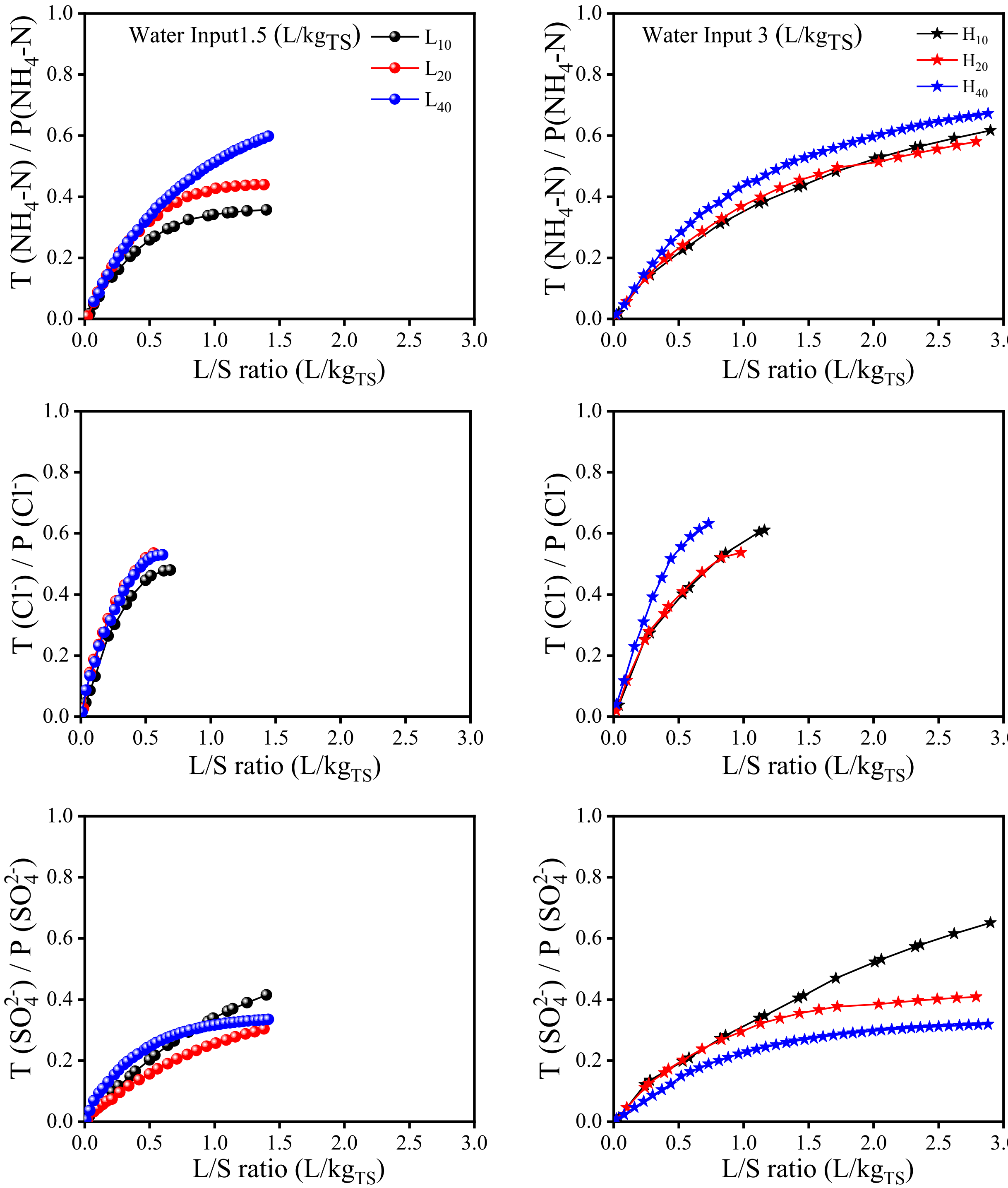

Figure 4. 7. The ratio between total inorganic contaminates collected from column leaching tests and the potential inorganic values generated from standard batch leaching tests. (T: total amount of values accumulated in column leachates; P: potential released values in batch leachates)

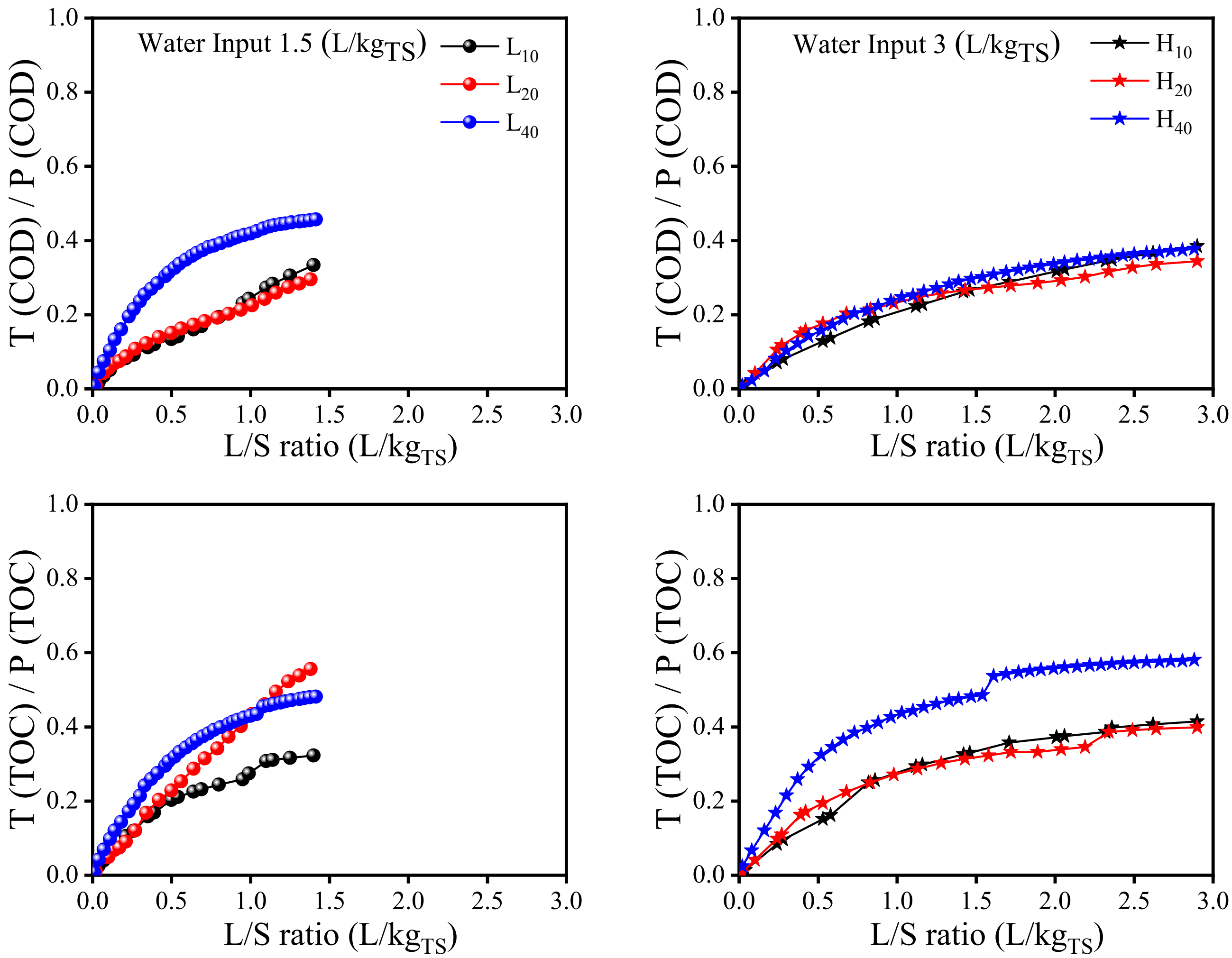

Figure 4. 8. The ratio between total organic contaminates collected from column leaching tests and the potential organic values generated from standard batch leaching tests. (T: total amount of values accumulated in column leachates; P: potential released values in batch leachates)

In general, the high frequency was found to contribute to high accumulative releasing amounts of both organic and inorganic contaminants, both in low-quantity and high-quantity irrigation groups: the accumulative amounts were in $L_{40} > L_{20} > L_{10}$, $H_{40} > H_{20} > H_{10}$. The only exception was sulphates. According to this result, higher the irrigation frequency, higher the reduction of the waste emission potential.

In general, for all parameters under all testing conditions, the cumulative contaminants extraction reached a plateau at values between 0.2-0.8, meaning that the accumulative emissions from column leaching tests are quite far from the potential emissions assessed by means of batch leaching tests, which are overestimating the emissions from a real landfill.

4.3.5 Standard Batch Leaching Test on Waste After the Column Leaching Tests

Standard batch leaching tests were conducted on the waste collected from column leaching tests in order to evaluate the residual emission potential. The results obtained are illustrated in **Table 4.5**. Comparing the quality of eluate from batch leaching tests of raw waste (**Table 4.4**) and of waste after column leaching test (**Table 4. 5**), the emission potential resulted to be significantly reduced after 30 years landfill simulation. In particular the highest emission potential reduction occurred in L tests and under high irrigation frequency (L_{40}, H_{40}), consistently with results from column leachate test. Those values are corresponding to the results gained in **Figures 4.7** and **4.8**: improving the irrigation frequency could enhance the extraction amount of contaminates, meanwhile, fewer amounts of contaminates remained in the waste (the waste in the column). Therefore, irrigation frequency should be taken into account for landfilling treatment.

Table 4. 5. Quality of eluates from standard batch leaching test of waste from column tests. (L represents a low-quantity of water irrigation (1.5 L/kg_{TS}), H represents a high-quantity of water irrigation (3 L/kg_{TS}). Numbers 10, 20 and 40 represent 10-time, 20-time and 40-time input frequencies during 40 days, respectively.)

Standard batching leaching test	TOC (mg/l)	COD (mg/l)	NH_4-N (mg/l)	Cl^- (mg/l)	SO_4^{2-} (mg/l)
Waste after column-L_{10}	13	78	18	< 35	122
Waste after column-L_{20}	12	120	4	< 35	99
Waste after column-L_{40}	< 10	78	2	< 35	< 10
Waste after column-H_{10}	12	102	14	< 35	< 10
Waste after column-H_{20}	13	112	13	< 35	60
Waste after column-H_{40}	< 10	74	8	< 35	< 10

4.4. Conclusion

Two types of leaching tests were carried out in the present study by means both column flow-percolations and standard batch leaching tests (UNI EN 12547-2) in view of assessing their significance in predicting the landfill sustainability.

Column leaching tests were performed under different L/S ratios (corresponding to one generation time under different rainfall/landfill conditions) and irrigation frequencies to assess their influence on the estimation of long-term emissions.

The results obtained allowed the following conclusions to be drawn:

- Eluates quality from standard batch leaching tests on raw waste demonstrated the waste sample used in the present study meets the limits for non-hazardous landfills. However, they are resulted to be not reliable for assessing the achievement of sustainability targets, being FSQ limits not always achieved in column leaching tests.
- The cumulative contaminants extraction from column leaching tests normalized to the maximum potential emissions (provided by the standard batch leaching test on raw waste) for all parameters under all testing conditions reached a plateau at values between 0.2-0.8, suggesting that the emission potential estimated by means of batch tests is overestimating the real emissions occurring under landfill conditions.
- Irrigation frequency influenced both organics and inorganics variation only under low water input conditions (L tests): higher the irrigation frequency, higher the flushing effects and faster the concentration variation. Conversely, no significant influence was detected under high water input conditions (H tests).
- Irrigation frequencies had a significant impact on the reduction of waste emission potential, and it could be taken into account to improve the in-situ treatments.

References

Alam, Q., Florea, M.V.A., Schollbach, K., Brouwers, H.J.H., 2017. A two-stage treatment for municipal solid waste incineration (MSWI) bottom ash to remove agglomerated fine particles and leachable contaminants. *Waste Management.* 67, 181–192. https://doi.org/10.1016/j.wasman.2017.05.029

Cossu, 2016. Back to Earth Sites: From "nasty and unsightly" landfilling to final sink and geological repository. *Waste Management.* https://doi.org/10.1016/j.wasman.2016.07.028

Cossu, R., Grossule, V., Lavagnolo, M.C., 2020. What about residues from circular economy and role of landfilling? *Detritus.* https://doi.org/10.31025/2611-4135/2020.13920

Cossu, R., Lai, T., 2013. Washing treatment of automotive shredder residue (ASR). *Waste Management.* 33, 1770–1775. https://doi.org/10.1016/j.wasman.2013.04.007

Cossu, R., Lai, T., 2012. Washing of waste prior to landfilling. *Waste Management* 32, 869–878. https://doi.org/10.1016/j.wasman.2011.12.005

Cossu, R., Lai, T., Pivnenko, K., 2012. Waste washing pre-treatment of municipal and special waste. *Journal of Hazardous Materials.* 207–208, 65–72. https://doi.org/10.1016/J.JHAZMAT.2011.07.121

Cossu, R., Stegmann, R., 2018. Solid Waste Landfilling: Concepts, Processes, Technology. Elsevier.

di Gianfilippo, M., Costa, G., Verginelli, I., Gavasci, R., Lombardi, F., 2016. Analysis and interpretation of the leaching behaviour of waste thermal treatment bottom ash by batch and column tests. *Waste Management* 56, 216–228. https://doi.org/10.1016/j.wasman.2016.07.034

Ettler, V., Johan, Z., 2014. 12years of leaching of contaminants from Pb smelter slags: Geochemical/mineralogical controls and slag recycling potential. *Applied Geochemistry* 40, 97–103. https://doi.org/10.1016/j.apgeochem.2013.11.001

Freyssinet, P., Piantone, P., Azaroual, M., Itard, Y., Clozel-Leloup, B., Guyonnet, D., Baubron, J.C., Baubron. 2002. Chemical changes and leachate mass balance of municipal solid waste bottom ash submitted to weathering. *Waste Management* 22 (2002) 159–172.

Galvín, A.P., Ayuso, J., Jiménez, J.R., Agrela, F., 2012. Comparison of batch leaching tests and influence of pH on the release of metals from construction and demolition wastes. *Waste Management* 32, 88–95. https://doi.org/10.1016/j.wasman.2011.09.010

Grathwohl, P., 2014. On equilibration of pore water in column leaching tests. *Waste Management* 34, 908–918. https://doi.org/10.1016/j.wasman.2014.02.012

Grathwohl, P., Susset, B., 2009. Comparison of percolation to batch and sequential leaching tests: Theory and data. *Waste Management* 29, 2681–2688. https://doi.org/10.1016/j.wasman.2009.05.016

Grossule, V., 2020. Final quality of a sustainable landfill and postclosure management. *Detritus* 13, 148–159. https://doi.org/10.31025/2611-4135/2020.13999

Grossule, V., Stegmann, R., 2020. Problems in traditional landfilling and proposals for solutions based on sustainability. *Detritus* 12, 78–91. https://doi.org/10.31025/2611-4135/2020.14000

Gupta, N., Gedam, V. v., Moghe, C., Labhasetwar, P., 2019. Comparative assessment of batch and column leaching studies for heavy metals release from coal fly ash bricks and clay bricks. *Environ Technol Innov* 16. https://doi.org/10.1016/j.eti.2019.100461

Jiao, F., Zhang, L., Dong, Z., Namioka, T., Yamada, N., Ninomiya, Y., 2016. Study on the species of heavy metals in MSW incineration fly ash and their leaching behavior. *Fuel Processing Technology* 152, 108–115. https://doi.org/10.1016/j.fuproc.2016.06.013

Kjeldsen, P., Barlaz, M.A., Rooker, A.P., Baun, A., Ledin, A., Christensen, T.H., 2002. Present and long-term composition of MSW landfill leachate: A review. *Crit Rev Environ Sci Technol.* https://doi.org/10.1080/10643380290813462

López Meza, S., Kalbe, U., Berger, W., Simon, F.G., 2010. Effect of contact time on the release of contaminants from granular waste materials during column leaching experiments. *Waste Management* 30, 565–571. https://doi.org/10.1016/j.wasman.2009.11.022

Luo, H., Cheng, Y., He, D., Yang, E.H., 2019. Review of leaching behavior of municipal solid waste incineration (MSWI) ash. *Science of the Total Environment*. https://doi.org/10.1016/j.scitotenv.2019.03.004

Mantis, I., Voutsa, D., Samara, C., 2005. Assessment of the environmental hazard from municipal and industrial wastewater treatment sludge by employing chemical and biological methods. *Ecotoxicol Environ Safety* 62, 397–407. https://doi.org/10.1016/j.ecoenv.2004.12.010

Maszkowska, J., Kołodziejska, M., Białk-Bielińska, A., Mrozik, W., Kumirska, J., Stepnowski, P., Palavinskas, R., Krüger, O., Kalbe, U., 2013. Column and batch tests of sulfonamide leaching from different types of soil. *Journal of Hazardous Materials* 260, 468–474. https://doi.org/10.1016/j.jhazmat.2013.05.053

Olgun, A., Atar, N., Wang, S., 2013. Batch and column studies of phosphate and nitrate adsorption on waste solids containing boron impurity. *Chemical Engineering Journal* 222, 108–119. https://doi.org/10.1016/j.cej.2013.02.029

Parodi, A., Feuillade-Cathalifaud, G., Pallier, V., Mansour, A.A., 2011. Optimization of municipal solid waste leaching test procedure: Assessment of the part of hydrosoluble organic compounds. *Journal of Hazardous Materials* 186, 991–998. https://doi.org/10.1016/j.jhazmat.2010.11.090

Pecorini, I., Baldi, F., Bacchi, D., Carnevale, E.A., Corti, A., 2017. Leaching behaviour of hazardous waste under the impact of different ambient conditions. *Waste Management* 63, 96–106. https://doi.org/10.1016/j.wasman.2016.10.037

Sazali, Norsuhailizah, Harun, Z., Sazali, Norazlianie, 2020. A review on batch and column adsorption of various adsorbent towards the removal of heavy metal. Journal of Advanced Research in Fluid Mechanics and Thermal Sciences 67, 66–88.

Tiwari, M.K., Bajpai, S., Dewangan, U.K., Tamrakar, R.K., 2015. Suitability of leaching test methods for fly ash and slag: A review. *Journal of Radiation Research and Applied Sciences* 8, 523–537. https://doi.org/10.1016/j.jrras.2015.06.003

5. General Conclusions

This thesis work was developed based on the concept of sustainable waste management. All efforts poured into this work mainly focus on achieving sustainable waste management and accomplishing the target of closing the material loop in the Circular Economy (CE).

Sustainability principle has been applied to guide the holistic waste management system due to the increasing need for resources. CE particularly highlights recycling materials from waste and reducing the consumption of raw materials, which has provided a global strategy for current solid waste management. However, the missing part in the concept of CE could not truly accomplish the closure of materials loop: landfilling disposal of non-renewable residues.

Although according to moral, ideological, and absolute principles, landfilling is regarded as an "bad, obsoleted" solution to waste management and ranks at the bottom of the waste hierarchy (Cossu, 2009., Cossu, et al., 2020), landfilling plays a fundamental role either depositing the increasing non-renewable residues in a low-cost way either offering the opportunity to recover resources in closing the loop of materials. therefore, landfills as the final sink storing non-recycled residues should be built with the aim of reducing diffusion of contaminates and preventing the long-term potential emission, in order to achieve the target to ascertain environmental safety and close materials loop in a sustainable way with respect to environment, economy as well as technology.

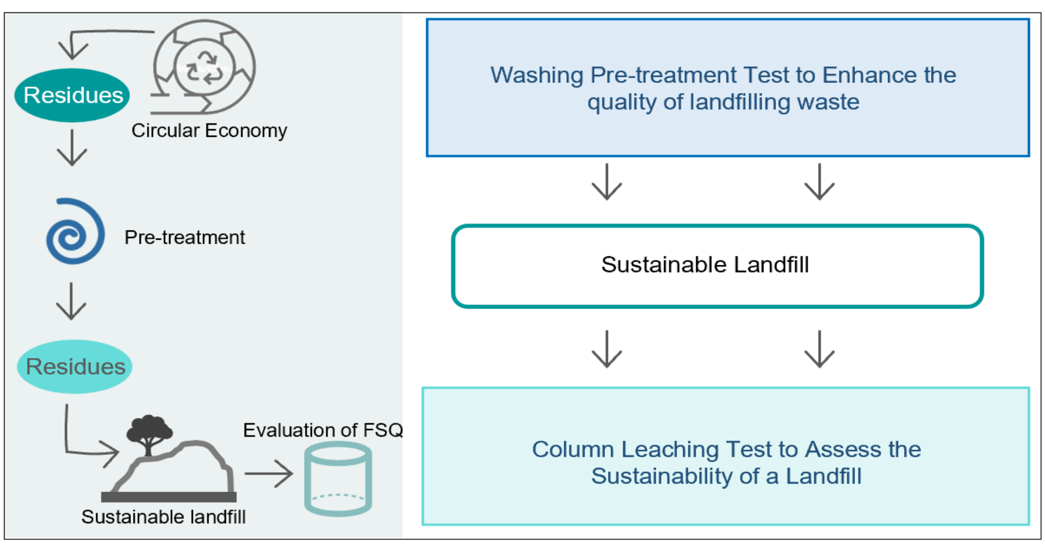

Figure 5. 1 The scheme of the holistic research plan.

A sustainable landfill relies on both pre-treatments and in situ treatments to stabilise the residual waste and immobilise the contaminants, achieving the FSQ within one generation. For those purposes, the thesis study developed a systematic work particularly focused on stabilising the buried residues to shorten the time achieving environmental equilibrium and assessing the sustainability of a landfill.

To stabilize landfilling residues and immobilize potential contaminants, a lab-scale waste washing pre-treatment was applied. Three types of residues were performed with washing procedures and were demonstrated with a significant reduction in contaminants, particularly in inorganic contaminants, confirming the effectiveness of the washing pre-treatment particularly in case of residues with high leachable inorganic contaminants. Simulation in column tests showed that time required to reach COD and ammonia FSQ limits was always shorter in case of washed residues compared to the raw ones, significantly reducing the long-term emissions. However, only the sample RP (after composting) was the only one respecting both COD and ammonia FSQ limits within 30 years. RB and RC samples of high contamination of not stabilised organics could not achieve FSQ within 30 years. Washing of waste proves a new strategy in waste management by controlling and reducing long-term emissions, as well as shortening the time to reach environmental equilibrium. However, to achieve a high washing efficiency and effective contaminant removal, biological pre-treatments or in situ treatment should co-process with the washing pre-treatment for removing unstable organic substances from the target waste. Besides, under lab scale testing conditions, both washing efficacy and landfill operation are optimised by the shredding of the waste material and by the small, controlled landfill simulation. When the process is scaled up, both washing efficacy and long-term impacts should be evaluated under pilot scale conditions of by considering a proper safety factor.

Based on obtained results achieving the FSQ by waste washing pre-treatment research, the significance of both column flow-percolations and standard batch leaching tests in view of assessing the landfill sustainability was investigated subsequently. Investigation results found that values from standard batch leaching tests could measure the limits of landfilling but are not enough to assess the sustainability of a landfill and overestimated the real emissions occurring under landfill conditions. Furthermore, results from column leaching tests demonstrated that irrigation frequency influenced both organics and inorganics variation only under low water input conditions instead of high water input.

Assessing the sustainability of landfilling by performing column leaching tests provides an optional and reliable method to tell the "landfilling truth" that batch leaching of short-time and high L/S could not reveal. The investigation related to irrigation frequency and quantity would also give a possible suggestion for in-situ treatment of landfill. Furthermore, the phenomenon of ammonia concentrations of high water input groups (H tests) approaching but could not achieve FSQ also provides new eyesight in waste management in tropic areas. Proper management of the water input quantities or frequencies should take into account to improve the sustainability of landfills within one generation.

References

Cossu, R. (2009). From triangles to cycles. *Waste Management*, 29(12), 2915–2917. https://doi.org/10.1016/j.wasman.2009.09.002

Cossu, R., Grossule, V., & Lavagnolo, M. C. (2020). What about residues from circular economy and role of landfilling? In *Detritus* (Vol. 9, Issue March, pp. 1–3). Cisa Publisher - Eurowaste Srl. https://doi.org/10.31025/2611-4135/2020.13920

Appendix

Experimental Equipment

The following equipment was applied either for experiments or analysis of samples.

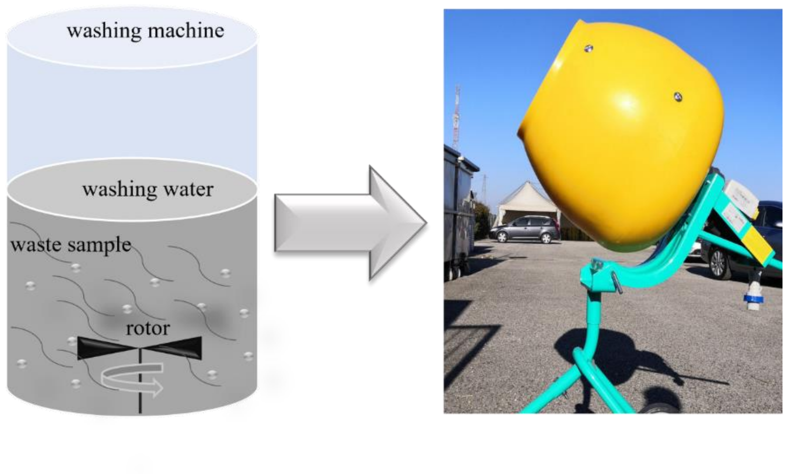

Figure appx. 1 Washing pre-treatment equipment, and washing process was continuously conducted at a speed of 22 rpm at room temperature.

Figure appx. 2. Apparatus for standard batch leaching test (UNI EN 12547-2: L/S=10, duration 24 h)

Figure appx. 3. The different sizes of sieves for trade composition and samples separated from the sieve.

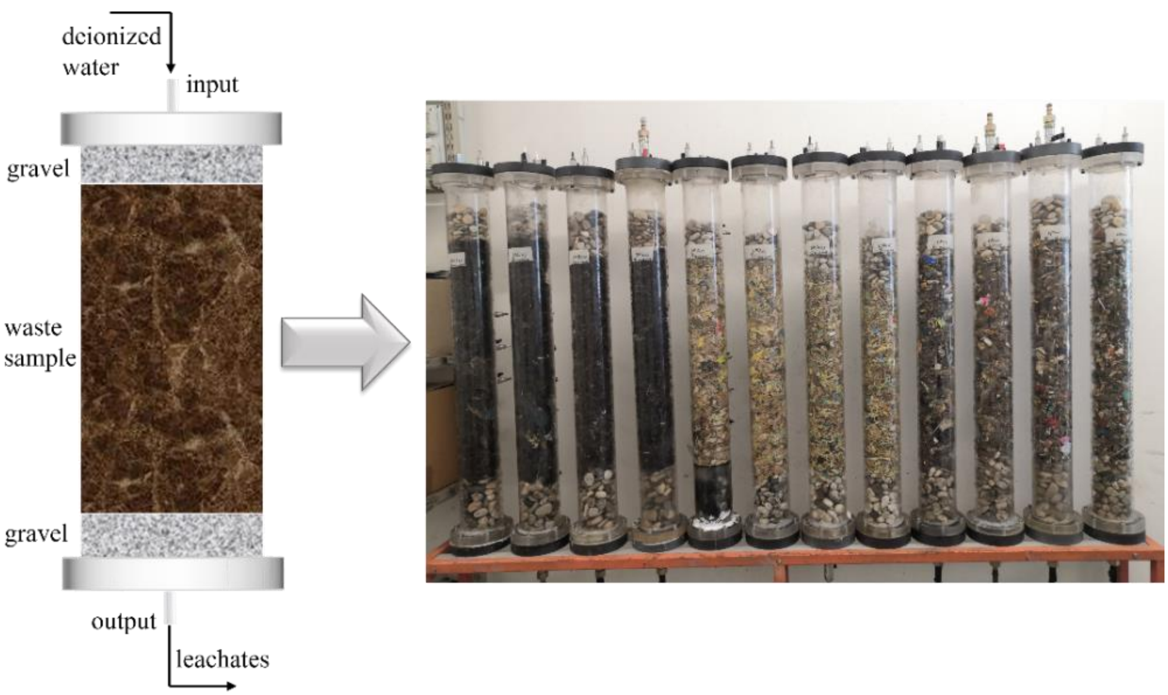

Figure appx. 4. Column test apparatus

Experimental Samples

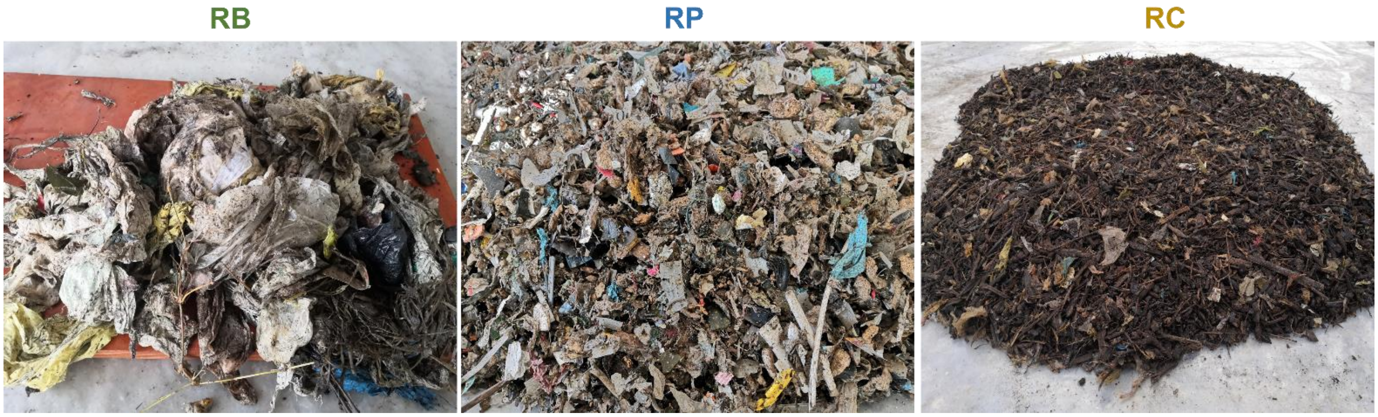

Figure appx. 5. Raw waste materials for waste washing experiments

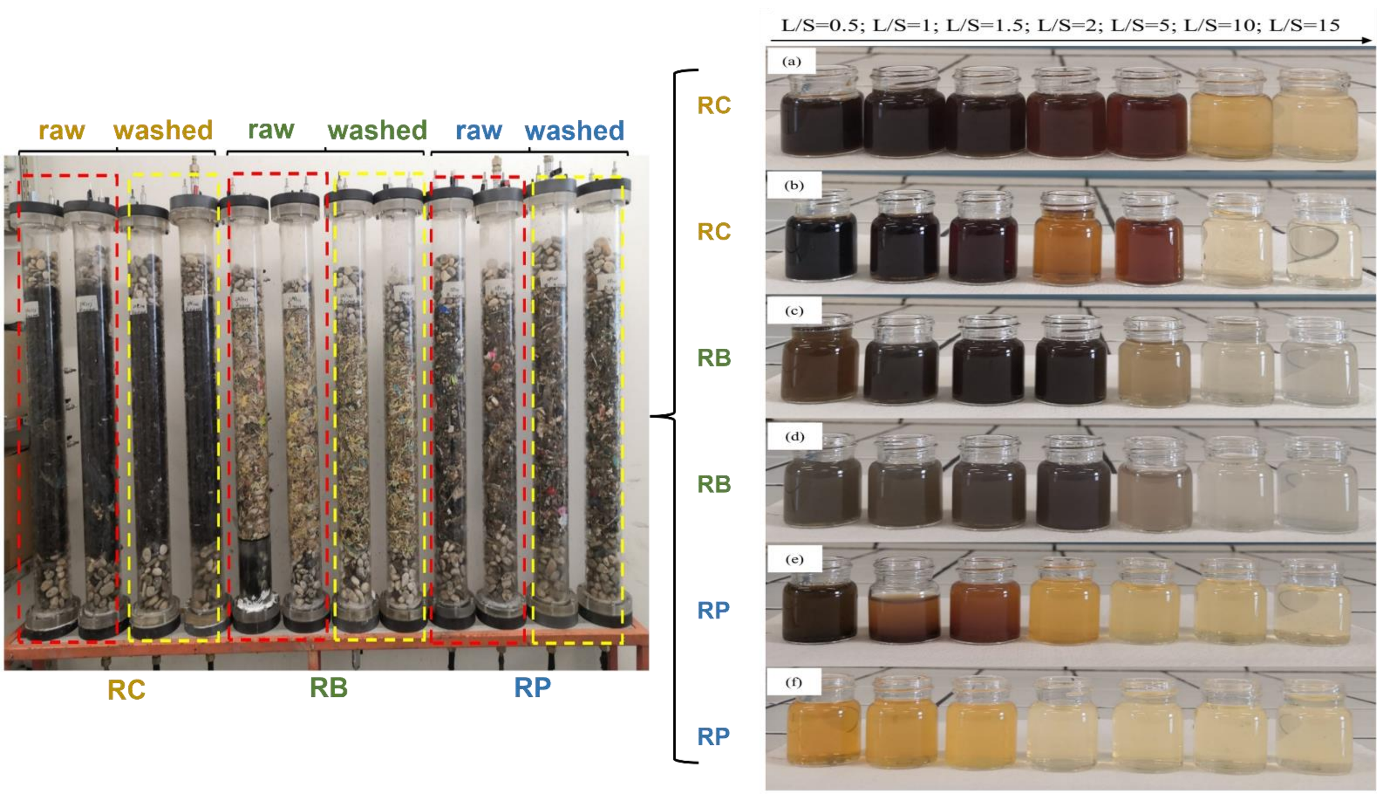

Figure appx. 6. Leachates collected at different L/S ratios from column tests carried out on both washed and unwashed residues.

Figure appx. 7. Waste sample collected from the old landfill (over 18 years)

Figure appx. 8. Leachates collected from column tests carried out on old-age residues.

Compositions of Waste Samples

Table appx. 1. Detailed compositions of RB waste (kg).

	sieve 100 mm			sieve 50 mm			sieve 20 mm			< 20 mm			Total amount
	1	2	**tot**	1	2	**tot**	1	2	**tot**	1	2	**tot**	
	kg	kg	**kg**	kg	kg	**kg**	kg	kg	**kg**	kg	kg	**kg**	**kg**
Paper	0.14	-	**0.14**	3.2	1.6	**4.8**	8.3	2.4	**10.7**	-	-	**-**	**15.2**
Glass	-	-	**-**	-	-	**-**	-	-	**-**	-	-	**-**	**-**
Putrescible	-	-	**-**	2.83	0.87	**3.7**	6.54	2.92	**9.46**	-	-	**-**	**13.2**
Plastics	7.7	2.52	**10.23**	5.5	1.7	**7.21**	2.17	0.59	**2.76**	-	-	**-**	**20.2**
Metals	-	-	**-**	0.03	0.05	**0.08**	-	-	**-**	-	-	**-**	**0.08**
Wood	0.34	0.1	**0.44**	0.18	0.09	**0.27**	0.08	0.11	**0.19**	-	-	**-**	**0.9**
Textiles	0.4	-	**0.37**	0.84	0.23	**1.07**	0.23	-	**0.23**	-	-	**-**	**1.67**
Undersieve < 20 mm	-	-	**-**	-	-	**-**	-	-	**-**	6.83	2.06	**8.89**	**8.89**
Total	-	-	**11.18**	-	-	**17.12**	-	-	**23.32**	-	-	**8.89**	**60.51**

Table appx. 2. Detailed compositions of RC waste (kg).

	sieve 100 mm			sieve 40 mm			sieve 20 mm			< 20 mm			Total amount
	1	2	**tot**	1	2	**tot**	1	2	**tot**	1	2	**tot**	
	kg	kg	**kg**	kg	kg	**kg**	kg	kg	**kg**	kg	kg	**kg**	**kg**
Paper	-	-	**-**	-	-	**-**	-	-	**-**	-	-	**-**	**-**
Glass	-	-	**-**	-	-	**-**	-	-	**-**	-	-	**-**	**-**
Wood	-	-	**-**	0.51	0.41	**0.92**	1.91	1.16	**3.07**	-	-	**-**	**3.99**
Plastics	-	-	**-**	0.16	0.12	**0.28**	0.16	0.11	**0.27**	-	-	**-**	**0.55**
Metals	-	-	**-**	-	-	**-**	0.03	0.04	**0.07**	-	-	**-**	**0.07**
Inerts	-	-	**-**	-	-	**-**	0.01	0.01	**0.02**	-	-	**-**	**0.02**
Textiles	-	-	**-**	0.02	0.04	**0.06**	0.02	0.02	**0.04**	-	-	**-**	**0.1**
Others	-	-	**-**	-	0.08	**0.08**	0.16	0.06	**0.22**	-	-	**-**	**0.3**
Undersieve < 20 mm	-	-	**-**	-	-	**-**	-	-	**-**	15.95	17.08	**33.03**	**33.03**
Total	-	-	**-**	-	-	**1.34**	-	-	**3.69**	-	-	**33.03**	**38.06**

Table appx. 3. Detailed compositions of RP waste (kg).

	sieve 40 mm				sieve 20 mm					< 20 mm				Total amount
	1	2	3	**tot**	1	2	3	4	**tot**	1	2	3	**tot**	
	kg	kg	kg	**kg**	kg	kg	kg	kg	**kg**	kg	kg	kg	kg	**kg**
Paper	0.02	-	-	**0.02**	0.03	0.01	0.01	-	**0.05**	-	-	-	-	**0.07**
Glass	-	-	-	**0**	0.3	0.11	0.15	0.14	**0.7**	-	-	-	-	**0.7**
Textile	0.02	-	-	**0.02**	0.01	0.02	-	-	**0.03**	-	-	-	-	**0.05**
Plastics	0.32	0.02	0.05	**0.39**	0.9	0.98	0.43	0.34	**2.65**	-	-	-	-	**3.04**
Metals	0.15	-	-	**0.15**	0.21	0.13	0.05	0.04	**0.43**	-	-	-	-	**0.58**
Inerts	-	-	-	**0**	0.02	0.01	0.01	0.01	**0.05**	-	-	-	-	**0.05**
Wood	0.01	0.05		**0.06**	0.3	0.01	-	-	**0.31**	-	-	-	-	**0.37**
undersieve < 20 mm	-	-	-	-	-	-	-	-	-	5.62	17.2	3.59	26.4	**26.4**
Total				**0.64**					**4.22**	-	-	-	26.4	**31.26**

Table appx. 4. Detailed compositions of residues from the old landfill (kg) (NB: the total weight has minus the weight of containers used for balancing the weight of samples)

	Sieve 100 mm				Sieve 80 mm				Sieve 50 mm				Sieve 20 mm				< 20 mm				Total
	1	2	3	**tot**	1	2	3	**tot**	1	2	3	**tot**	1	2	3	**tot**	1	2	3	**tot**	
	kg	kg	kg	**kg**	kg	kg	kg	**kg**	kg	kg	kg	**kg**	kg	kg	kg	**kg**	kg	kg	kg	**kg**	**kg**
Plastics	4.66	8.79	4.65	**14.44**	3.64	4.79	2.79	**7.56**	3	3.68	3.22	**6.24**	4.81	5.26	4.1	**10.51**	-	-	-	**-**	**61.95**
Paper	-	0.95	0.87	**0.78**	0.82	0.77	0.55	**0.58**	0.55	0.77	0.55	**0.31**	0.67	0.77	0.73	**0.61**	-	-	-	**-**	**33.28**
Metals	1.35	0.9	-	**1.21**	1.6	0.64	1.03	**1.71**	1.1	0.69	1.35	**1.58**	0.67	1.07	0.67	**0.85**	-	-	-	**-**	**28.46**
Inerts	2.26		1.58	**2.8**	1.25	3.5	1.87	**5.06**	4.24	5.39	5.47	**13.54**	10.23	9.86	11.25	**29.78**	-	-	-	**-**	**73.45**
Textiles	2.14	3.38	-	**4.48**	1.6	0.96	1.55	**2.55**	1.65	1.24	1.44	**2.77**	10.04	0.86	1.09	**10.43**	-	-	-	**-**	**33.82**
Undersieve < 20 mm																	32.04	40.33	40.8	**113.17**	
Total	**7.63**	**11.24**	**4.84**	**23.71**	**5.61**	**7.36**	**4.49**	**17.46**	**7.24**	**8.47**	**8.73**	**24.44**	**23.12**	**14.52**	**14.54**	**52.18**	**32.04**	**40.33**	**40.8**	**113.17**	**230.96**

Operations for Physical and Chemical Analysis

Total Solid (TS) and Volatile Solids (VS) Analysis

Total solid (TS) analysis was performed for determining the net dry mass of waste samples. Each waste sample was placed in crucible and was weighted by using an analytical balance, and then was dried in the oven at the temperate of 105 ˚C for 12 hours. After drying, waste samples were weighted by using analytical balance, and TS of samples was caculated by the following equation 12. Volatile solid (VS) analysis was perfermed with the dry waste samples gained from TS analysis, as shown in **Figure appx. 9**. Waste samples were placed in the muffle furnace at the temperate of 550℃ for 3 h. When the temperature of the samples in the muffle furnace dropped to room temperature, VS was calculated by equation 13. Each sample was prepared in triplicates for both TS and VS analysis.

The equation used to determine the TS in a solid sample (IRSA-CNR Q.64/84 vol.2 n°2):

$$TS\ [\%] = \frac{W_{105} - W_c}{W_s} \cdot 100 \qquad (12)$$

Where W_{105} = weight of the sample and of the container after 12 h at 105 °C (g); Wc = weight of the container (g); Ws = weight of the sample analysed (g).

Equation used to calculate the VS in a solid sample (IRSA-CNR Q.64/84 vol.2 n°2):

$$VS\ [\%] = \frac{W_{105} - W_{550}}{W_{105} - W_c} \cdot 100 \qquad (13)$$

Where W_{105} = weight of the sample and of the container after 12 h at 105 °C (g); W_{550} = weight of the sample and the container after 3 h at 550 °C (g); Wc = weight of the container (g).

Figure appx. 9. Total solid and volatile solid analysis.

Dissolved Organic Carbon (DOC)

Dissolved organic carbon (DOC) defined as the organic matter that is able to pass through a 0.22 µm filter. DOC concentrations in the eluates derived from the leaching tests of the raw and washed waste samples and in the washing waters was measured by the method of IRSA-CNR 29/2003 vol.2 n°5130.

Respirometric Index (RI_7)

The seven-days respirometric index (RI_7) was determined by means of the Sapromat apparatus (**Figure appx.10**). According to the procedure followed, the samples preparation required a moisture content in each sample equal or higher than 50%; for this reason, depending on the TS content in the sample, the respective amount of water to be added was calculated according to the following equation:

$$V_{H2O} = W_s \cdot \frac{M_f - M_i}{1 - M_f} \qquad (14)$$

Where V_{H2O} = volume of water to be added to the sample in order to reach a moisture content equal to 50%; W_s = weight of the sample (g); M_f = final moisture content of the sample (%); M_i = initial moisture content of the sample (%).

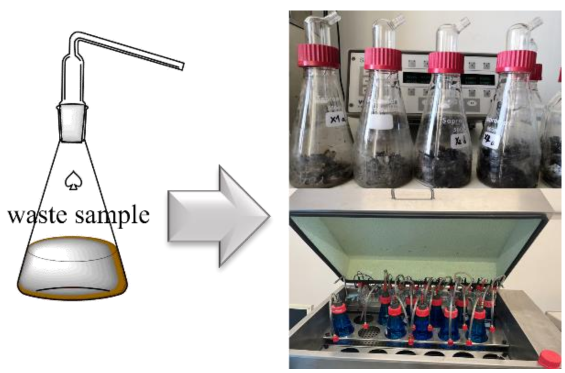

Figure appx. 10. RI_7 samples preparation.

Total Kjeldahl Nitrogen (TKN)

Waste samples were analyzed by total Kjeldahl nitrogen (TKN) to determine both the organic and the inorganic forms of nitrogen. The analysis started with concentrated sulfuric acid digestion of samples organics, converting organic nitrogen to ammonia. It requires that samples boiled in concentrated sulfuric acid, potassium sulfate, and a copper catalyst to convert the organic nitrogen to ammonia, the procedure as shown in **Figure appx.11**. The samples collected from distillation procedure were titrated with boric acid to determine the ammonia. TKN content in liquid samples was calculated by means of an automatic titration (IRSA-CNR 29/2003, vol.2 n°5030) according to the following equation:

$$TKN\ [mg_N/L] = \frac{V_{tit} \cdot N_{tit} \cdot 14000}{V_S} \qquad (15)$$

Where, as for the ammonia nitrogen calculation, V_{tit} = volume of the titrant solution used (ml); N_{tit} = normality of the titrant solution (N); *14000* = equivalent weight of nitrogen; V_S = volume of the sample analysed (ml).W

In order to determine the TKN content in solid samples, the following equation was used (IRSA-CNR Q.64/85, vol.3 n°6 mod.):

$$TKN\ [mg_N/kg] = \frac{V_{tit} \cdot N_{tit} \cdot 14000}{W_S} \qquad (16)$$

Where V_{tit} = volume of the titrant solution used (ml); N_{tit} = normality of the titrant solution (N); *14000* = equivalent weight of nitrogen; W_S = weight of the sample analysed (g).

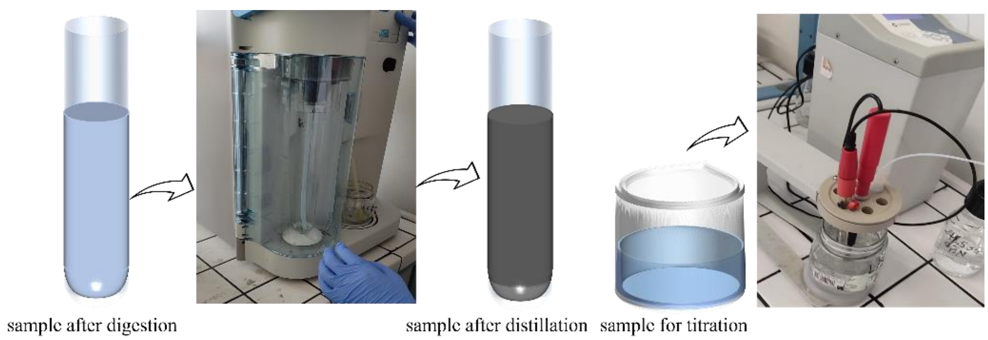

Figure appx. 11. TKN analysis procedures.

Total Organic Carbon (TOC)

Total amount of carbon in organic compounds of both solid and liquid waste samples was measured by total organic carbon (TOC) analysis. The measurement was completed with the specific equipment (Shimadzu TOC-VCSN analyzer) seen in **Figure appx.12**.

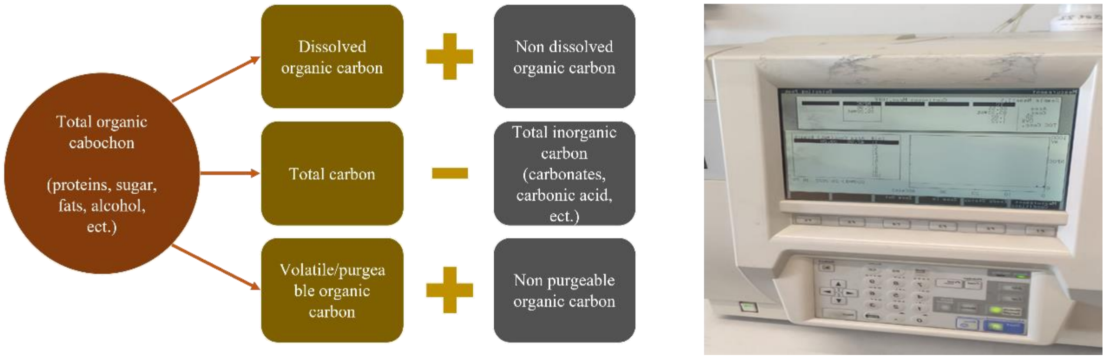

Figure appx. 12. TOC measurement and the equipement.

Chemical Oxygen Demand (COD)

Chemical Oxygen Demand (COD) analysis was applied to determine the oxygen-depletion capacity of liquid samples contaminated with organic waste matter. Liquid samples obtained from leaching eluates of raw and washed residues were measured with the wet chemistry method (IRSA-CNR 29/2003 vol.2 n°5130). The analysis procedure involved a two-hour digestion at high heat under acidic conditions in which potassium dichromate acts as the oxidant for any organic material present in a water sample. Silver sulfate was present as the catalyst and mercuric sulfate acted to complex out any interfering chloride. Following the digestion, the extent of oxidation was measured through indirect measurement of oxygen demand via electrons consumed in the reduction of Cr^{6+} to Cr^{3+}. The prepared samples finally were done by titration or spectrophotometry. And all the processes could be seen in **Figure appx.13**.

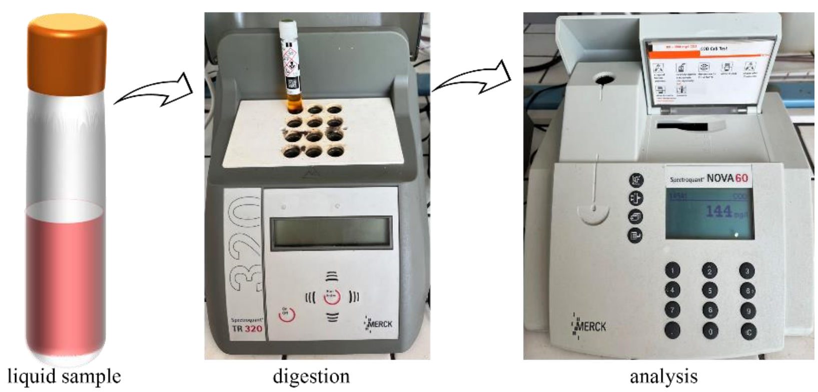

Figure appx. 13. COD analysis devices.

Biochemical Oxygen Demand (BOD_5)

The amount of organic pollutants in the samples was determined by the test for biochemical oxygen demand (BOD), while in this study, the test of BOD_5 with the duration of 5 days was used for analysing eluates from raw and washed waste batch leaching tests, measuring the oxygen consumed by bacteria from the decomposition of organic matter. The procedure followed was the IRSA-CNR 29/2003 vol.2, n° 5120A.

Chlorides (Cl^-)

Chlorides (Cl^-) of samples were detected by the method of IRSA-CNR 29/2003, vol.2 n°4090B. When a few drops of a silver nitrate solution were added to a slightly acidic aqueous solution that contains chloride ions, a white precipitate of silver chloride will form, while in this study, an automatic titration is used to measure the amount of Cl^- (**Figure appx.14**). The equation used in order to determine the content of Cl^- in liquid samples by means of the following:

$$Cl^- \, [mg_{Cl}/L] = \frac{V_{tit} \cdot N_{tit} \cdot 35457}{V_S} \quad (17)$$

Where V_{tit} = volume of the titrant solution used (ml); N_{tit} = normality of the titrant acid (N); 35457 = equivalent weight of chloride; V_S = volume of the sample analysed (ml).

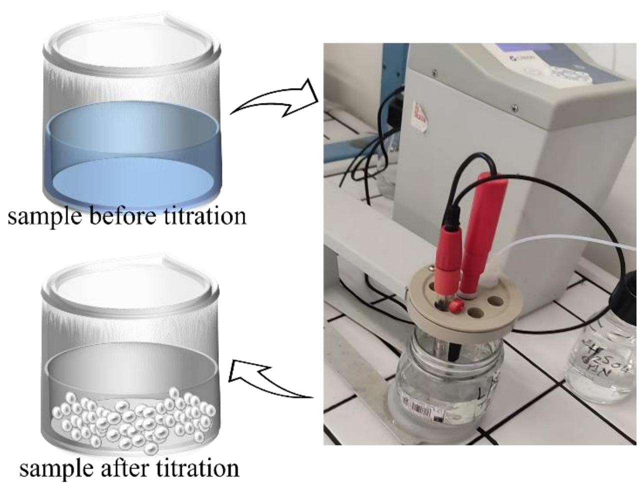

Figure appx. 14. Chlorides analysis.

Heavy Metals determination and pH

The determination of heavy metals in liquid samples was conducted by analysing Cd, Cr, Cu, Fe, Mn, Ni, Pb, and Zn according to the methods of IRSA CNR 29/03 vol. n°3010+2020.

pH was an important parameter of liquid samples and was measured by an automatic machine according to the method of IRSA-CNR 29/2003, VOL 1, N. 2060.

Ammonia Nitrogen (NH_4-N) Analysis

Ammonia nitrogen (NH_4-N) of leachates generated from tests were determined by means of an automatic titration (IRSA-CNR 29/2003, vol.2 n°5030). NH_4-N concentration in the liquid samples was calculated by the following equation:

$$NH_4\text{-}N\ [mg_N/L] = \frac{V_{tit} \cdot N_{tit} \cdot 14000}{V_S} \qquad (18)$$

Where, as for the ammonia nitrogen calculation, V_{tit} = volume of the titrant solution used (ml); N_{tit} = normality of the titrant solution (N); *14000* = equivalent weight of nitrogen; V_S = volume of the sample analysed (ml).

SO_4^{2-} analysis

SO_4^{2-} in liquid samples was determined using the method of IRSA-CNR 29/2003, vol 2, n. 4140_B. Liquid samples were prepared by mixing with 10 ml of glycerin solution and 5 ml of barium chloride solution, and then the mixture was stirred for 2 minutes on a magnetic stirring machine. The absorbance of the sample against the blank at a wavelength of 390 nm was read by an ultraviolet spectrophotometer. All the selected liquid samples should be colourless and non-cloudy in order to avoid the interference of colour.

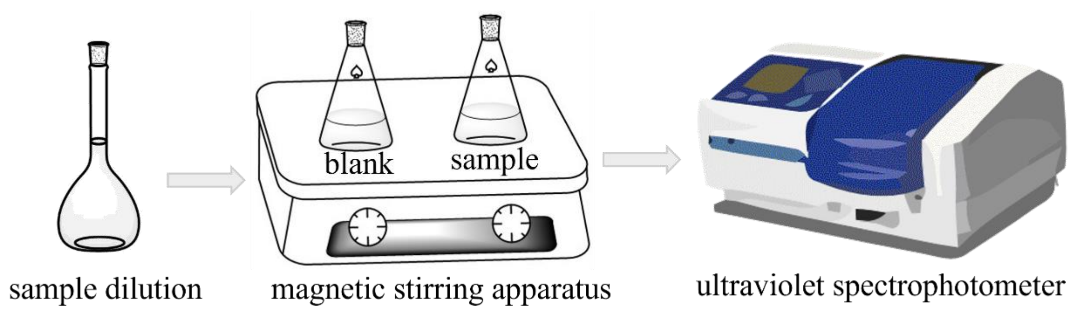

Figure appx. 15. Sample preparation of SO_4^{2-} analysis and apparatus.

Acknowledgements

I would like to express my great gratitude to my primary supervisor, Prof. Maria Cristina Lavagnolo, who guided me throughout my project. I am so grateful for her careful supervision and powerful support for my scientific research as well as my personal growth. In the past three years, Prof. Lavagonlo offered me tons of help not only in my program but also in my life. She plays a role of a patient and professional tutor, and a kind and warm-hearted friend as well.

I would also like to thank my co-supervisor Dr. Valentina Grossule. She gave me a lot of support and helped me to develop my experimental tests, as well as offered me fundamental consultants for the following discussion of the obtained results. She also shared with me lots of useful experiences both in scientific research and daily life. Deserved thanks to the researcher of LISA lab, Annalisa Sandon, for her selfless contribution to the whole laboratory and her patience with all my experimental operations as well as her daily care of me.

A special thank goes to all the LISA Lab family for their warm-hearted support, selfless friendship, generous meals, great complicity and each moment shared with me. I really feel grateful for having such lovely friends: Rachele, Rossanna, Paula, Valentina, Giovanni, and Salwa in LISA lab, who accompanied me and brought me a lot of surprises as well as coloured my life.

Beyond all and mostly, a super thanks to my family, my parents and my sister, thanks for their patience and unique support during my three-year oversea journey.

I really feel lucky and grateful to have the chance to do my fantastic journey in Padova and in Italy. Each moment I experienced on this land will brave me and encourage me to face an unknown future and start my new journey.

致谢

人的一生会因为选择和坚持，而获得可以与即定命运不同的路线，一条按照自己意愿发展的路线。

在我还不足以称得上有经验的人生里，我曾做过很多很多选择。决定出国求学、来意大利帕多瓦大学的 LISA-LAB 攻读博士学位，是我在过去的所有岁月里做得最美好的一次选择。因为在意大利学习生活的这三年，虽说不长，但却让我重新构建了对客观世界的认知以及对自身的了解；即使如今自己的见识依旧浅薄，但在这里的经历却教会了我包容与成长的真谛。

一路走来，算不得一帆风顺，在某种意义上，甚至可以算得上是有些曲折，毕竟没有几个留学生会一来留学就会遭遇诈骗而不得不与对方对簿公堂；而后又经历天灾（新冠肺炎）人祸（小型车祸）以及至亲重病、离世以及三年不得归家等种种突发事件。但，我却依旧觉得我如此幸运又如此被命运眷顾，因为在我记忆里留下来的，尽数都是一回想起来就让人热泪盈眶的美好的事物与经历，还有可爱的人们。

我不知道自己是否算得上所谓的“寒门子弟”，如果出身是在下雨能在路上泥巴洼里捉鱼的农村，从小就要干各种各样农活，还要学会摆摊赚学费、本科硕士都会靠做兼职而不要家里一分补贴就算“寒门”的话，我大概也是能算的。因为以上这些，我不仅经历过，而且还因为太过熟稔，还经常被村里人当成某种教育的素材。但是我却从未因为这个身份而自卑过，一丝一毫也没有。相反，我常常因为有这样的经历而感激，因为经历过此前种种，所以才能更能体会当下美妙的每一秒，才会珍惜和感恩生命里出现的每一份美好，以至于现在的我不会那么容易错过幸福。

非常感谢三年前面试了我还决定录取我的导师 Prof.Lavagnolo，感谢她在这三年间给予我无论是生活还是学习上的诸多帮助；感谢一直与我并肩作战、教会我我良多的 Dr.Grossule，感谢她作为一个前辈、一个朋友、一个小导师给予我的鼓励与帮助；还要感谢一直任劳任怨照顾 LISA-LAB 中每一员的 Annalisa，感谢她作为一个没有亲缘关系的长辈，给予我的温暖和关爱；也要感谢 LISA-LBA 中与我同行、与我共餐、与我并肩的每一个可爱的小伙伴，感谢他们这三年来的包容与照顾，让我渐渐融入这个大家庭、在这里尽可能地无忧无虑完成我的学业。也要感谢这三年来，在帕多瓦、在意大利遇到的朋友们，感谢大家给我勇气和信心，以及带给我无数个热泪盈眶的瞬间。还要感谢千里之外的父母、阿姐与亲友，感谢来自他们的爱与温暖让我时刻感受被惦记、被牵挂的美好。也想要感谢这二十八里生命里出现过的每一个曾带给我欢乐与感动的人，感谢你们让我知晓人类的情感是如此伟大与深不可测。也想，感谢那个一路走来，虽然莽撞但却勇敢的自己，感谢自己的坚持与乐观，让自己看到了更大的世界，遇见了更多的美好。

前路虽不可知，但此刻的自己却有信心前行。因为我知晓，无论将来遇到何等难以攀登的高峰，但以一惯之的努力，不懈怠的人生，微小的积累便就能最终得以改变结果。