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Paradigm shifts in incinerator ash, sediment material recovery and landfill monitoring

Evan K. Paleologos
Professor, Engineering and Project Management Graduate Program, Abu Dhabi University, Abu Dhabi, UAE (Orcid:0000-0002-3582-2288) (corresponding author: evan.paleologos@adu.ac.ae)

Rainer Bunge
Professor, Department of Environmental Engineering, Eastern Switzerland University of Applied Sciences, Rapperswil, Switzerland

Gisela Weibel
Senior Research Assistant, Institute of Geological Sciences, University of Bern, Bern, Switzerland

Claudia Vitone
Associate Professor, Department of Civil, Environmental, Land, Building Engineering and Chemistry – Dicatech, Polytechnic University of Bari, Bari, Italy (Orcid:0000-0001-6529-4167)

Michael Ploetze
Senior Scientist, Department of Civil, Environmental and Geomatic Engineering, ETH Zürich, Zurich, Switzerland (Orcid:0000-0001-9833-4196)

Rossella Petti
Post-doc Researcher, Department of Civil, Environmental, Land, Building Engineering and Chemistry – Dicatech, Polytechnic University of Bari, Bari, Italy (Orcid:0000-0003-0111-3480)

Eugeniusz Koda
Professor, Institute of Civil Engineering, Warsaw University of Life Sciences – SGGW, Warsaw, Poland (Orcid:0000-0002-3895-960X)

Brendan C. O'Kelly
Associate Professor, Department of Civil, Structural and Environmental Engineering, Trinity College Dublin, Dublin, Ireland (Orcid:0000-0002-1343-4428)

Anna Podlasek
Assistant Professor, Institute of Civil Engineering, Warsaw University of Life Sciences – SGGW, Warsaw, Poland (Orcid:0000-0003-0326-5672)

Magdalena Daria Vaverková
Associate Professor, Faculty of AgriSciences, Mendel University in Brno, Brno, Czech Republic; Associate Professor, Institute of Civil Engineering, Warsaw University of Life Sciences – SGGW, Warsaw, Poland (Orcid:0000-0002-2384-6207)

Theo Sarris
Science Leader, Health and Environment Division,Institute of Environmental Science and Research (ESR), Christchurch Science Centre, Christchurch, New Zealand (Orcid:0000-0002-1429-2185)

Venkata Siva Naga Sai Goli
Research Scholar, Department of Civil Engineering, Indian Institute of Technology Bombay, Mumbai, India (Orcid:0000-0003-1916-725X)

Abdel-Mohsen O. Mohamed
Professor, Uberbinder, Inc., Seattle, WA, USA; EX Scientific Consultants, Abu Dhabi, UAE (Orcid:0000-0002-0971-6940)

Alexander Puzrin
Professor, Department of Civil, Environmental and Geomatic Engineering, ETH Zürich, Zurich, Switzerland (Orcid:0000-0002-9566-8841)

Jan Winkler
Assistant Professor, Department of Plant Biology, Faculty of AgriSciences, Mendel University in Brno, Brno, Czech Republic (Orcid:0000-0002-5700-2176)

Maurizio Iler Marchi
Team Leader, GPI Global Product Innovation Italcementi Heidelbergcement Group, Bergamo, Italy; Technical Operations Manager, Innovhub, Milano, Italy

Prithvendra Singh
Research Scholar, Department of Civil Engineering, Indian Institute of Technology Bombay, Mumbai, India

Louise Weaver
Science Leader, Health and Environment Division, Institute of Environmental Science and Research (ESR), Christchurch Science Centre, Christchurch, New Zealand (Orcid:0000-0002-3750-868X)

Devendra N. Singh
D.L. Shah Chair Professor for Innovation, Department of Civil Engineering, Indian Institute of Technology Bombay, Mumbai, India (Orcid:0000-0003-3832-1507)

The current paper discusses some selected developments for efficient management of the globally increasing quantity of waste. Incinerator bottom ash (IBA), the heavier ash generated during incineration of municipal waste, is currently utilised in two distinct ways. One pathway is not to fragment IBA and use it as a building material in road construction. This results in reduced landfilled residual but low metal recovery. The other way is to crush the mineral aggregate, thus maximising metal recovery but resulting in higher landfilled, end material. Here emphasis is placed on the second approach as implemented in Switzerland together with the economics and need for improvement of metal recovery from IBA and fly ash. The second theme reports on the viability of recycling mussel shells as a partial substitute for cement to stabilise dredged marine sediments mechanically. This can reduce the consumption of natural resources and lower the amount of binders used in sediment stabilisation practices. Finally, the adequacy of EU's requirements regarding monitoring of groundwater pollution from landfills is assessed, and recommendations are provided to use bioindicators to determine the impact of landfills on surrounding vegetation.

Keywords: bio-monitoring/dredged sediments/fly ash/heavy metals/incinerator bottom ash/landfills

Introduction

The municipal solid waste (MSW) generated globally amounted to 2.01 billion t annually, with an estimated one-third of it not disposed in an environmentally safe manner. This is expected to increase by almost 70% by 2050, reaching 3.4 billion t, with Sub-Saharan Africa, South Asia, the Middle East and North America doubling their waste generation by that time. Waste management among countries is strongly income dependent, with waste in high- and upper-middle-income countries disposed of in engineered landfills, incinerated (prevalent in land-constrained, high-income countries) or recycled in controlled facilities, whereas lower-income countries rely at 93% on open dumping (Kaza *et al*., 2018).

The 27 Member States of EU in 2018 generated 2337 Mt of waste from all economic activities, with households generating 191.6 Mt (8.2%) of this quantity (Eurostat, 2021). In EU, although the overall landfill rate (waste sent to landfill as a proportion of waste generated) decreased by 3% between 2010 and 2018; depending on the waste category, these rates varied. Household waste and similar waste by small businesses and office buildings disposed of in landfills decreased by 51%, but combustion waste, such as waste from flue gas purification and slags and ashes from waste incineration, saw an increase of 16%. The latter resulted from the 'expansion of combustion capacity in the EU, [and] tighter conditions for the material utilization of combustion residues …' (EEA, 2021). It is critical therefore to analyse some of the practices in the waste incineration sector, in terms of waste generation, together with material recovery during these activities (Paleologos *et al.*, 2018).

During incineration of MSW, the heavier ash generated, which does not rise but drops down, is called 'incinerator bottom ash' (IBA), while the fine particles ash that rise up with flue gases is named 'fly ash' (FA). IBA comprises mainly minerals, metal pieces and unburnt material. Under the conditions of waste incineration, most metals oxidise, if at all, only superficially and then pass through the incinerator largely intact. Consequently, IBA represents a valuable resource for recyclable metals. The recovery of these metals is not only ecologically sensible but, in view of rising metal prices, also interesting economically. The 100 000 t of IBA generated globally every day contains potentially recoverable metals equivalent to about €8 million per day, of which about 50% is indeed realised by way of IBA processing (Holm and Thomé-Kozmiensky, 2018).

In Europe, it is common to recycle IBA as construction material, particularly in road construction (Blasenbauer *et al.*, 2020). For this purpose, IBA is processed to remove the metal pieces. In this case, the processing that is usually carried out is relatively 'material friendly' and aims to avoid the fragmentation of coarse mineral particles, which are much more valuable as construction material than, for example, fine sand. On the other hand, many countries are abandoning the recycling of mineral residues from IBA to focus on maximising metal recovery.

At the same time, although copper (Cu) accumulates to >90% in the IBA, large amounts of zinc (Zn) (60 wt.%), lead (Pb) (50 wt.%) and cadmium (Cd) (90 wt.%) evaporate and are transported in the flue gas, accumulating in the FA. Due to the high concentration of organic and inorganic pollutants, FA has to be disposed under strict regulations and at great costs or has to be treated properly.

Treatment of FA before disposal is essential due to contamination with organic and inorganic pollutants. Stabilisation/solidification (S/S) and thermal methods (vitrification, melting and sintering) are often used to attain environmentally stable materials. Such methods do not reduce the concentration of pollutants but reduce leachability and hence the potential risk to the environment. The presence of elevated alkali chlorides and sulfates and the high concentration of heavy metals in FA lead to technical problems during S/S processes and melting, and therefore, combined methods with a pre-washing steps are often used. Such technologies have economic advantages, but no recovery of materials is possible. The treatment of FA and thus the recovery of heavy metals lead to considerable ecological and economic benefits compared with direct deposition. The sections headed 'Recovery of metals from IBA' and 'Metal recovery from incinerator FA' provide an insightful exposition of the practices in Switzerland for recovering metals from IBA and FA.

Recent studies in Italy and Switzerland have analysed the utilisation of two marine wastes: shells and dredged sediments, which thus far have been landfilled. Mussels account for almost 10% of the global fishery industry and 14% of the value of the aquaculture production. Large quantities of mussel shells are landfilled worldwide, while studies have suggested their utilisation from the cosmetic to the fertiliser industry. At the same time, management of dredged sediments is complex, particularly when these have been contaminated by various sources. The total amount of sediments dredged from marine basins in Europe has reached 200 million m^3/year (SedNet, 2011). This is a critical problem where conflicts arise between port development, conservation of protected ecosystems, tourism and sustainable use of sediment resources within an integrated coastal zone management policy. Novel developments in the utilisation of these marine wastes are explored in the section 'Recent developments in marine waste and biosolid management' together with some practices in the disposal of solid sludge from waste-water-treatment (WWT) plants. Finally, the section headed 'Insights on landfill groundwater monitoring and bio-monitoring' considers some aspects of regulations regarding landfill monitoring and expounds on bioindication, the use of biological indicators as a measure of the environmental effects of landfills on surrounding ecosystems (Koda *et al.*, 2022).

Recovery of metals from IBA

IBA consists of the following main components (Figure 1): 32% mineral fraction >2 mm (single particles of glass, porcelain, tiles, ceramics, stone chippings etc.); 27% slag >2 mm (solidified, partially melted and sintered material); 11% native metals >2 mm (the breakdown of the most important metals is shown in Figure 2); 3% unburnt organic material >2 mm (e.g. books, leather, wood) and 27% grit (fine-grained particles <2 mm, consisting of all the aforementioned materials plus metal oxides, fillers from paper and plastics, wood ash etc.).

The 'mineral fraction' includes all mineral materials >2 mm that are not slag. It consists of about one-third glass and porcelain with a size of mostly 4–64 mm. The glass fraction can vary significantly, depending on how much glass bypasses the waste incineration process through separate municipal collection and direct recycling (Astrup *et al.*, 2016).

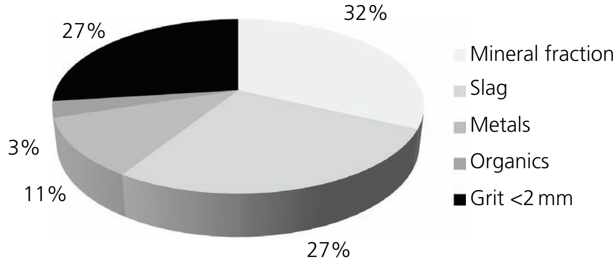

Figure 1. IBA contains some 11% potentially recoverable metal pieces that represent a value of approximately €80/t of IBA (source: Rainer Bunge)

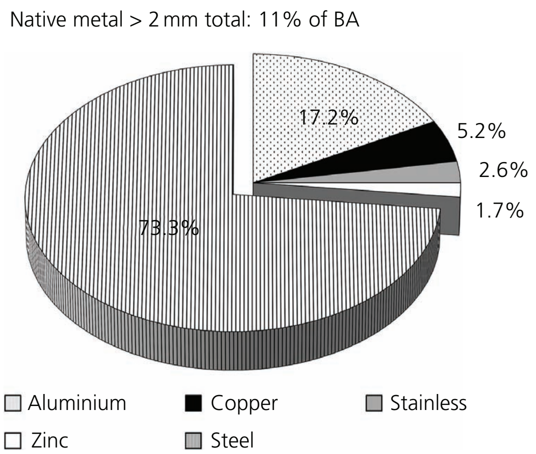

Figure 2. Breakdown of the relative amounts of the most important metals. With the state of the art, most metal pieces with a size exceeding 2 mm are potentially recoverable (source: Rainer Bunge)

Slag particles form in places in the fire bed where temperatures are high enough to exceed the softening temperature of glass. Their formation depends on the calorific value and composition of waste, as well as on combustion conditions. In particular, the amount of air supplied through the grate and the intensity of stoking are important. The slag particles are quite dense, often have a ‘glassy’ appearance, and they display a brown/blackish colour. They are also quite resistant to leaching and contain about 40% iron oxides, which makes them weakly magnetic (Astrup *et al*., 2016).

In terms of typical particle size distribution, the figure by Šyc’s *et al*. (2018) is often quoted, showing a particle size of around 5 mm. The authors of this paper believe that Figure 3 with a mean of about 10 mm, which is based on numerous studies in Switzerland (Bunge, 2019), is more representative of ‘typical’ IBA at least in Europe. The ‘grit’ is material <2 mm (Figure 3) with a composition similar to that of the total IBA in terms of metals, slag and mineral content. In contrast to the coarse-grained fractions, the grit contains about 2% water-soluble substances. As soon as the bottom ash (BA) is exposed to moisture, a multitude

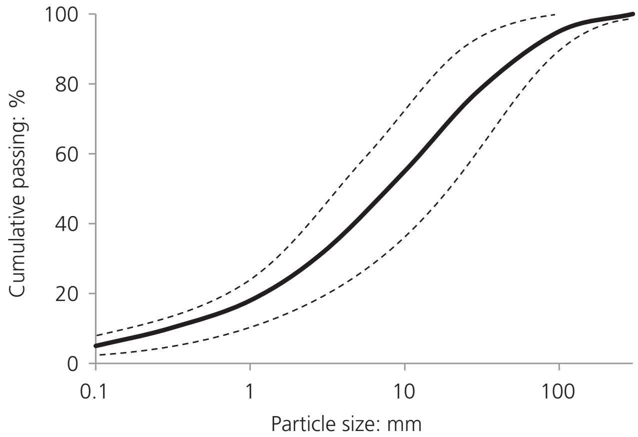

Figure 3. Particle size distribution of IBA. The mean particle size is around 10 mm (Bunge, 2019). The dashed lines represent the boundaries between which the particle size distribution of BA typically lies

of chemical reactions are triggered, as a result of which the IBA sets due to the formation of new minerals. Such minerals are formed not only, for example, by pozzolanic reactions (calcium silicates) but also by precipitation (e.g. gypsum) or by absorption of carbon dioxide (CO_2) from ambient air (limestone). The formation of new mineral phases leads to progressive solidification of the BA. One such reaction is the carbonation of calcium hydroxide ($Ca(OH)_2$), which consumes up to 10 kg carbon dioxide/t BA (approximately 5 m^3 carbon dioxide/t). In the course of several months of curing, the wet BA solidifies into a solid block. During this process, the IBA, mineral fractions and metals are gradually encapsulated by a matrix of solidifying grit with slightly cementitious properties. This is undesirable with regard to the recovery of metals, as they must be liberated later from the encapsulating matrix before they can be separated (Astrup *et al*., 2016; Holm and Thomé-Kozmiensky, 2018).

In Switzerland, which aims to recover metal material rather than using IBA in construction activities, this necessitates that the mineral-encapsulated metal pieces be liberated by crushing the surrounding mineral matrix. Only then can the native metal pieces be separated, by means of magnetic and eddy current separators from the mineral matter. The mineral residue from this processing is very fine grained as a result of the aggressive comminution processes and cannot be utilised as a building material in the form of ‘gravel substitute’. It is thus usually landfilled (Holm and Thomé-Kozmiensky, 2018).

Thus, there are two opposing approaches with regard to IBA treatment (Blasenbauer *et al*., 2020).

- The ‘building material’ variant, based on recycling of the mineral material (EU system). In this case, aggressive comminution is avoided. The metals are merely removed as impurities to recycle the minerals as road construction material. The recovery rate of ferrous pieces >2 mm is about 80% and that of non-ferrous metal pieces >2 mm is about 40%.

- The alternative is ‘maximised metal recovery’, in which the mineral aggregate is crushed to liberate the trapped metal pieces. Consequently, it is no longer usable as a building material and must be landfilled (Swiss system). With this strategy, some 65–95% of the non-ferrous metal pieces larger than 2 mm are recovered from the BA and recycled.

Although there are hardly any two IBA-processing plants in the world with identical flowsheets, many plants are configured somewhat similarly to Figure 4. In these simple plants, the focus is on the building material approach described earlier.

The IBA discharged by means of a wet extractor is technically difficult to treat directly due to the formation of caked layers of moist fines in the machines. The IBA is therefore first stored after wet extraction for some 10–30 days, during which it dries out internally. Some of the moisture is then consumed by the formation of new minerals. Some moisture is also being evaporated due to the high temperatures within IBA heaps that are the result of exothermic processes due to the corrosion of iron (Fe) and aluminium (Al) (Astrup *et al*., 2016; Šyc *et al*., 2018).

In an IBA-processing plant, a first magnetic separation takes place, during which coarse iron particles are removed. Then, screening takes place, for example, at 63 mm. The material >63 mm contains coarse metal pieces and unburnt material that are sorted manually. This is followed by another magnetic

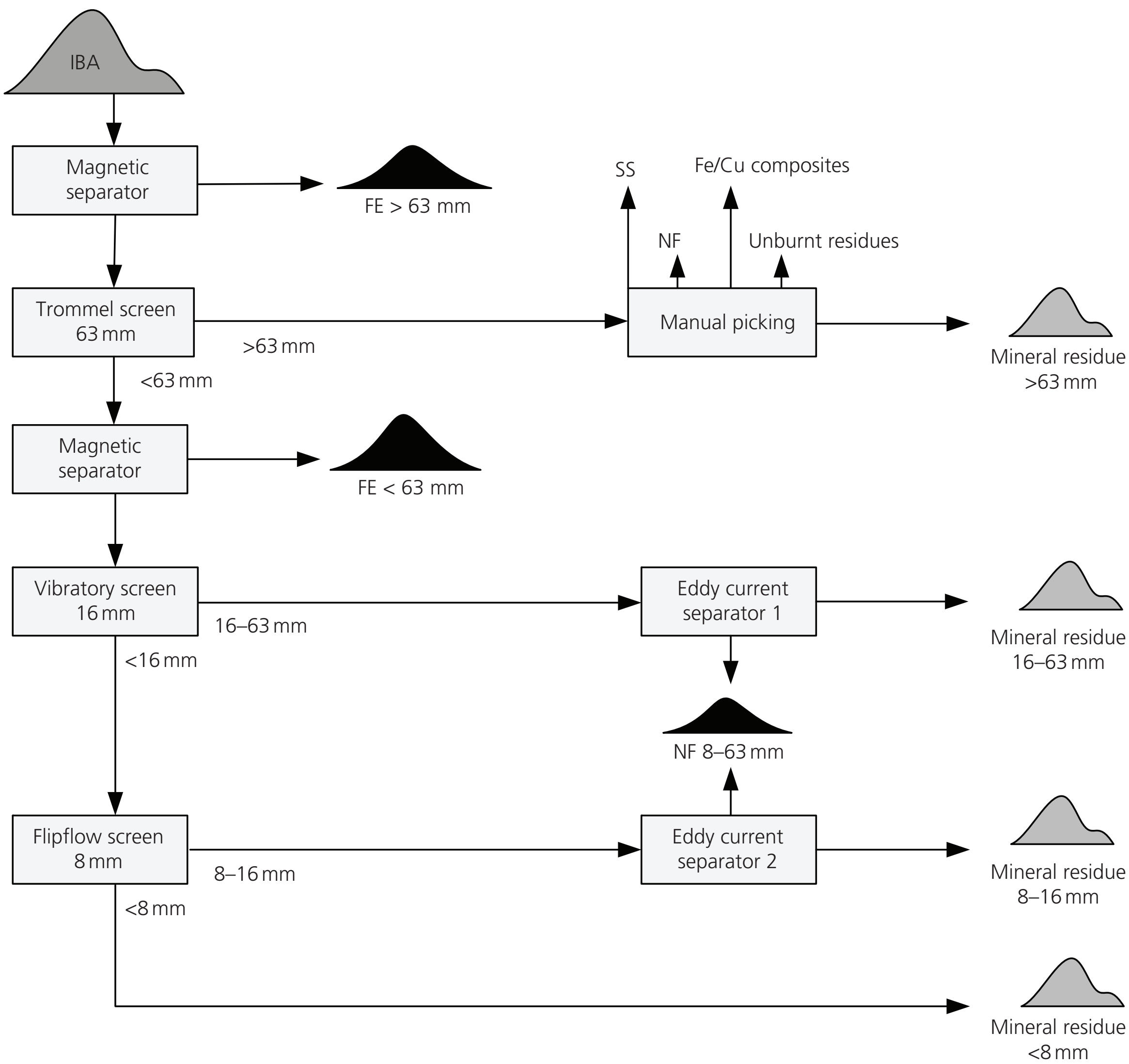

Figure 4. Basic flowsheet of an IBA-processing plant. FE, magnetic steel; NF, non-ferrous metals (aluminium, copper etc.); SS, sewage sludge

separation and another screening process, for example, at 16 mm. Non-ferrous metals are recovered from the coarse material fraction by means of eddy current separation (ECS). The fines are separated by a further screening stage (e.g. at 8 mm), and the coarse material of this screening is freed from non-ferrous metals by means of ECS (Astrup *et al.*, 2016; Šyc *et al.*, 2018).

If the approach of maximised metal recovery is pursued, yields of up to 95% of all metal pieces >2 mm can be achieved and even metal pieces 0.5–2 mm can be recovered (Bunge, 2019). Besides to the basic flowsheet outlined in Figure 4, a plant geared for maximised metal recovery will be furnished with (*a*) a crushing device, usually an impact crusher; (*b*) another train of magnetic and eddy current separators processing the material <8 mm; and (*c*) in some cases, a sensor sorter for the extraction of stainless steel particles.

The cost of processing the IBA by means of the flowsheet outlined earlier is on the order of €12/t (Bunge, 2019). Added to this are transport costs and landfill costs for the material <8 mm. The processed mineral fractions are given away usually free of charge. Depending on the region in Europe, a small remuneration can be obtained. On the other hand, sometimes an additional payment to the recipients of the IBA mineral fraction is required. Revenue is collected from the gate fee for accepting the IBA from the incinerator plus the earnings for the metals sold into the scrap trade. The gate fee is usually on the order of local landfill fees.

If the focus is on maximised metal recovery, and consequently the IBA mineral aggregate needs to be landfilled, the effort for processing of the IBA and disposal of the residual mineral aggregate is significantly higher. This is, however, compensated for by the increased revenues from the recovered metals. Figure 5 shows the revenues for the sale of the metals from a particular Swiss plant. Of course, these revenues depend strongly on current metal prices, which are quite volatile. The high concentration of gold in the fine-grained fractions, which originate mainly from electronic products, is striking.

The metal content in waste increases with the gross domestic product (GDP) of the region in which the waste incinerator operates. This effect is particularly pronounced for copper. The concentration of copper in MSW comes mainly from electronic equipment, and therefore, the copper content of the incinerated waste appears to be a good indicator of the level of development of the region in which an incinerator operates. As the global GDP increases, the amount of incinerated waste will increase. The overall amount of IBA produced will also grow with a consequent rise in the metal content to be recovered. Current trends point in the direction that the recycling of mineral aggregates as building materials will probably be possible only in the future, if they are washed to eliminate soluble pollutants. In the course of wet processing, it will also be possible to use density separation processes, by means of which very small pieces of heavy metals

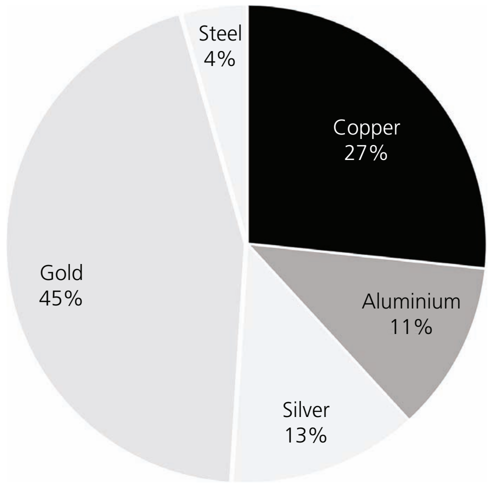

Figure 5. Revenue from the sale of metals extracted by a high-tech IBA-processing plant (based on London Metal Exchange metal prices 2021) (source: Rainer Bunge)

can be recovered very effectively from IBA, particularly copper and gold.

Metal recovery from incinerator FA

About 2% of the MSW input mass accumulates as FA, which consists primarily of aluminosilicates, oxides, soluble salts, heavy metals and toxic organic compounds. Heavy metal fractionation between IBA and FA depends on the composition of MSW, the binding form of the metals and the operating conditions of the incinerator. Metal transfer to FA is favoured by higher furnace temperatures and thus increasing amounts of dust particles as well as elevated chlorine and sulfur concentrations in the flue gas (Jakob *et al.*, 1996; Morf *et al.*, 2000). Ash particles are collected at different stages during the flue gas cleaning process. The empty pass ash (EA) is the first solid residue from the flue gas pass that is often disposed together with IBA. In the further course of the flue gas cleaning process, flue gas passes through the boiler, which is equipped with heat exchanger tubes. By passing through the boiler, the flue gas is cooled from approximately 650 to 250°C. The boiler and later the electrostatic precipitator act as dust removal systems, where the different boiler ashes (BOAs) and the electrostatic precipitator ash (ESPA) arise. BOA and ESPA are usually collected together and referred to as FA.

In general, the treatments of FA can be grouped into three classes: (*a*) S/S methods, (*b*) thermal methods and (*c*) leaching methods. Acidic FA leaching appears to be the most effective method for heavy metal separation and recovery (e.g. Katsuura *et al.*, 1996; Ludwig *et al.*, 2005; Tang and Steenari, 2015; Yang *et al.*, 2012). Acidic leaching using scrub water from the wet flue gas cleaning (Fluwa process) has been established on an industrial scale since the 1990s and represents the state of the art in Switzerland (Bühler and Schlumberger, 2010; Schlumberger *et al*., 2007; Vehlow *et al*., 1990). The depleted residue (filter cake) after the Fluwa process has less impact on the environment and can be deposited together with the IBA. The FA in most other countries is directly deposited in underground storages or extracted with water (neutral leaching) to remove water-soluble salts before S/S with cement.

To increase the metal leaching recoveries from FA and therefore minimise the pollutant load to landfills, knowledge of the binding forms of heavy metals and the enclosed phases controlling the metal leaching in FA is essential. FA consists of the following main components: (*a*) refractory particles that pass the incineration process without modification, (*b*) newly formed high-temperature phases surrounded by characteristic dust rims, (*c*) metals that are carried along with the flue gas and enriched in mineral aggregates and (*d*) metals that are vaporised and condensed as chlorides or sulfates.

About 40 wt.% of the phases in FA are present a crystalline form. This is remarkable when considering the rapid formation and cooling processes taking place during incineration and transport in the flue gas. Refractory particles such as aluminium foil, remaining carbon particles, quartz fragments and hollow glassy cenospheres are characteristic of FA. Newly formed high-temperature phases such as gehlenite ($Ca_2Al_2SiO_7$) and wollastonite ($CaSiO_3$) are mostly surrounded by a characteristic dust rim. Such reaction products are often found in contact with refractory particles (quartz and feldspars) that form the core of the mineral association. Heavy metals are either transported in the flue gas in particulate form (e.g. iron fragments and brass) or vaporised and condensed as complex chlorides or sulfates in FA (Weibel *et al*., 2017).

The chemical composition of FA from different incineration plants varies significantly, mainly depending on waste input and season. A high industrial waste content during incineration results in a higher concentration of heavy metals and a lower concentration of calcium (Ca) in FA compared with that of FA arising from processes where household waste prevails. The average chemical composition of FA is dominated by calcium oxide (CaO) (270 000 mg/kg), chlorine (Cl) (130 000 mg/kg), sodium oxide (Na_2O) (100 000 mg/kg), silicon dioxide (SiO_2) (80 000 mg/kg), potassium oxide (K_2O) (70 000 mg/kg) and sulfur trioxide (SO_3) (13 000 mg/kg). The metal concentration in FA varies between 10 and 15 wt.%. Of the recoverable elements, zinc (average 36 000 mg/kg) is the most abundant, followed by lead (8000 mg/kg) and copper (2000 mg/kg) (Zucha *et al*., 2020).

FA presents a large potential for economically viable heavy metal recovery. Mainly the elements zinc, lead and copper are of interest for recycling. Compared with physical processes for the recovery of metals from IBA, chemical processes show an effective way to recover metals from FA. Acidic leaching is the most effective FA-treatment method for heavy metal separation and recovery. Heavy metals are thereby transferred into an aqueous solution and separated using appropriate methods. Nowadays, more than 60% of the FA in Switzerland is treated according to the Fluwa process. The efficiency of heavy metal leaching depends on the binding environment of the metals, pH value of ash slurry, redox conditions, liquid-to-solid ratio, temperature and leaching time (Weibel *et al*., 2017). With an optimised Fluwa system, recovery rates of 50–80% zinc, 50–90% lead and 30–70% copper are possible (Schlumberger, 2021). The addition of an oxidising agent such as hydrogen peroxide (H_2O_2) during FA leaching keeps the redox-sensitive metals (mainly lead and copper) in solution, leading to significantly higher depletion factors. The resulting ash slurry is separated into a metal-depleted filter cake and a metal-enriched leachate. The filter cake is deposited together with IBA, and the metalliferous leachate either is fed to a WWT plant (hydroxide sludge precipitation) or can be used for direct metal recovery (e.g. Flurec process).

The Flurec process (Quina *et al*., 2018) offers a means of recovering high-purity zinc from the heavy-metal-enriched leachate coming directly from the Fluwa process (Schlumberger *et al*., 2007). Thereby cadmium, lead and copper are separated from the leachate by a cementation process. For this purpose, zinc powder is added to the leachate as a reducing agent and metals more noble than zinc are separated as a metallic cementate. Due to the high lead load of 50–70%, the cementate can be sent directly to a lead smelter, where the remaining heavy metals are also recovered in the lead production process. The remaining zinc is selectively separated and purified from the leachate by solvent extraction followed by an electrolytic zinc recovery process. Since 2012, this process has been implemented in a Swiss MSW incineration plant and allows the recovery of 300 t of zinc annually. The revised Swiss Waste Ordinance prescribes the recovery of metals from FA from 2026 onwards (Swiss Federal Council, 2015). The implementation requires high investments for plant operators. Therefore, the construction of a central hydroxide sludge-treatment plant with integrated metal recovery, similar to the Flurec process, is planned in Switzerland (SwissZinc).

An important factor for operators is the cost of treatment and disposal of FA. The costs of acidic leaching by the Fluwa process have remained constant over the past 20 years and are on the order of €50/t. The entire FA-treatment process brings additional costs for transport and landfilling of the residues, which include the disposal of the leached filter cake at €110/t and of the hydroxide sludge at €90/t. Not to be neglected also are investment and amortisation costs of FA treatment, which are variable depending on the size and age of the plant and are in the range of €100–200/t. In total, this results in treatment costs of approximately €400/t FA when performing the Fluwa process (Schlumberger, 2021).

If the metals are recovered after the Fluwa process, a yield of €95/t FA can be achieved (Schlumberger, 2021). Of this, 75% is due to the recovery of zinc, which is produced in high-quality form (>99.995%) in the Flurec process. Metal recovery also eliminates the landfill costs of the hydroxide sludge (€90/t FA). Taking all Swiss FA (80 000 t/year), a resource potential for zinc of around 2000 t per year can be achieved, which corresponds to about 15–20% of the total annual zinc import to Switzerland.

To promote circular economy, heavy metal recovery efficiency from FA needs to be improved further. This can be achieved by modified process conditions or by an optimisation of the FA input. Specifically, ash fractions with poor extraction properties should be excluded from the leaching process and directly deposited together with the IBA. It has been shown that BOA fractions show characteristics close to those of IBA with a high acid-neutralising capacity or heavy metals bound in poorly soluble form (Wolffers, 2021). The properties of such ashes mean that a lot of additional acid must be used to mobilise the few metals available during the leaching process. Another challenge applies to precious metals such as copper, which reacts very sensitively to the Eh–pH conditions during acidic leaching. To increase the extraction yields of these metals, the focus must be kept on stable leaching conditions.

Recent developments in marine waste and biosolid management

An important issue related to marine waste entails the sustainable management of dredged sediments. The fate of dredged sediments depends on the disposal options chosen by holders (e.g. port authorities) since different pieces of national legislation (water, habitat, waste, soil) may apply. In most cases, dredged soils are classified as ‘waste’ and the disposal in large fill-in basins is often used with concomitant high environmental risks. Although in Europe, no directive exists to address specifically the disposal of dredged sediments, the 2008 EU Waste Management Law (European Council, 2008) provides some alternative management options. Many Member States’ waste regulations reflect the basic principles of the EU waste framework directive European Commission, 2022) that prioritise the reuse, recycling and recovery of sediments. Dredged sediments, however, need important treatments to improve their properties before any reuse. Research activities are underway to improve knowledge about the physical and hydromechanical properties of dredged sediments when treated for reuse as secondary raw materials. Since the 1950s, several solutions have been proposed for improving the physical–mechanical characteristics of soils through the use of hydraulic binders (e.g. Broms and Boman, 1977; Croft, 1967; Dalla Rosa *et al*., 2008; Dumbleton, 1962; Federico *et al*., 2015; Herzog and Mitchell, 1963; Locat *et al*., 1990, 1996; Mitchell, 1981). The problem with these solutions, however, is in the considerable quantities of carbon dioxide released into the atmosphere for the production of binders themselves. The most recent scientific literature (e.g. Assadi-Langroudi *et al*., 2022; García *et al*., 2017; Islam *et al*., 2017; Papadimitriou *et al*., 2017; Smith *et al*., 2017; Wang *et al*., 2015; Yao *et al*., 2014; Yoobanpot *et al*., 2018) has reported alternative mechanical and chemical stabilisation methods to identify those that combine effectiveness, durability, cost-effectiveness and environmental sustainability.

Mussels account for almost 10% of the global fishery industry where, for example, in southern Italy, where one of the world’s largest mussel farms is located, 2 t of mussel shells is transported daily for landfilling and an additional 10 t illegally disposed of to avoid landfilling costs. The procedures for mussel shell disposal are complex, as they represent a difficult food waste to manage. On the one hand, mussel shells are considered wet waste because of their animal origin, whereas, on the other hand, the calcium carbonate they are made of is non-biodegradable with very long disposal times. When mussel shells are illegally disposed of in the sea, they can harm ecosystems and can cause environmental problems by accumulating on shorelines (Jović *et al*., 2019).

The use of mussel shells has been suggested for various applications. Shells are used for soil fertility improvement (Paz-Ferreiro *et al*., 2012); for treatment of acid mine drainage (Uster *et al*., 2014); for adsorption of dyes and heavy metals (Papadimitriou *et al*., 2017); as de-icer grit for street and pavements, drainage layer in green roofing structures and raw shell biofilter (Morris *et al*., 2019); for formulation of pharmaceutical and construction field material; as polymer filler (Hamester *et al*., 2012); for biodiesel production (Buasri *et al*., 2015); and so on. Furthermore, preliminary studies report the beneficial effect of mussel shells in the manufacturing of green building products and as additives to traditional binders (e.g. Martínez-García *et al*., 2017). The recent literature shows that various types of cement mortar can be prepared by replacing quarry limestone aggregate with limestone obtained as a by-product from waste of the mussel cannery industry (Li *et al*., 2015; Mo *et al*., 2018). Shells are converted following three production steps: (*a*) washing process to remove salt components from seashells, (*b*) heating and calcination at a high temperature (500°C) to remove water and organic materials and (*c*) grinding of the calcined product. Mortars obtained by using mussel shell limestone exhibit a more packed microstructure, which facilitates the setting of cement and results in improved mortar strength. The enhanced mechanical properties of the new mortars allow the cement content in the final mortar composition to be decreased and production costs to be reduced. However, these products do not imply a reduction in energy consumption since mussel shells need to be calcinated before their reuse.

A Swiss and Italian group’s research, who are co-authors of this paper (Cotecchia *et al*., 2021; Petti *et al*., 2022; Roque *et al*., 2021; Sollecito *et al*., 2021; Tang *et al*., 2021; Vitone, 2019, 2021), has proven the viability of recycling shells, as a partial substitute for cements, to stabilise dredged marine sediments mechanically. The original binders, named CemShells, were obtained by mixing commercial cements and mussel shell powder (SP). SP was prepared by grinding and heating at 105°C the raw material – that is, without calcination. SP was prepared by heating the raw material at 105°C – that is, without calcination. After that, the mussel shells were ground and sieved to obtain a powder characterised by clay fraction = 28.51%, silt fraction = 67.55% and sand fraction = 3.94%. The group's investigations at the laboratories of Politecnico di Bari (Italy); ClayLab, ETH Zurich; and Italcementi–HeidelbergCement Group were carried out on samples obtained by mixing together (*a*) fine-grained dredged sediments, (*b*) different types of cements and (*c*) SP in partial replacement of the total amount of binder (Figure 6). The multiscale geo-chemo-hydro-mechanical experimental characterisation of CemShell-based solutions to stabilise soft sediments mechanically aimed to close material loops between dredged sediments and mussel shell management.

The soil used in this research was a dredged sediment (Sed) collected from the IV pier of the Port of Taranto in the south of Italy. The sediment was essentially a silty clay with medium plasticity (PI = 28.61%) and a negative consistency index (CI = −0.71) (Table 1).

CemShell was prepared by partial replacement of the cement binder with SP. Specifically, mussel shells and two types of cements were used: the traditional type I Portland cement 52.5R (PC) and the more sustainable commercial binders type III blast-furnace cement 42.5 N (TGC). The latter is characterised as a green product because of its carbon dioxide emissions being lower than 550 kg/t and the use of at least 30% of recycled material coming from pre- or post-consumption.

Table 1 and Figure 7 show the Atterberg limits and CIs of the natural sediment and the different mixtures after 28 days of curing. In the table, Sed8PC and Sed8TGC are the mixtures formed by mixing sediments with 8% PC, and TGC, respectively. The table also shows the results relative to the mixtures of the sediment with 6% PC or TGC cement and 2% SP (i.e. mixtures

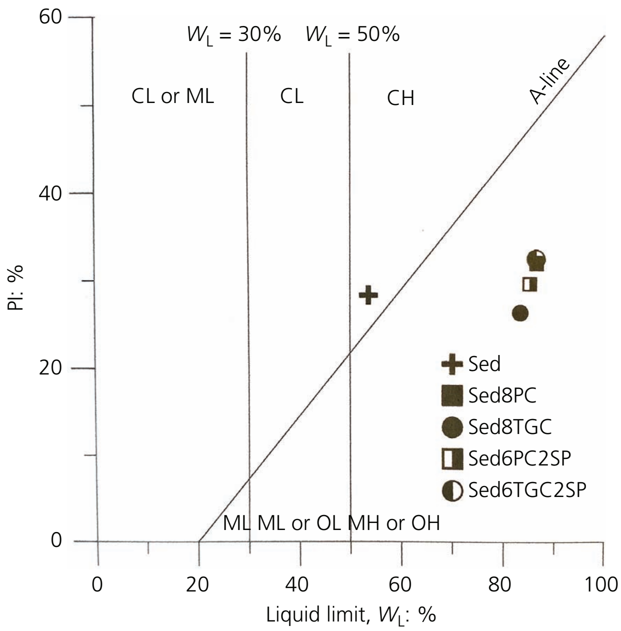

Figure 7. Casagrande plasticity chart of natural sediment and treated sediment after 28 days of curing (source: Rossella Petti)

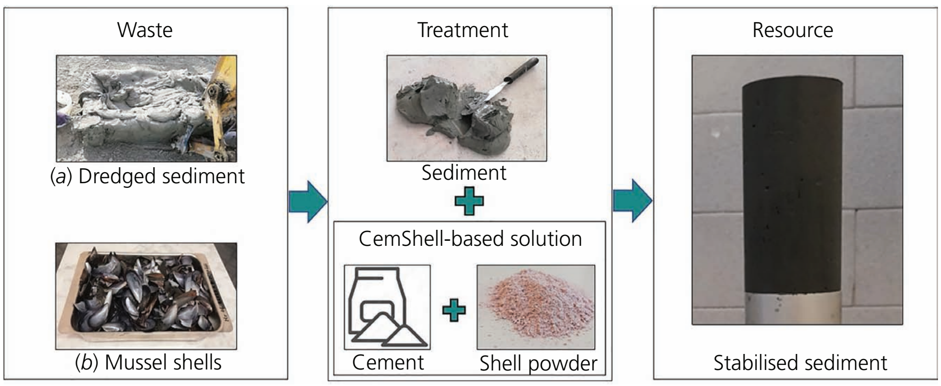

Figure 6. CemShell-based stabilisation solutions (source: Rossella Petti)

Table 1. Physical and state properties of natural sediment and treated sediment after 28 days of curing

Mixture	W_L: %	PI: %	W_0: %	CI
Sed	53.59	28.61	74.0	−0.72
Sed8PC	86.50	32.20	66.95	0.61
Sed8TGC	83.36	26.51	67.82	0.58
Sed6PC2SP	85.17	29.83	66.47	0.63
Sed6TGC2SP	86.42	32.66	66.48	0.61

CI, consistency index (CI = (W_L − W_0)/PI); PI, plasticity index; W_L, liquid limit; W_0, initial water content

Sed6PC2SP and Sed6TGC2SP, respectively). As expected, the use of cement (either PC or TGC) causes an increase in the CI of both the mixtures, which becomes equal to 0.61 and 0.58 for Sed8PC and Sed8TGC, respectively. The replacement of part of the cement with SP seems to cause a further, although small, increase in CI in both the Sed6PC2SP and Sed6TGC2SP mixtures compared with the CIs of the corresponding traditional ones (Sed8PC and Sed8TGC, respectively). In the CemShell-based mixtures, the lowest water contents are recorded. As for the standard mixtures, the CemShell treatments also induce changes to the liquid limit and the plasticity index (PI) so that the soil classification passes from clay of high plasticity (CH) for the untreated sediment to silt of high plasticity (MH) for the treated sediments (USCS) (ASTM, 2017).

The role of SP in the mixture microstructure was analysed by comparing the scanning electron microscopy (SEM) micrographs of natural sediments, standard mixtures and CemShell–sediment mixtures. Figure 8(a) shows the complex system of the marine sediments and the many domains, which are either in edge-to-face or in edge-to-edge contact. Where the soil microstructure is visible, stacks of domains interbedded with zones of random and chaotic fabric can be detected. In Figure 8(b), the fabric of the sediment sample treated with only TGC is shown. The image shows the evident flocculated structure of the treated sediments and the needle shape of ettringite. Figure 8(c) shows the fabric of the sediment treated with TGC–CemShell. The image confirms that SP is completely encapsulated in the cement–sediment structure, acting as a connector thanks to the elongated shape of

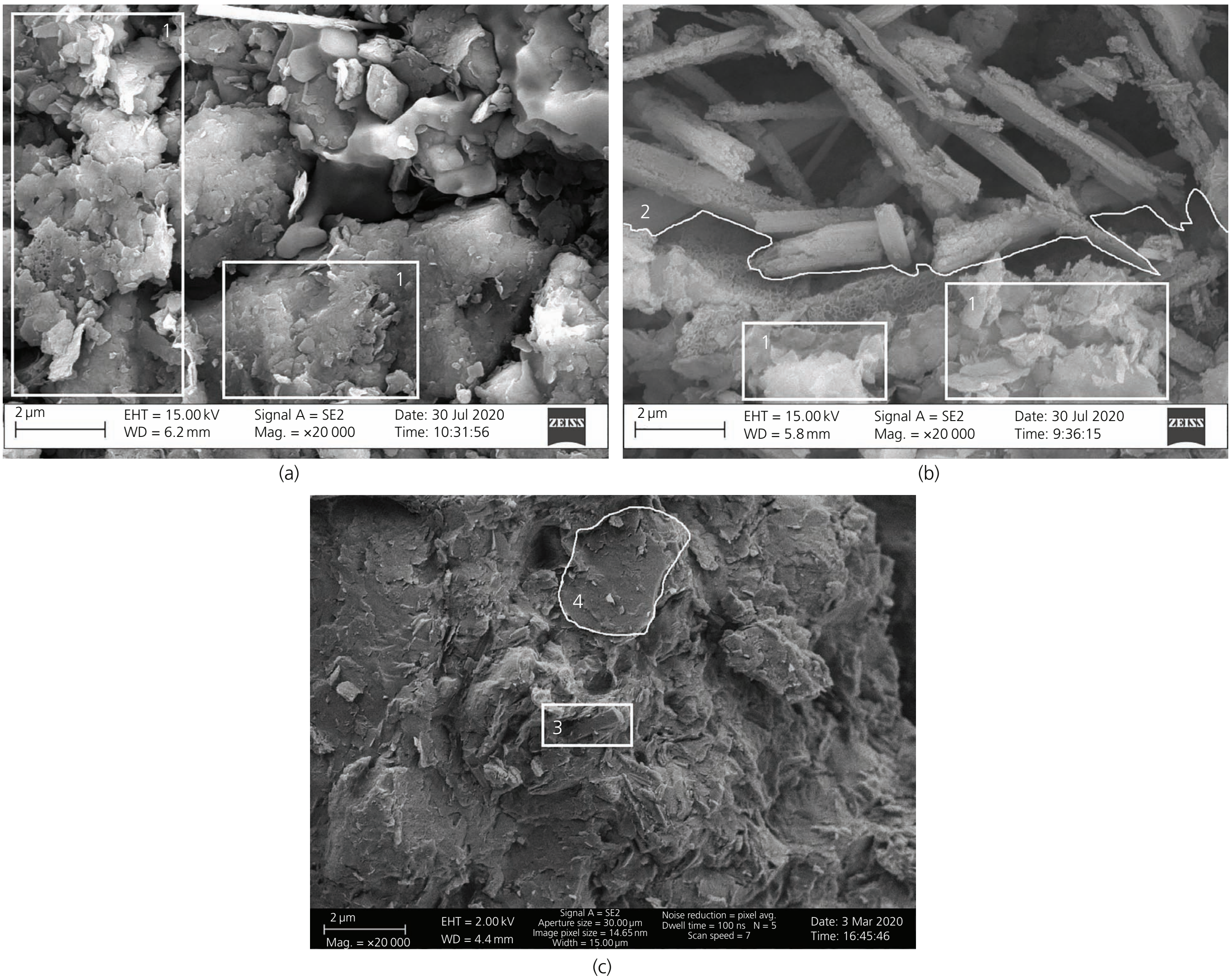

Figure 8. (a) SEM micrograph of untreated sediment Sed: 1, clay particles. Magnification ×20 000 (source: Rossella Petti). (b) SEM micrograph of the Sed8TGC mixture: 1, clay particles; 2, ettringite. Magnification ×20 000 (source: Rossella Petti). (c) SEM micrograph of the Sed6TGC2SP mixture after 28 days of curing: 3, mussel shells; 4, CH plate. Magnification ×20 000 (source: Rossella Petti)

the mussel shell fabric. Moreover, the presence of a portlandite–CH plate (i.e. a typical product of cement hydration) is evident.

Such solutions provide a viable alternative that can reduce the consumption of natural resources (e.g. crushed rock, sand and gravel, extracted through highly impactful quarrying or river exploitation activities) and lower the amount of binders used in traditional sediment stabilisation practice. This work is consistent with circular economy concepts, through actions that ensure the use of secondary resources for harbour water-management activities, necessary to maintain operational navigation depths, as well as transform waste in another industry/value chain through cascading material reuse.

An additional issue with the disposal of biosolid material relates to sewage sludge (SS), the amount of which that is produced annually has increased dramatically in recent decades because of population growth, urbanisation, stricter regulations on the quality of waste water discharge and the demand for new municipal WWT plants and the continuous upgrading of existing facilities. Dewatered SS and biosolids are challenging and unconventional geomaterials, generally with extremely high water and organic matter (OM) contents, very low shear strength, very high compressibility, swelling and shrinkage potentials, very to extremely low permeability and propensity to degrade and produce biogas for pH < 11, such that their compositions and hence engineering behaviour/properties change over time (O'Kelly, 2016). Management of these increasing amounts of sludge materials is a key issue. Principal disposal options include (*a*) land application (grassland, forests and agricultural croplands) (for biosolids) and soil redevelopment/reclamation; volume reduction by mechanical means followed by (*b*) thermal treatment or (*c*) landfilling; and (*d*) indefinite storage in sludge lagoons. Currently, option (*a*) is the preferred route in many EU Member States, given the valuable nutrients of these materials, such as nitrogen, phosphorus and OM, although there are real and emerging issues of land contamination caused by accumulation of high microplastic contents, heavy metals, pathogens and pharmaceutical residues that are concentrated in the SS and biosolid residues by WWT processes (Goli *et al*., 2022; O'Kelly *et al*., 2021). Therefore, inappropriate handling of these materials severely threatens public health and causes serious environmental issues. For disposal option (*d*), there is a legacy of SS impoundment in large sludge lagoons, some being up to 25 m or more deep, with associated potential instability issues due to very low shear strength and biogas build-up causing increases in excess pore-water pressure. Recent experiences have demonstrated that dewatering/consolidation through the vacuum preloading process, chemical conditioning combined with vacuum preloading and sludge solidification are ideal ground improvement options for these ultra and very soft deposits. While disposal into sanitary landfills is presently often the least used method, sludge monofilling, whereby these materials are mechanically dewatered and then allowed to solar dry in evaporation lagoons at WWT plants before their placement and compaction in dedicated engineered monofills (O'Kelly *et al*., 2020), represents an option to maximise the storage potential and provide for future resource recovery and nutrient recycling. Chemically stabilised SS and biosolid materials can be used as construction material as landfill cover, road sub-base, embankment and core fill, provided that special protection is in place if a risk of leaching/flow to adjoining water bodies exists. Rather than conventional cement and lime, there is increasing emphasis on reusing industrial by-products, including FA and slag materials and bauxite residue, for S/S to achieve desired properties, although most of these results are project specific and relate mainly to the use of new binders and SS from specific sources. Besides pathogen inactivation, odour control and toxic compound removal, proper sludge treatment/disposal could also achieve a significant decrease in greenhouse gas (GHG) emissions. For instance, Wei *et al*. (2020) found that of the four main technical sludge disposal routes in China, in terms of GHG emissions, incineration $\gg$ land application > sanitary landfills > land application > building material production. It could be argued that land applications are merely a preferred sludge disposal alternative when compared with landfill or incineration, with a minimal agricultural loss if the total volume of solids produced was reduced and/or a separate more beneficial use was found for them. For instance, new thermochemical and liquid extraction technologies may better recover energy and metals and reduce sludge quantities while inactivating pathogens and destroying organic pollutants (Mulchandani and Westerhoff, 2016). While this shift from 'removal and treat' to 'recovery and reuse' may increase the economic cost of sludge treatment, it could be balanced by cleaner environment and valuable resource benefits for society, cognisant of the fact that future strategies need to be adapted to local economies and the geographical context (Shaddel *et al*., 2019).

Insights on landfill groundwater monitoring and bio-monitoring

As the move towards material recovery from waste is promulgated, it is of interest also to assess existing regulations aiming to protect the environment from MSW disposal. Thus, for example, in the EU, the landfill directive (EC, 1999, 2018) mandates that all Member States must reduce to 10% the total quantity of MSW directed to landfills by 2035, a goal already achieved by ten European countries (EEA, 2021). Despite such encouraging developments, existing landfills have a life of several decades, and hence, monitoring of the processes taking place in them and the pollution risks they pose constitutes a necessity that extends way beyond the above timeline.

Annex III of the 1999 EU directive (EC, 1999) specifies the monitoring requirements during the operation and post-closure phases of a landfill. These include the collection of meteorological data; sampling of the leachate and surface water volume and composition and of gas emissions; and data on the structure, composition and settling of the landfill body. Of interest is the text in the annex on groundwater monitoring, which requires 'at least one measuring point in the groundwater inflow region and two in the outflow region' (EC, 1999), the number of which can be increased depending on the hydrogeologic conditions of a particular site. Although the sampling frequency of groundwater level is defined for both the operational and after-care phases to be at a minimum semi-annually, the frequency for groundwater sampling of recommended parameters (pH, total organic carbon, phenols, heavy metals, fluoride, arsenic and oil/hydrocarbons) is left open to site-specific conditions, not mandating a maximum period between consecutive samplings. The above requirements have not been amended by the 2018 EU directive (EC, 2018).

However, experience from landfill monitoring schemes in several countries has shown that operators rarely conduct studies to determine the number, location, depth and design of monitoring wells and the frequency of sampling to optimise the likelihood of detecting potential landfill leakages. In particular, for landfills located adjacent to farming areas or containing different cells, developed at different times, with variable waste, the existence of a small number of downstream wells makes it extremely difficult to detect and differentiate pollution sources, to conduct effective remediation and to allocate the cost to the appropriate polluter. Besides, several numerical studies (e.g. Papapetridis and Paleologos, 2011, 2012) have shown that in relatively heterogeneous soils (variance of log(K: hydraulic conductivity) equal to 2) and even with low/medium dispersion (α_T = 0.01 m), the probability of detecting (P_d) groundwater contamination remains small, even with the use of a large number of monitoring wells and with frequent sampling (Figure 9). It is clear that future amendments of the EU directive on landfilling of waste should provide minimum standards on the design of groundwater monitoring wells and sampling frequency.

Recently, research has used the type of plant species (bio-monitoring) to monitor landfill impacts (Koda *et al.*, 2022; Vaverková *et al.*, 2022). Landfills have the potential to lay the foundations of new ecosystem types on their surrounding areas, posing potential risks on neighbouring native ecosystems (Vaverková *et al.*, 2019).

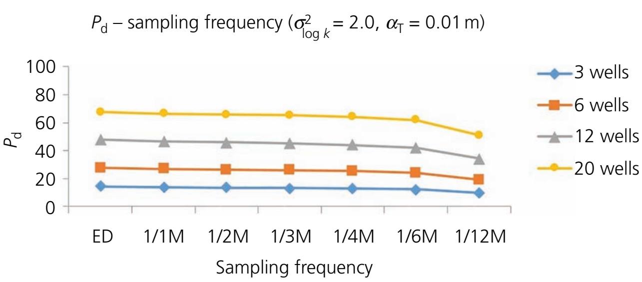

Figure 9. Probability of detecting groundwater contamination (P_d) against the number of downstream monitoring wells and frequency of sampling. ED, every day; 1/*x*M, once every *x* months (source: Evan K. Paleologos)

The first of such risks is the high representation of invasive plant species leading to biodiversity loss (Winkler *et al.*, 2021). On landfills, these plant species may be sources of diaspora, and their further spread to nearby natural ecosystems or agricultural lands can impair the food chain and other relationships among living organisms. These invasive species include *Arrhenatherum elatius*, *Calamagrostis epigejos*, *Impatiens parviflora* and *Tanacetum vulgare* (Vaverková *et al.*, 2022). Many species developing around landfill sites produce allergenic pollens that may be directly harmful to the health and quality of life of people living not only in the vicinity of landfills but also at more distant areas (Vaverková *et al.*, 2019). The second risk is the contamination of plants with hazardous substances released from wastes, which may hamper these plants in joining the food chain. This particularly applies to entomophilous plants and those plant species producing fruits and seeds (e.g. *Solidago canadensis*, *Sambucus nigra*, *Reynoutria sachalinensis*, *Robinia pseudoacacia*, *Oenothera biennis*) or species spread by windborne seeds (*Acer negundo*, *C. epigejos*, *S. canadensis*) (Vaverková *et al.*, 2019; Winkler *et al.*, 2021).

Man-made habitats constitute a new environment for vegetation with different environmental conditions. The vegetation near landfills is not stable in terms of its species composition. Common field weeds have been identified at landfills sites in Poland, with the most important being *Capsella bursa-pastoris*, *Cirsium arvense*, *Convolvulus arvensis*, *Elytrigia repens*, *Equisetum arvense*, *Galium aparine*, *Chenopodium album*, *Thlaspi arvense* and *Tripleurospermum inodorum* (Vaverková *et al.*, 2022). The presence of landfill leachates alters the composition of plant species with increasing representation of species more tolerant to salinisation (e.g. *Atriplex prostrata*, *A. tatarica*, *C. glaucum* and *Portulaca oleracea*) and a decreasing share of glycophytes (Koda *et al.*, 2022). Moreover, landfilling leads to the creation of a new type of geologic environment and vegetation based on allochthonous plant species introduced from other areas (Winkler *et al.*, 2021). Vaverková *et al.* (2022) confirmed that ruderal vegetation around landfills consist of xerophilous and thermophilic vegetation: *Arabidopsis thaliana*, *Fallopia convolvulus*, *Galeopsis pubescens*, *Lapsana communis* and others. This work confirmed that conditions at landfills differ from adjacent habitats, and hence, vegetation monitoring can contribute to the evaluation of the impacts of landfills on the environment.

Conclusions

An exposition is provided herein of some selective, recent developments in waste management, focusing on metal recovery from incinerated bottom and FA, the utilisation of mussel shells for treatment of marine sediments and recommendations on existing landfill monitoring requirements. The heavier ash that is generated during incineration of municipal waste, the IBA, in most countries is not fragmented for use as building material in road construction. This results in reduced landfilled residuals but low metal recovery. What is reported here is the practice in Switzerland to maximise metal recovery by applying processes that can result in yields of up to 95% of all metal pieces >2 mm. Additionally, presented are the novel practices implemented in the same country for metal recovery from the lighter ash contained in the flue gas, the FA, primarily of the elements zinc, lead and copper, together with the economics of and need for improvement of current processes.

New research studies are also being detailed, regarding the treatment and reuse of dredged sediments and of mussel shells. The former create problems because of the contaminants that they may contain, and the latter because their calcium carbonate composition is non-biodegradable with very long disposal times. The combined use of both is presented, indicating the viability of recycling shells as a partial substitute of cement to stabilise dredged marine sediments mechanically. This has the potential to reduce the consumption of natural resources and the concomitant land degradation and lower the amount of binders used in traditional sediment stabilisation practices. Recent developments in biosolid management, including chemical stabilisation of SS and biosolid materials employing industrial by-products for use as construction materials, are presented.

The paper concludes by providing recommendations for amending existing landfill monitoring regulations. The need for specific language on groundwater monitoring requirements is stressed together with the suggestion to use, in addition to existing measures, bioindicators to determine the impact of landfills on surrounding vegetation. Monitoring of vegetation (bio-monitoring) can positively contribute to the assessment of the environmental impact of landfills for a more effective management of landfills and other facilities related to waste management. It is recommended that vegetation monitoring be carried out during the main growing season. The specific date of the main growing season is determined by local climatic conditions and varies according to latitude. The distance between the landfill and the surrounding ecosystem is important in terms of the dispersal of some plant species. The potential for natural dispersal is significant up to approximately 5 km from a landfill. Access roads where waste is brought in are also important for the spread of plants. Currently, no strict regulations exist in the EU with respect to the vegetation management of landfills. Considering the authors' many years of experience on vegetation monitoring data, it is suggested that, besides to other types of monitoring, landfills be subjected to regular vegetation bio-monitoring too.

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