Toxic Fallout

Waste Incinerator Bottom Ash in a Circular Economy

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Abstract

Bottom ash is fallout from the grate of mass-burn waste incinerators. Large quantities are produced and this residue has negative value. Visible proportions of sand, glass, and stones make it appear, on the surface, to be low hanging fruit for use in a circular economy; but bottom ash also contains appreciable quantities of toxic 'high level of concern' elements and persistent organic pollutants.

A secondary 'fallout' occurs when these substances leach from bottom ash into its surroundings across a range of conditions and timescales. The waste incineration industry fails to mention these facts when advertising bottom ash as a 'green' building material. In comparison to direct airborne pollution from waste incinerators, bottom ash has gone somewhat under the radar, making it ripe for greenwash.

This report uses independent empirical research to evidence that incinerator bottom ash is insidiously hazardous and underregulated. Risk is heightened by the fact that testing methods for its use as a building material are outdated. A list of fifteen concerns for public health and safety is provided in relation to the use of waste incinerator bottom ash in cement-based products and as road/pathway aggregate. Calls for the support of its use within a circular economy are premature, and, as per the precautionary principle, all ongoing usage should cease. Examination of independently analysed bottom ash provides a diagnostic on the operational steady state of waste incinerators, incidentally raising concerns about operational compliance with emissions legislation and the capacity of incinerators to produce benign bottom ash when fed with municipal solid waste.

1 Introduction

In Nature's biosphere, something's discarded effluence is something else's resource. All naturally occurring 'waste' is readily consumed in the efficient process of elemental recycling that operates at the Earth's surface. Within moments, creatures set about its consumption in earnest. Waste does not occur in nature because nature abhors inefficiency.

In contrast, civilisation in the 21st century has implemented an economic system which is proactively inefficient in terms of how it utilises its natural resources (the finite budget of chemicals that form the Earth's lithosphere and biosphere and the energy contained within their chemical bonds) by seeking to expedite disorder and create temporary, localised financial gain. In doing so, it has taken human endeavour above and beyond stability - i.e. the natural recycling of elements within a finite budget -; and has thrown it into the unstable realm of a throwaway society where, in an attempt to satiate this requirement, greater consumption of goods, services, and fuel must occur in greater volumes than the year before, thus creating increasingly larger amounts of waste.

Prompted by numerous environmental concerns directly arising as a result of this system, and of the logic to transition away from it, a number of ideas have been proposed which, rather than directly challenging the fundamentals of the system, suggest a reconciliation. One of these is 'sustainable development' (Spaiser et al., 2017). Another is 'circular economy' (Ellen MacArthur Foundation, 2014). Waste incineration is considered to be outside of the circular economy (ibid.). Reasons are that it is a destructive process which provides 'back-end pull' for waste generation accentuated by contractual lock-ins (Muznik, 2017).

Currently, the European Union (EU) is examining whether the use of modern waste incinerator bottom ash could be worthy of investment support within a future circular economy. The matter is being discussed as part of a wider EU Taxonomy (EU, 2020). To be aligned, suitable activities must make a "substantial contribution" to at least one of six objectives:

1. Climate change mitigation,
2. Climate change adaptation,
3. Sustainable use and protection of water and marine resources,
4. Circular Economy Transition,
5. Pollution prevention and control, and
6. Protection and restoration of biodiversity and ecosystems;

while simultaneously they must ***"do no significant harm"*** to any of the other objectives; in other words, progress towards one objective must not be made at the expense of another. Compliance is assessed against specified "technical screening criteria", which require that evidence is ***"science based"*** [sic], and ***"developed via a robust methodology"*** (PSF, 2021).

It is not the objective of this report to consider the de-merits of waste incineration within the circular economic model. It is its aim to provide evidence against the aforementioned criteria: specifically, the use of bottom ash in both 'unbound' aggregates (i.e. for roads and paths) and bound composites (e.g. cement based products like concrete and blocks). The topic has wider relevance to the legislative, permitting, and planning sectors where claims are put forward by the incinerator industry that bottom ash can have ***"many applications"***, can be ***"carbon negative"***, and even that it can assist with ***"Climate change adaptation and greenhouse gas emissions"*** (Powerfuel, 2020).

In this report, the hazard (if any) posed by the use of incinerator bottom ash is assessed using independent, empirical, peer-reviewed scientific literature. Specifically, the total concentrations of toxic substances in bottom ash and their propensity to leach out into the environment from subsequent products and applications. Current regulatory and testing safeguards within a European context are investigated, while drivers and motivations for the proposed use of bottom ash are also discussed.

2 Background to Bottom Ash

In the mid-1800's, prior to the first municipal solid waste incinerator (MSWI) patent (Clark, 2007) societal waste comprised mainly dust, ashes, and cinders (ca. 80% - the residue from fire grates), along with lesser quantities of vegetative matter, excrement, bones, and animal carcasses; plus minor amounts of ceramics, rags, paper, and metals (Tanner, 2006). This detritus was frequently piled up within the boundaries of rapidly expanding urban areas, and these refuse heaps were considered to be of some value (Dickens, 1865). People lived among them, scavenging was permitted, and in one city at least a fee was charged for the privilege (Melosi, 1973). Once all 'valuables' had been removed, the leftover ash and cinders were commonly used as a sub-base for paths and carriageways; indeed, in 1848, the whole of London's Great Dust Heap (Figure 1) was reportedly sold to Russia for building the streets of Moscow (Tilley, 2014).

Figure 1. King's Cross, London: the Great Dust-Heap, next to Battle Bridge and the Smallpox Hospital. Watercolour painting by E. H. Dixon, 1837 (Wellcome Collection, no date).

Modern incinerator bottom ash is markedly different from the ash and cinders which were used as a road base in the 1800s. Municipal solid waste (MSW) now includes ubiquitous quantities of plastics and their additives, along with plastic/metal composites such as printed circuit boards and other petrochemically coated substances like paper, packaging, and waste wood (Conesa et al., 2021). A recent report listed over 2400 substances in waste plastic that are identified as of potential concern because they meet one or more of the persistence, bioaccumulation, and toxicity criteria in the EU (Wiesinger et al., 2021).

The majority of modern waste incinerators are mass-burn, grate-fired systems, and the most massive quantity of residue that they produce is 'fallout' from the main grate - 'bottom ash'. Though incinerators are not built to harvest bottom ash, their purpose is to create it: the word's etymological route is a process for 'converting to cinders'. Some incinerators recover a quantity of the energy contained in waste, so-called Waste-to-Energy (WtE) or Energy-from-Waste (EfW) plants. But the waste to electricity efficiency is very low, at $\eta \leq 0.3$, essentially meaning that at least 70% of the chemical functionality in waste is lost in the process of 'converting to cinders' (Neuwahl et al., 2019).

In modern incinerators, approximately a third of the input waste is incombustible or goes uncombusted (Bielowicz, et al. 2021). This equates to about a quarter of the input mass becoming bottom ash (Bunge, 2019; Hulgaard and Vehlow, 2011). The balance - a smaller amount of solid residue - becomes entrained in the combustion gases and is either emitted into the atmosphere (Particulate Matter Research Group, 2019) or captured by gas cleaning modules (Vehlow, 2015). These entrained particles are termed fly ash and air pollution control residues (APCr) and are not part of this report.

The focus of legislation in Europe has been to minimise these airborne emissions, lately implemented via the Industrial Emissions Directive (IED) (EU, 2010). This requires that the post-combustion gas [author's emphasis] must be subjected to at least 850°C for a minimum of 2 seconds even under the most unfavourable of conditions, and that the bottom ashes/slag have total organic carbon (TOC) content of <3 wt% or their loss on ignition (LOI) is less than 5 wt%. Limit values exist only for pollutant concentrations in the airborne emissions and APCr system wastewater. The combustion environment above an incinerator grate is a hostile one to monitor and, though little is known about localised variations, temperatures above the grate are believed to oscillate around 900°C (Bunge, 2019).

At the macro-scale, bottom ash is mostly (between 50 – 97%) amorphous material, stones, shards of glass, chunks of metal, and sandy grit (Buchholz and Landsberger, 1995; Caviglia et al., 2019). The amorphous fraction is often referred to as 'slag' and 'clinker'; a product of high temperatures in the combustion zone at which substances melt, aided by elements from groups 1 and 2 of the periodic table which are fluxing agents (Miles et al., 1995). The words 'slag' and 'clinker' are often used as synonyms for bottom ash. Chemically, bottom ash has a pH in the 11-12 range (Bunge, 2019). Major constituents (ca. 90%) are oxides of sulphur (S), silicon (Si), calcium (Ca), iron (Fe) and aluminium (Al) bound, among which are numerous minor elements from different chemical groups, many of which are very toxic (Simon et al., 2021; Vateva, and Laner, 2020). Bottom ash also has some pure metals and a fraction of these are commercially extractable (Bunge, 2019).

Commercial extraction of metals is influenced by how bottom ash is temporarily stored upon discharge. Some incinerators have quenching systems (a water-filled tank) while others operate dry capture, often with a period of open air stockpiling known as weathering or ageing, each of which can alter bottom ash form and chemistry. Both ferrous (Fe) and non-ferrous (NFe) metals are extractable, but this refers to only unoxidised constituents (i.e. pure, native metals) and not to metal oxides which are grouped with the mineral constituents. Full recovery of all metals is not possible, with the remainder along with metal oxides left within what is sometimes called the 'mineral fraction' of bottom ash. This 'left-over' bottom ash residue is the subject of this report.

3 Method and Hazard Identification

Research was framed by two hypotheses:

1. The use of incinerator bottom ash could substantially contribute to the transition to a circular economy; and
2. Its utilisation will do no significant harm.

The research methodology was a literature review, with papers selected by date of publication from 2019 onwards, and only those which contained results derived from empirical research. Datasets were limited to samples of bottom ash produced by the incineration of MSW, i.e. household and commercial/industrial waste; studies reporting on special 'hazardous waste' incinerators were excluded. Also excluded were publications either directly commissioned by industry, co-authored by, or co-funded by industrial sponsors. The scope was set within Europe, defined geographically; but, for organic substances, it was extended to include empirical studies from other continents which evidenced compliance with EU legislated operational minima and/or Best Available Techniques (BAT).

The potential hazards of bottom ash are a function of its intrinsic chemistry. Further hazards are created by the interaction of bottom ash with the chemistry of its external environment when applied in product form. Risk is assessed also as a function of the legislative framework of safeguards, if any, which govern product manufacture and point of use. Literature commonly expresses the chemical hazard by two metrics: a) the 'total concentration', which is the quantity per unit mass of specific elements and compounds; and b) the mobility of these elements and compounds from bottom ash, termed 'leaching concentration', and defined as the mass of substance per unit volume of liquid used to irrigate the sample.

In this report, chemical hazard identification was based on EU REACH (Registration, Evaluation, Authorisation and Restriction of Chemicals). All the substances listed in Table 1 were present in the bottom ash as reported by the studies which comprise this report. All are considered as High Level of Concern by fulfilling one or more of hazard criteria under EU REACH (namely: very bioaccumulative; carcinogenicity; mutagenicity; reproductive toxicity; endocrine disruption; specific target organ toxicity upon repeated exposure; and chronic aquatic toxicity), combined with the large volumes produced, as identified by Wiesinger et al. (2021). Phase change data is provided in Table 1 so that inferences can be drawn on the conditions inside the waste incinerator and by the presence and form of the substances in bottom ash.

Table 1. Selection of elements* found in MSWI bottom ash from studies in this report, and which are considered as High Level of Concern (Wiesinger et al., 2021). * = Cl^- is an ion and SO_4^{2-} is an ionic compound. ** Halkidiskis et al., 2019; Wiesinger et al., 2021.

Element	Melting Point	Boiling Point	Origin in MSW**
Arsenic (As)	Sublimes at 616°C		Used in electronics and glass, wood preservative. Biocide in plastics.
Barium (Ba)	729°C	1637°C	Antioxidant, colourant, filler, heat and UV stabiliser in plastics.
Bromine (Br)	-7°C	59°C	Major constituent of flame retardants in plastics, foams and textiles.
Cadmium (Cd)	321°C	756°C	Heat stabiliser, antioxidant and pigment in plastics. Used in metal plating and batteries.
Cobalt (Co)	1495°C	2870°C	Catalyst and pigment in plastics. Widely used in magnets and metal alloys.
Chloride (Cl^-)*	n/a	n/a	Plasticiser, heat stabiliser, colourant, antioxidant and catalyst in plastics. Major constituent of polyvinyl chloride (PVC). Wood preservative.
Chromium (Cr)	1860°C	2672°C	Catalyst and pigment in plastics. Used in metal plating.
Copper (Cu)	1084°C	2567°C	Biocide and pigment in plastics. Present as wiring in most electrical goods.
Lead (Pb)	334°C	1740°C	Colourant, antioxidant, UV and heat stabiliser in plastics. Present in batteries, metal goods, glass, electronics.
Mercury (Hg)	-39°C	357°C	Catalyst, colourant, cross-linking agent, filler and biocide in plastics.
Molybdenum (Mo)	2617°C	4612°C	Catalyst, cross-linking agent and flame retardant in plastics.

Nickel (Ni)	1453°C	2732°C	Catalyst and biocide in plastics.
Antimony (Sb)	631°C	1635°C	Main use is as a flame retardant in plastic, Also plastic catalyst, antioxidant and pigment.
Sulphate (SO$_4^{2-}$)*	n/a	n/a	Filler, colourant, heat and UV stabiliser in plastics.
Tin (Sn)	232°C	2270°C	Biocide and antioxidant in plastics. Used as flame retardant, and in metal plate, glass, ceramics.
Vanadium (V)	1887°C	3377°C	Antioxidant in plastic. Also a lubricant in plastic manufacture. Level of concern = vanadium oxide.
Zinc (Zn)	420°C	907°C	Multiple uses as plastics additive: filler, heat stabiliser, flame retardant, slip agent, pigment.

In addition, a number of organic chemical groups are present in bottom ash and are also considered as hazardous. These are commonly known as Persistent Organic Pollutants (POPs) and most are listed within the Stockholm Convention, though some are not. POPs are long-lasting, toxic, known to bio-accumulate within higher trophic levels, and each may comprise many species with similar properties (known as congeners). These are detailed separately within §5.2.

4 Results: Legal Framework of Standards and Testing Methods

Currently in Europe, the use of waste incinerator bottom ash as a building material is fragmented (Blasenbauer et al., 2020). Some countries (Norway) prohibit its use, while one (The Netherlands) uses all that it creates. Some nations use none even though it is permitted, while many have regional variations which range in usage from $0 \le$ wt % ≤ 100. Some have use-specific requirements (e.g. in Italy bottom ash can be used in cement, bricks and expanded clay without treatment or testing, but for road use leaching tests specify only the presence of some heavy metals (Caviglia et al., 2019). Five out of twenty six EU countries provide no regulation at all, while two (Ireland and Luxembourg) prohibit domestic use but allow export (Blasenbauer et al., 2020).

The collective European Standards for building aggregate and cement-based products (EN12620, EN13139, EN13043, and EN13242) employ the general term ***"dangerous substances"***. This refers to total concentration only (no leachate), is unspecific, and is marked as ***"informative"***, rather than ***"normative"***, for the purposes of linking with other EU Directives, none of which provide adequate safeguards - Art. 53 of EU (2010) discusses the minimisation of residues, stating that ***"appropriate tests"*** should be carried out to establish their polluting potential. The matter is delegated to countries, yet no harmonised testing method exists, an issue that was identified more than a decade ago (Blasenbauer et al., 2020).

Of those countries which require leaching tests for bottom ash prior to use as a building aggregate, details are summarised below (Blasenbauer et al. 2020):

- Eleven countries use batch tests, based on EN-12457 – a method not designed for bottom ash as a building aggregate – and which uses water as leachant[1] (EN,2002/2003). This varies between countries via the following parameters:
    - the volume of eluent per mass of sample, quantified as liquid to solid ratio (L/S). Countries use either 2 or 10 l.kg^{-1}, and one country (Belgium) requires a two-stage test. All have a duration of 24 hours . Seven countries apply this test to grain sizes of <4mm, while three countries (plus one region in Belgium) apply it to grain sizes <10 mm.
- Four countries use a column or percolation test, with an elution rate from 0.1 to 10 l.kg^{-1}. Particle size is not specified in Finland and Sweden, while it is set at <4 mm in The Netherlands and two regions of Belgium. Test duration is not specified.
- The Netherlands are unique in also having a monolith test which applies only to bound aggregates. This involves placing a sample of bottom ash in water for 64 days. Particle size is not specified and L/S varies depending on the monolith size.

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[1] 'Eluent' or 'leachant' is the known mass of liquid which flows through, or is agitated within a close vessel in contact with, the known mass of bottom ash, in turn capturing some portion of the toxins. Following the analysis, the quantity of toxins retained by the 'eluate' are determined as their test specific 'leaching/leachate concentration'.

The broad opinion among scientific authors is that the leaching tests are inadequate and provide insufficient safeguards. For example (Tiberg et al., 2021):

"To what extent the leaching from and presence of metals in the mineral fraction of bottom ash poses a threat to the environment and how this should be evaluated is still not entirely clear."

And, (Simon et al., 2020):

"The standard leaching tests mobilize only small amounts of the complete reservoir of certain substances in incinerator bottom ash."

Further independent observations are provided in the following section which compares empirical bottom ash analyses with the various European limit values for total concentration and leaching concentration. Information on limit values was obtained from Blasenbauer et al. (2020) and Glauser et al. (2021) unless otherwise stated. No comment is made on the suitability of limit values because (Blasenbauer et al., 2020):

"It cannot be concluded whether a specific limit value is too high or too low, since it is unknown how limit values were defined in each country."

5 Results: Empirical Research

5.1 Potentially Toxic Elements

Table 2. shows the studies which met the scope of the literature review. These were from six European countries. Note in particular in Table 2 the detail of sample preparation (ageing, metals extraction, weathering).

Table 2. Datasets which report empirical studies analysing potentially toxic elements in MSW incinerator bottom ash.

Reference	Sample provenance	Sample preparation	Method	Analyte
Bielowicz et al., 2021.	WtE plant, Poland.	Stockpiled indoors for two weeks, post Fe and NFe metals extraction. Sampling of 20kg from 350-400kg each week, analysis over 36 weeks.	Leaching to EN12457 with L/S of 10 $l.kg^{-1}$.	Ba, Cl^-, Cr, Cu, Mo, Pb, Sb, SO_4^{2-}, Zn.
Glauser et al., 2021	Two Swiss MSWI/bottom ash treatment plants: 1 wet discharge, ageing and metals extraction; 2. 'enhanced' dry treatment and metals extraction.	1 tonne, post-metal, sampled at 5-6 times over two days from each plant. Pieces of metal and unburned matter >5 mm removed. All size fractions crushed to <0.25 mm.	Leaching tests: 1. Batch at 10 $l.kg^{-1}$, a) with de-ionized water, and b) with CO_2 saturated water (reduced pH). 2. Column with de-ionized water at L/S of $0.1 \leq l.kg^{-1} \leq 10$.	Cl^-, Cu, Pb, Zn (leachate) As, Cl^-, Cd, Cr, Co, Cu, Ni, Pb, Sb, Sn, V, Zn (total concentration).
Mantovani et al., 2021.	WtE plant, Italy	Five samples totalling 30kg taken over five days. Dried for 24 hours then sieved to various grain sizes.	Total concentrations only.	Numerous.
Simon et al., 2021.	BA treatment facility, Germany.	Fe and NFe separated, no ageing. Wet separation and	6 year leach test experiment using	Cl^-, Cr, Cu, Mo, Sb, SO_4^{2-}, V.

		sieving to grain size range of 25 ≤ mm ≤ 45.	simulated rainwater in a lysimeter.	
Tiberg et al., 2021.	Six different Swedish WtE plants.	Metal extraction then aged outdoors for at least four months or treated to pH 10.	pH dependent leach test on grain sizes: 5.5 ≤ mm ≤ 8.5.	Al, Cu, Fe, Zn.
Kalbe and Simon, 2020.	Bottom ash treatment facility, Germany.	Post Fe and NFe metals extraction, no ageing. Wet separation and sieving to size range 25 ≤ mm ≤ 45.	Four different leaching tests: simulated rainwater in a lysimeter of 2.96 l.kg^{-1}, column up to 9.6 l.kg^{-1}, batch tests of 2 l.kg^{-1}, and 10 l.kg^{-1}.	Numerous
Vateva and Laner, (2020)	German MSWI plant (wet discharge system).	1900 kg taken in 9 days over a four week period. Compared 'as received', with 4 month aged, grain sizes: <0.063 ≤ mm ≤ 31.	Batch leaching test EN12457 at 10 l.kg^{-1}.	As, Cd, Cl^{-}, Cr, Cu, Hg, Ni, Pb, SO_4^{2-}, Zn.
Alam et al., 2019a	Different incinerators in The Netherlands.	Natural weathering for six weeks, then dried, and sieved to ≤ 0.125 mm.	Batch leaching test EN12457-2, at 10 l.kg^{-1}di-ionized water.	Numerous
Alam et al., 2019b	Different incinerators in The Netherlands.	No metals extraction, weathering for six weeks, dried at 105°C, and sieved to 3 grain sizes in range: 0.125 ≤ mm ≤ 4.	Column leaching test to EN 7383:2004, compared to sequential test of acidic, reducing, oxidising stages.	Numerous
Caviglia et al. 2019	Incinerator in Italy.	Single grab sample. Grain size sieved to 0.063 ≤ mm ≤ 20.	Batch leaching test to EN12457 with de-ionized water at L/S of 10 l.kg^{-1}.	Numerous

5.1.1 Comparison between Leaching Test Methods

Glauser et al. (2021) showed how the different leaching test methods of Switzerland and The Netherlands produced incompatible results for numerous potentially toxic elements from the same sample. Statistically significant correlations ($R^2 \geq 0.95$) were only found for Cu and Cl^{-} using deionized water as eluent. There was no statistically significant correlation for Zn ($R^2 = 0.65$), while for Pb the average concentration in the leachate from the Swiss batch test was 2.5 times higher than in the Dutch column test ($R^2 = 0.55$), thus evidencing the relative leniency of the Dutch test.

None of the bottom ash samples met Swiss regulations for landfill due to the total concentrations of heavy metals Cr, Cu, and Pb in some grain size fractions, but in particular Sb which exceeded the threshold for all size fractions, by 11 times in fine fractions, and even by 1.5 to 3 times when the bottom ash was subjected to 'enhanced treatment'. To this, the authors dryly observed that it is for a ***"good reason"*** that the Swiss landfill regulations only have limit values for TOC and certain non-ferrous metals, otherwise bottom ash samples would fail to comply. In general, their results showed that (Glauser et al., 2021):

"Disposal on landfills with lower requirements and recycling of bottom ash as raw material for cement clinker is not possible without applying further treatment steps."

With regard to the Dutch column leaching tests, only Cl^-, Cu, Pb, and Zn were reported, but all size fractions of bottom ash failed for Cl^-, while 62% of the samples also failed for Cu, and 38% failed for Pb. Of this, the authors commented how the Dutch column test, using deionized water, was unrepresentative because it neglected changing pH conditions over time.

Different leaching test methods using the same sample were also compared by Kalbe and Simon (2020). Some toxic elements such as Cd, Co, Ni, and Pb were shown to leach out of the bottom ash in greater quantities under column and lysimeter tests than under batch tests; while others such as Cl^-, Sb, and Sn leached out in greater quantities under batch rather than column testing, supporting the findings of Glauser et al. (2021). They explained their use of a lysimeter in comparison to the common batch or column tests (Kalbe and Simon, 2020):

"The results from lysimeter experiments are closer to real field conditions than the column test due to larger sample size and overhead irrigation rather than up-flow conditions".

As shown in Table 2, no European country uses a lysimeter for leaching tests.

Quantifying the range of results highlights the great variations that exist between batch and column test methods and hence how these permit some countries to use bottom ash while others cannot. Measured as a percentage difference in total concentration (C) across the batch and column tests, each using the same sample $((C_{batch} - C_{column})/C_{batch})$ x 100, values differed by the following amounts: As = 63%, Cl^- = 44%, Sn = 52%, and Sb = 52%. This again quantifies the relative leniency of the Dutch column test, though for some other elements the values were negative evidencing the reverse.

Again of relevance to The Netherlands' use of column or monolith leaching tests, Allam et al. (2019a) assessed a bottom ash sample using the deionized water batch tests commonly applied in other countries. Their results showed that leachate concentrations of Cu, Cr, Mo, and Sb, along with Cl^- and SO_4^{2} from batch tests, would have put the samples above the legal threshold in The Netherlands for the use as a building aggregate. They went on to discuss options to improve bottom ash, and did not advocate weathering because:

"The formation of weathering phases reduces the leaching of potentially toxic elements for the short to mid-term; however these weathering phases are stable only in a limited pH range."

The same research group compared the Dutch column leaching tests for granular building material with a sequential leach test method (Allam et al., 2019b). In a sequential test, the sample is exposed to different conditions and therefore provides a 'worst case scenario'. No European country uses the sequential test.

Allam et al. (2019b) found that the ≤4 mm sample breached the Dutch leach test limit values for Cl^-, Cu, and Mo. They also referred to their own previously published studies from 2016 and 2017, which showed that leaching of Cr, Ni, Pb, Sb, and Zn ***"commonly exceeds the limit for use in non-isolated applications".*** With the sequential leach tests, Zn in particular was highly mobile during the conditions of low pH, while Cr, Cu, Ni, and Sb all showed high mobility during the oxidising conditions. The authors attributed this to the complexation of many toxic elements with humic material, which then become mobile when organic matter is destroyed under oxidisation. These results further evidence that the sterile leach tests are unrepresentative of real life conditions, in this case due to interactions with organic matter, and particularly as an unbound aggregate for road and footpath construction.

More results are provided below, drawing from other studies. To avoid a bland repetition of summaries, further results have been grouped to focus on specific limitations of the leach tests used across Europe in relation to the hazards of bottom ash.

5.1.2 Ageing, pH, Buffering, and Humic Material

When Glauser et al. (2021) changed the eluent of the batch test to lowered pH (using CO_2 saturated water) the differences in leachate concentrations were much greater: Zn mobility increased in the batch test by 15 times in comparison with deionized water eluent. It was also observed that Cu leachate concentrations were higher when dissolved organic matter was present even under alkaline conditions (statistical correlation of $R^2 = 0.90$ between Cu concentration and dissolved organic carbon). The authors noted a high buffering capacity within the smaller fractions when CaO was present, resulting in temporary stability of both Cu and Zn.

Vateva and Laner (2020) compared their leach test results against two current and one draft German standard for building aggregate. Leachate concentrations of Cl^- and SO_4^{2-} in the samples were above the limit values for most grain fractions, while leachate concentrations of Cr, Cu, and Pb were also above limit values. They concluded that the ageing process of four months did not improve the quality of the bottom ash with regard to Cl^- and SO_4^{2-}. Interestingly, the authors observed, though did not explore, a ***"substantial variation in the content of unburned organic matter over the whole nine days of sampling"***. In summary:

"The processed bottom ash was not suitable as a whole, neither as a construction material in constrained structures nor as an aggregate in concrete because it did not comply with limit values in current German regulations...Legal compliance of bottom ash as a construction material was not facilitated by longer aging [and] further manipulation or processing would be required to reduce the contents of soluble salts as well as to minimise residual metal contents."

And:

"The processed bottom ash, as a mixture, did not comply with current German limit values for use as a construction material mainly due to excessive soluble salt contents. Coarser grain size fractions were less contaminated, resulting in an utilisable potential of less than 30% of the bottom ash as a construction material."

The same draft German aggregate limit values were used as a reference by Simon et al. (2021), results which were the culmination of a six-year experiment with irrigation of a bottom ash sample by simulated rainwater (600 $mm.a^{-1}$) in a lysimeter. The lysimeter allows for temporal analyses which batch tests do not (Kalbe and Simon, 2020). In these tests, the pH did not change (9.9 ± 0.5), thus substances in the bottom ash acted as a temporary pH buffer. Even with ageing, short term release of high quantities of Cl^- and SO_4^{2-} were observed, which, along with Cu and Mo, greatly exceeded one of the limit value categories. Both Sb and V continued to be mobile throughout the six years and approached the limit values, as stated:

"Our experiment shows that the release of Sb and V from incinerator bottom ash is not minimised over the time of almost six years. Thus long-term use of incinerator bottom ash e.g. in secondary building materials can pose a potential risk to the environment".

They went on to say that no economically viable technical measures for the targeted depletion of Sb and V are available. And, in comparison to the new draft ordinance for mineral waste, the limit values were not reachable even for wet treated bottom ash (Simon et al. 2021).

In the experiments of Kalbe and Simon (2020), the substrate in the lysimeter was maintained in the alkaline range ($8.5 \leq pH \leq 10.5$). Though the authors say that in this limited range no correlation can be made between pH and Sb mobility, they did say that, at the end of the experiment, (six years) the cumulative release of Sb was still increasing. Sb release was shown to increase with decreasing Ca, as Ca forms less soluble compounds such as $CaCO_3$ (calcium carbonate) due to ageing. Thus, ageing to stabilise bottom ash will not only have an inverse effect on Sb mobility, but pro-active treatments such as adding Ca compounds would be unlikely to solve the problem as, in the long term, these will transform to $CaCO_3$ anyway (Kalbe and Simon, 2020).

Tiberg et al. (2021) confirmed that both Zn and Cu were more mobile below pH 8.5 often as much as four orders of magnitude. They also noted that leachability of these elements above neutral pH was governed by other parameters, but that humic matter increases their mobility. The authors did not refer the results to legal implications because "***clear guidance is lacking, and practice differs between countries***". They specifically referred to the limitations of the Waste Framework Directive.

5.1.3 Incinerator Instability and Hazard as a Function of Grain Size

Bielowicz et al. (2021) sampled bottom ash over a period of 36 weeks and during this time all elements studied at some stage exceeded the national leachate limit values for *"processing outside the plant"* by Polish legislation.[2] For Cl^-, the value was never below the limit at all and reached a maximum at seven times the limit value, while other significant maxima were for Ba (11.5x above the limit) and Sb (21x above the limit), with Zn also exceeding the limits on four dates; while the mean for Sb was more than twice the limit value, and exceeding the limits more often than not. The mean value for Pb concentration in the leachate exceeded the limit value by 70%. This broad variability in concentrations evidenced that the incinerator plant provided little or no constraints on toxic substances in the bottom ash. This was corroborated elsewhere by Simon et al. (2020) who observed that: *"The concentrations especially of trace metals can vary by an order of magnitude".*

Some authors suggest that removing the finer fractions of bottom ash would lead to reduced toxicity (Alam et al., 2019a). But this is not always correct. Kalbe and Simon (2020) showed that the smallest fractions (<25 mm) did not necessarily contain the most toxic elements, with approximately twice the amount of Br, Co, Cr, and Ni in the largest (0.25 ≤ mm ≤ 45) cut. Vateva and Laner (2020) also measured Cr and Pb in greater quantities in larger (>31.5 mm) and smaller (4-8 mm) fractions; Cd, Ni and Zn in smallest fractions; and, for Sn and Sb, partitioning exhibited a random spread. They also found greater concentrations in the >4 mm fraction for Cl^-, Cr, and SO_4^{2-} when the sample had been aged.

Mantovani et al. (2021) detected higher concentrations of Cr and Pb in the largest (>16 mm) fraction, while the highest concentration of Ni was in the 8 – 16 mm cut. A surface coating of finer fragments was found to cover the larger clasts and these were easily separated during transport and sieving, evidencing the friable nature of potentially toxic element release after sieving. Another interesting finding was that the LOI values measured as a function of size fraction: all greatly exceeded the EU (IED) requirements, with the lowest being 9.4% and the highest 26.4%. They concluded against the worth of sieving and screening because of potentially toxic elements (such as Cu, Ni, Zn, Pb, Sn) in all categories of carbonates, sulphates, amorphous, and LOI residues.

Caviglia et al. (2019) found that due to the presence of Cu, Italian limit values were exceeded for all size fractions below 10 mm. Other interesting findings were that LOI values exceeded EU regulations (at 6.2%); and that concentrations of most heavy metals (namely Cu, Ni, Pb, Sr, Zn) were in the mid-range (2 - 8 mm) grain sizes.

5.2 Persistent Organic Pollutants and Microplastics

Only one paper met the scope, from a research group in Norway. The methodology was therefore extended by year and to encompass recent references from outside Europe. Datasets are shown in Table 3.

Table 3. Datasets of empirical studies on organic toxins in MSW incinerator bottom ash.

Reference	Sample provenance	Sample preparation	Method	Analyte
Arp, et al., 2020	Norwegian incinerators via a waste handling facility	Samples taken over a year.	Leaching test with distilled water, as per EN12457 at 10 $l.kg^{-1}$. Shake time increased to 28 days.	PCBs (7 congeners).
Morin et al., 2017	Twelve Norwegian waste handling facilities	Sampled over one year.	Batch leaching test with distilled water, compliant with EN12457 at 10 $l.kg^{-1}$.	BFRs (PBDEs)

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[2] Following personal correspondence with author: "Construction, reconstruction or renovation of railway structures and track beds, embankments, railway and road embankments, road foundations in motorways, impermeable coverings, bowls of earth settlements, cores of hydrotechnical structures and other structures and construction works, including foundations".

			Shake time increased to 28 days	
Lin et al., 2014	Two incinerators in Taiwan	Four samples per day (every 6 hours) over one week. Size fractions < 0.075 ≤ mm ≤ 9.5	Batch leaching test (shaking bottle) agitated for 18 hours with 'fluid' of pH 2.9.	PBDEs
Wang et al., 2010	Incinerator in Taiwan	Three samples per day (every 8 hours) over 3 days. Extractable Fe and NFe metals, 'stones and glass' removed.	Total concentrations. Samples compared to other ash samples downstream of the combustion chamber.	PBDD/Fs (12 congeners). PBDEs (30 congeners).
Liu et al., 2021	Three Chinese incinerators	24 samples taken over two years.	Total concentrations plus supernatant after centrifuge and pH 'stabilisation' to 7-8.	PFAS.
Hsieh et al., 2018	Incinerator in Taiwan	Taken directly from the incinerator over four years.	Total concentrations.	PCDD/Fs.
Chen et al., 2006	Two incinerators in Taiwan	Sampled 4 times per day at 2-hour intervals over one week.	Total concentrations. Size < 0.25 ≤ mm ≤ 9.5	PCDD/Fs.
Yang et al., 2021	Sixteen MSWIs and one bottom ash treatment plant in China	31 bottom ash sampled loads during stable operation.	Total concentrations.	Microplastics

5.2.1 PCBs

Polychlorinated biphenyls (PCBs) were widely used in paint and electronic capacitors until this was restricted in the 1970s. Many remain in circulation as legacy pollutants, but only three European countries (The Netherlands, Belgium, and Czech Republic) assess for the hazard of PCBs in building aggregate, and none assess for PCBs in leachate.

Arp et al. (2020) compared total concentrations of PCBs and their leachability from bottom ash and fly ash against a number of other substances from waste recycling facilities in Norway. They found that mean total concentration of PCBs were much greater in bottom ash (28 ± 34 μg kg^{-1}) than in fly ash (0.3 ± 0.8 μg kg^{-1}) - 93 times greater taken at the mean, while bottom ash showed the second highest leachability of PCBs for all samples studied. These results likely underestimate the true leach hazard, for the study only considered the freely dissolved fraction and not those PCBs associated with particles or dissolved organic carbon: the authors cite previous research that PCBs are mainly associated with particles and dissolved organic carbon by ***"up to 80 - 99.9%"***. These mean total concentrations would have breached the limit value for the Czech Republic and one region in Belgium, which, if considering the full range, all limit values were exceeded. Of note is the wide variation in concentrations over the one year period of sampling.

5.2.2 PCDD/Fs

Polychlorinated dibenzo-p-dioxins/dibenzofurans (PCDD/Fs) are halogenated poly-aromatic compounds, commonly termed 'dioxins'. They produce a range of harmful effects including carcinogenicity, teratogenicity, immunotoxicity, and embryotoxicity. But, of the twenty six countries reported by Blasenbauer et al. (2020), only one of these (Germany) stipulated an assessment of PCDD/F total concentration in bottom ash prior to its use as a building aggregate. No country required an assessment of PCDD/Fs in bottom ash leachate.

PCDD/Fs are more concentrated in fly ash than in bottom ash, but in bottom ash they are still appreciable (Hsieh et al., 2018). Over a four-year sampling period, the mean concentrations of PCDD/Fs in bottom ash (1.48 $ng.g^{-1}$) were approximately three fifths that of fly ash (2.56 $ng.g^{-1}$). Importantly, since the quantities of bottom ash produced were (mean average) three times greater than the quantities of fly ash, bottom ash was the main repository of incinerator PCDD/Fs. Again of note was the wide range of total PCDD/F concentrations in the bottom ash over the sampling period, evidencing the inconstancy of the MSWI process to stabilise PCDD/F fallout. This was expressed by the authors on a WHO_{2005}-TEQ basis[3], with concentrations varying by a factor of seven.

Chen et al. (2006) analysed total concentrations of PCDD/Fs in post metals extraction and air-dried bottom ash from two modern incinerators in Taiwan, described as ***"the most effective technique for PCDD/F emission control"***. Measured on an I-TEQ basis, they found that particles ≤0.6 mm accumulate amounts of PCDD/Fs at a level potentially hazardous to the environment and so unsuitable for use in ***"soil, road sub-base and construction blocks"***. Based on fertilizer limit values in Germany, one sample of bottom ash (grain sizes ≤0.6 mm) exceeded the limit values for application to pasture land, while larger grain sizes (≥2.36 mm) were borderline.

5.2.3 BFRs: PBDEs and PBDD/Fs

Brominated flame retardants (BFRs) are widely used in electronic circuit boards and plastic packaging such as microwave trays and coated textiles (Weidlich, 2021). Their concentrations in plastic range from 3 to 15% (Hennebert, 2021). Some are banned and regulated in the EU, but legacy products are in circulation, so will likely remain in waste for decades to come (ibid.). Polybrominated diphenylethers (PBDEs) are a subgroup of BFRs; many are endocrine disruptors; immune system toxicants; and form PBDD/Fs during low temperature, low oxygen regions above a MSWI grate (Weidlich, 2021). Evidence is that PBDD/Fs are as highly toxic as their better known chlorinated analogues, and more hazardous than the PBDEs from which they formed (Conesa et al., 2021). No European country currently tests for PBDEs or PBDD/Fs in building aggregate, while PBDD/Fs are neither monitored nor is there any limit for these substances in waste incineration residues.

After assessing the total concentration and leachability of PBDEs from a variety of waste handling sites, two of which were bottom ash from a modern Norwegian waste incinerator, Morin et al. (2017) found that total concentrations were approximately an order of magnitude higher in bottom ash than in fly ash, while leachate concentrations were also approximately an order of magnitude higher from bottom ash than fly ash. The study, again, did not consider the leached PBDEs sorbed to colloids or humic acids, but noted that the leaching results were likely ***"biased low"*** due to equilibrium not being reached over the experimental time period. The authors observed that the total concentration of BFRs remained constant between bottom ash and feedstock MSW indicating that they were not destroyed by incineration:

"Bottom ash contains concentrations of flame retardants that cannot be considered negligible [and that] this may need to be considered when landfilling bottom ash, or utilizing it in other purposes, such as filling materials."

In Lin et al. (2014), the PBDE total concentrations were 2 to 19 times higher in bottom ash than in fly ash, further evidencing that PBDEs were not completely destroyed during the incineration process. These total concentrations were two orders of magnitude higher than in urban and rural soils, more distributed in larger (0.25 ≤ mm ≤ 1) particles, and leached out at a rate approximately four orders of magnitude higher than the original waste material. Total concentrations in the bottom ash were 29 ≤ $ng.g^{-1}$ ≤ 243, but there are currently no limit values against which to compare them. The authors qualified the hazard by stating that after passing through the incinerator, the highly brominated leachable PDBEs readily degrade to lighter brominated variants, resulting in enhanced toxicity via increased uptake and bioaccumulation. They also commented that humic solutions enhance the leachability of PBDEs from

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[3] TEQ = Toxicity equivalent. The sum of the products of the concentration of each compound multiplied by its Toxic Equivalence Factor (TEF). TEQ is applied due to the commonly shared toxicity across a suite of compounds to represent how their combined toxicity is additive. Two common metrics exist: I-TEQ based on TEF determined by an expert international group, and WHO-TEQ based on TEFs determined by the World Health Organisation.

bottom ash impacting on its use in ground works. Consequently, they advised caution on the proposed use of incinerator bottom ash as construction material.

The most highly cited study on POP leachability from bottom ash is by Wang et al. (2010). The authors sampled residues of bottom ash and fly ash from two air pollution control modules, and ash from two post combustion zones (economizer and superheater) three times per day at eight-hour intervals for three days, on a system ***"recognized as the most effective technique for PCDD/F emissions control"***. They found that bottom ash had the highest PBDD/F and PBDE content of all residues, supporting that PBDEs were not destroyed by the incineration process; and also that PBDD/Fs were created by it giving higher concentrations than in the input waste. The content of PBDEs in bottom ash were between one and three orders of magnitude higher than in reference soils. They concluded that reutilization of incinerator bottom ash would contribute these substances to the environment.

5.2.4 PFAS

Polyfluoroalkyl substances (PFAS) have been produced since the 1940s and are widely used as flame retardants, and as water and oil repellents in and on plastics (Liu, et al., 2021). They are sufficiently soluble in water to be taken up by plants (Ghisi et al., 2019). None of the European countries reported on by Blasenbauer et al. (2020) tested for PFAS in bottom ash for use as a building aggregate.

PFAS are also not completely destroyed during the incineration process. Liu et al. (2021) found that bottom ash from two out of three incinerators sampled was enriched in PFAS at three times greater concentration than in fly ash. The authors concluded that bottom ash constitutes an important vector for PFAS into the environment; that more study is needed on the thermal transformation of PFAS; and that techniques to destroy PFAS within incinerators need to be developed. Another important finding was the widely varying concentrations of PFAS sampled in bottom ash over the two years.

5.2.5 Microplastics

The hazard posed by microplastics in bottom ash is a very new and underresearched topic. Yang et al. (2021) showed that incineration does not terminate microplastics and their presence in bottom ash ranged from 1.9 to 565 particles per kg, or up to 102,000 microplastic particles per metric ton of waste incinerated. The study was from sixteen modern MSW incinerators established or upgraded to ***"advanced technology"*** in the last ten years and obtained during stable operation. For all samples the LOI was below 3.2%, thus well within the <5wt% stipulated by the EU. The largest fractions of microplastic in bottom ash were identified as from packaging and building materials (polypropylene and polystyrene) indoctrinated with flame retardants thus making them resilient to high temperatures. Unfortunately, the study did not analyse for specific flame retardants or any POPs within the microplastics. There are currently no standardised test methods for determining plastic content in solid matrices, not least bottom ash, and there are no bottom ash/aggregate limit values for microplastics.

6 Discussion

6.1 Processing Influences and Implications

To understand bottom ash composition and its production mechanisms, one must first understand that waste is a poor fuel (Hulgaard and Vehlow, 2011). MSW is highly heterogeneous making its combustion a very complex phenomenon involving thousands of chemical reactions (Chagger et al., 2000). In theory, elements such as Cd and Hg with boiling points lower than the grate temperature should not be present in bottom ash, while others such as Pb and Zn with higher boiling points should always fall out through the grate. However, this does not follow, with also As, Br, Cd, even Hg in bottom ash (studies in this report, plus Buchholz and Landsberger, 1995; Meima et al., 1999; Klymko et al., 2016). Even recent authors question how volatile substances find their way there (Glauser et al., 2021). But it is basic reactor engineering that localised that hot and cold regions occur due to endothermic drying and pyrolysis reactions that sap the internal temperature, along with rich and lean (i.e. oxygen enriched and oxygen depleted) pockets, combined with physical mass and heat transfer limitations, resulting in improper conditions for burning both spatially and temporally; while the presence of many metallic elements such as Cu and Sb create catalysis, speeding up unfavourable reactions that form chlorinated and brominated dioxins (Ebert and Bahadir, 2003; Weidlich, 2021). Added to this there are numerous plastic goods in waste which are impregnated with flame retardants (Table 1), thus not only resistant to thermal treatment, but many of which convert to more toxic forms. This is seen by the range of substances which bottom ash contains, evidencing exceedingly high temperatures (molten Cu) along with substantially low (even unburned sewage sludge is emitted) (Bunge, 2019).

Bottom ash treatment is still a fledgeling industry, progressing largely since the 1990s, and with no two treatment plants the same (Bunge, 2019). Processing is usually undertaken offsite, by a different commercial entity, and frequently after shipping across regional or national borders (Arkenbout, 2019). Mehr et al. (2021) describe how modern plants have extraction efficiencies of between 29 ≤ % ≤ 92 for five metals only, while for Pb it is only 16%. As Simon et al. (2021) state:

"The recovery of elemental metals is still a challenge in terms of recovery rate and purity".

Increased recovery of metals can be achieved by extra comminution, but this would adversely affect the residue's value as an aggregate where the integrity of larger particles must be maintained (Bunge, 2019). It would also increase the risk of toxic dust creation, a problem which is somewhat mitigated by wet discharge predominating in Europe. Wet capture creates new mineral phases, which in theory leads to some stabilisation of heavy metals, but at the same time binds elements into a mineral matrix making them unextractable with current technology (Vateva and Laner, 2020). Dry extraction creates its own problems, one of which is friability and dust containment since the dust is loaded with heavy metals (Bunge, 2019). Airborne dispersal of Pb is identified as a particular critical risk factor with road and sub-base applications (Van Praagh et al., 2018). Adverse environmental consequences of bottom ash dust release have also been reported in recent case studies from The Netherlands where it is claimed that open air transportation and handling (without precautions) resulted in marine fauna endocrine disruption at a UNESCO site (Arkenbout, 2019).

In the EU BAT document for waste incineration (EU, 2019), screening and sieving of bottom ash is recommended. However, the findings of this study do not totally concur with these recommendations, since toxic elements are widely spread across all size fractions and types of bottom ash clast. Similarly, the same document recommends weathering/ageing, and while formerly it was perceived that longer duration was better, Germany for example is moving towards shorter ageing periods, which leads to higher metal extraction potential but also higher solubility of salts and less stable mineral phases (Vateva and Laner, 2020).

Weathering/ageing can also lead to the detachment of finer particles and hence increase the mobilisation of toxins (Alam et al. 2019a). Furthermore, weathering increases the mobility of Sb from bottom ash over long timescales (Kalbe and Simon, 2020). Scientific understanding on the subject is still weak and cannot yet adequately guide praxis.

Some authors tested secondary treatment but this is not totally beneficial and has associated climate/cost impacts. Caviglia et al (2019) found that, after subjecting bottom ash to temperatures up to 1000°C, leachate concentrations of Cu, Pb, and Zn were reduced, but there were two orders of magnitude increase in leachate concentrations of Cr and Ba, and one order of magnitude increase for Al.

The increased mobility of Al is interesting and important for those companies who promote cement bound 'green' products - such as building blocks - made from incinerator residues. The matter is of concern not just for toxic substances leaching out but also structural safety, particularly where high treatment temperatures are used in manufacture (as per Caviglia et al., 2019). Aluminium reacts with water and, over time, releases hydrogen, which can lead to both fires and swelling of the block product from which it is made, thus deteriorating compressive strength (Allegrini et al., 2015; Bunge, 2019). This was supported by studies such as Vateva and Laner (2020) where metallic Al content was above 1% in all grain fractions of bottom ash treated with current BAT; and, even at this level, it was said to impair the utilisation of bottom ash as bound aggregate. Elsewhere, Tiberg et al. (2021) reported concentrations of Al of 5-6% of the total composition of the bottom ash even after metals separation.

When such products are brought to sale in Europe, they come via a weak, outdated, fragmented, and therefore unsatisfactory regulatory system of standards and testing methods. The few nationally approved leach tests represent a compromise between the desire to replicate real conditions and the wish to minimise testing time and avoid crushing (Blasenbauer, 2020). But, as shown in this report, they are not comprehensive of all toxic substances, they only represent short-term and relatively sterile conditions, thereby providing at best only a makeshift snapshot rather than a long-term assurance against product integrity and public safety. By discounting changes in pH and the influence of humic matter they also provide spurious results: High acidity (low pH) of the eluent increases the concentration of metals in the leachate and therefore overestimates the leaching potential, but slightly acidic conditions are buffered by the alkalinity and therefore temporarily stabilise it, leading to underestimation of species mobility. This also undermines the safety of bound products, since Portland cement is known to continue ageing through its lifetime and convert to $CaCO_3$ (Haselbach, 2009). Thus, bound bottom ash products are unlikely to remain stabilised over time within cement-based blocks or concrete claimed to be initially safe. This would also then lead to the fallout of toxins.

An unexpected and incidental finding of this report was the range in LOI values in excess of the legal minima. Surprisingly, this report is believed to be the first to use bottom ash (made available by independent testing) as an incinerator diagnostic. It casts doubts on whether incinerators fed with modern MSW are fit for the purpose of creating environmentally-benign, and therefore usable, bottom ash; but also on the efficacy of current incinerator monitoring and operational stability. They supplement previous concerns expressed by bottom ash treatment plant operators regarding large fluctuations in the quality of ash sent to them and general trends of decreasing quality (Arkenbout, 2019). Others have suggested that sources of POPs could result from MSW incinerator operators not applying BAT (Weber et al. 2019); while periods of Other Than Normal Operating Conditions (OTNOC) have been offered as a further supposition to account for high pollutant emissions (Arkenbout et al., 2018). This report suggests that, in fact, instability may be commonplace even during periods of steady state, a matter which requires further investigation.

6.2 Drivers and Other Motivations

In one of the more readable practical accounts on bottom ash treatment, its author concludes that the push for using bottom ash after metals extraction has nothing at all to do with environmental concerns (Bunge (2019):

"using dry processed [sic] ***BA as a construction material is exclusively driven by commercial interest hiding behind a fig leaf of environmental commitment."***

He is referring to the high cost of landfill and, perhaps, also the legal impositions of reporting and monitoring prior to disposal of a commodity which has negative value (EU, 2004). Similar findings were reported by Arkenbout (2019), where the Dutch environment inspectorate concluded that a high risk of fraud comes from the waste industry due to the negative market value of bottom ash, and indicated a clear problem with current implementation of regulations (Arkenbout, 2019). It went on to say that:

"Due to a lack of commercially viable options to 'clean' the bottom ash to acceptable levels of toxins (POPs and heavy metals), it is simply not done."

There have been case studies, such as Byker (Newcastle), and Jezera (Czech Republic) where the application of incinerator ash led to local soil contamination and POP bioaccumulation, the latter accidentally supported by EU funding (Petrlik and Bell, 2020; Swedish Environmental Protection Agency, 2011; Arnika, 2021). More recently, drawing on further case studies from The Netherlands, it was concluded that (Arkenbout, 2019):

"Though research is limited, what exists indicates strong concerns for public safety and the environment" [It urgently called for further research and...] "until then, any 'useful' application of bottom or fly ash should be suspended."

Trade appears to be a driver for the use of bottom ash rather than domestic usage. Austria has no need for bottom ash as a construction material (Blasenbauer et al., 2020). The same applies to Switzerland, where vast amounts of unpolluted aggregate are generated as a surplus to excavation work (Glauser et al., 2021). When one looks at European trading statistics, a handful of countries (particularly The Netherlands, Germany, Norway, and Belgium) are major traders in natural gravel and sand, with an economic turnover in billions of US dollars per year (Leal Filho et al., 2021). The Netherlands in particular is the third largest global exporter of sand (OEC, 2021). Combined with the unharmonised and fragmented testing methods already reported, the building aggregate standards take a laissez-faire approach which puts the onus of risk assessment on the producer, advising testing only ***"when required or in case of doubt"***, while product control merely reverts back to the inappropriate EU Directives (EN, 2008):

> ***"It is the producer's responsibility to ensure that if any dangerous substances are identified their content does not exceed limits in force according to the provisions valid in the place of use of the aggregate."***

All of which creates a high level of risk to public and environmental health. The main factor controlling bottom ash use in Europe currently is not whether it is scientifically shown to be safe, but rather which leaching test method is chosen and which country or region is the point of sale. Any financial support for 'green' use of bottom ash would likely encourage the movement of hazardous material away from containment in landfill and towards countries with either no, or at least more lenient, environmental regulations. Developing countries seem particularly vulnerable. Some protection is offered by bottom ash being listed on Annex II of the Basel Convention, which seeks to minimise the transboundary movements of hazardous wastes; but not all countries are party to this, and transboundary shipping is permissible with prior consent.

Bottom ash does not have End of Waste status (Blasenbauer et al., 2020). But, according to EU rules, it may be classified as non-hazardous if proven by testing in relation to fifteen hazard classes (EU, 2014). However, there is no harmonised testing method (Blasenbauer et al., 2020). In one Dutch study for the incinerator industry, it was shown that bottom ash was in breach of limit values for the EU waste classification by Pb total concentration, being particularly vulnerable to H10 (toxic for reproduction) and H14 (ecotoxicity) (Klymco, et al. 2016). This confidential report was followed one year later by guidance explaining how the different methodologies for determining H14 could provide different (i.e pass or fail) hazard classifications, despite the limit value breach (Klymko et al., 2017).

The hazards of bottom ash were identified in the 1990s. Meima et al. (1999) found that Cd, Cu, Mo, and Pb showed leachability which was independent of pH, with Zn (all samples) and Cd showing highest leachability at low pH. The sequential leaching methodology is also not new. It was used by Buchholz and Landsberger (1995) who found that leaching of Zn was deemed to be particularly significant due to relatively high quantities in bottom ash; As, Cd and Pb leached in mildly acidic conditions and were assigned as ***"long term leaching hazards"***; while As and Pb were present in the greatest range of compounds, making their containment more difficult.

Yet, nearly three decades later, and despite the evidence presented in this report, one still finds that the facts about toxins in bottom ash are totally missing in waste incinerator industry bottom ash 'fact sheets' (CEWEP, 2019). While they also go unmentioned in waste incinerator planning/permitting applications (see §1).

Recently, the United Nations Environment Programme Special Rapporteur published a document on the environmentally sound management and disposal of hazardous substances and waste. Particular criticism was directed at the waste industry (Orellana, 2021):

> ***"Examples abound of disinformation campaigns developed by companies and industries in order to retain their market share at the expense of the rights of people, including workers, consumers, individuals and communities who are exposed to hazardous substances."***

Among thirty nine recommendations, the following seem relevant (ibid.):

> ***"Design policy interventions to address the risks and harms of hazardous substances on the basis of the best available scientific evidence."***

"Respond to scientific breakthroughs by updating and revising protection measures regarding toxics in a timely manner."

"Apply the precautionary principle in all policy-making and regulatory contexts in which the relevant scientific evidence concerning hazardous substances is inconclusive."

6.3 Findings in Relation to the EU Taxonomy

This research finds that considerations of bottom ash making a substantial contribution to a circular economy transition are premature and unproven, and the hypothesis that the use of bottom ash in civil engineering applications would *"do no significant harm"* is refuted. This is by reference to the following EU Taxonomy objectives:

- Sustainable Use and Protection of Water and Marine Resources;
- Pollution Prevention and Control; and
- Protection and Restoration of Biodiversity and Ecosystems.

The use of bottom ash is seen to be excluded by Art 17, 1, d, iii of EU (2020):

"The long-term disposal of waste may cause significant and long-term harm to the environment."

With respect to Climate Change Mitigation from the use of bottom ash, and while excluding the greenhouse gas emissions directly associated with waste incineration, only one author commented on it, though some mentioned the energy demands of bottom ash treatment without comparison (e.g. Mehr et al., 2021). Bunge (2019) stated that the CO_2 emissions between landfill and the use of bottom ash in place of gravel and sand in building applications cannot be expressed in any meaningful way due to the environmental damage caused by the leaching of metals once *in situ.*

Of relevance is an earlier study from Allegrini et al. (2015) who used empirical data from leaching tests based on bottom ash obtained from a Danish bottom ash processing plant. With this they modelled the toxicity impact for metal leachate only (not POPs) via three categories: carcinogenic human toxicity; non-carcinogenic human toxicity; and freshwater ecotoxicity from the proposed utilisation of bottom ash in nine different construction scenarios. Adverse impact was shown for all three categories in eight out of the nine scenarios: a significant contribution to human carcinogenic toxicity and freshwater ecotoxicity was identified with the inclusion of bottom ash in concrete; while Cr dominated the human carcinogenic impact; and As and Zn were more influential in the non-carcinogenic toxicity category. The high impact of Cr was caused after carbonating concrete specimens were used as a road sub-base, leachate values for Cr and selenium (Se) did not comply with release limit values from concrete specimens set by the Danish government, while leachate from the same sample was not compliant prior to ageing due to excessive release of Pb, and Cu had the greatest adverse impact on freshwater ecotoxicity.

6.4 Alternatives and Recommendations

If waste incineration (a linear activity) remains for some time within a circular economy transition, then better upstream source separation of waste to remove plastics which contain toxins and/or which are known to produce POPs during waste incineration seems essential. Removal of plastics from waste incinerators would, however, have adverse consequences for internal temperature because plastics are of relatively high calorific value. Also halogens - the precursors to many POPs - will remain widely distributed in other waste substances such as coated wood and textiles (see Table 1). A robust system of traceability for toxins in waste would appear to be a preferential activity for funding, and this would offer wider benefits to a circular economy by providing a mechanism to reduce dissemination and repeated cycling of toxins. A better option is, however, to reduce the generation of waste.

Until then, an overhaul of regulatory standards and best practice is needed to better reflect science and the precautionary principle before any further use of bottom ash is sanctioned. This should involve sequential (worst case scenario) leaching test methods and more comprehensive testing of toxic substances in leaching and total concentration analyses with a focus on long-term, life-cycle stability, along with greater European harmonisation.

6.5 Limitations

It is possible that some empirical studies were overlooked during this review. The absence of recent European-based empirical research on POPs is perplexing, and cannot be explained other than by the hypothesis that it reflects a prevalent academic funding environment in Europe, with a trend for research with commercial enterprise potential - in this case, increased bottom ash metals extraction. This would also explain the lack of research studies on toxic organics, which would likely elicit no such immediate benefit.

Only one study reported on a single grab sample. The rest provided results of long-term bottom ash sampling and analysis (maximum six years), thus strengthening the value of these results. Variations in waste composition could increase the potential for error, but this is constrained by European nations having broadly similar waste demographics (Hoornweg and Bhada-Tata, 2012). Results for organic toxins from Asian incinerators might need treating with some caution, though the plastic fraction in waste is similar globally (ibid.).

A number of studies are considered as underrepresenting the true hazard of bottom ash. For example, Glauser et al. (2021) picked out unburned organic material >5mm from the bottom ash prior to sampling; while Bielowicz et al. (2021) excluded certain samples which contained Pb exceeding national limit total concentrations, and despite this their results still showed Pb concentrations which exceeded the leachate limit value by 70%. Some studies were limited by the range of elements analysed: Vateva and Laner (2020) did not analyse Sb, an element which was shown to exceed the leachate limit values elsewhere for similar tests (Glauser et al. 2021, Simon et al. 2021).

7 Conclusions

Arising from this study, there follows a list of concerns for public health and safety relating to the use of incinerator bottom ash residues in 'circular' applications:

1. Current standards for safety are outdated. In the EU, the use of bottom ash is inadequately regulated; rather there exists a hotchpotch of, at best, autonomic rules and guidelines, with many countries having no requirement for testing.
2. Bottom ash contains significant total concentrations of elements which are a 'high level of concern' based on EU REACH hazard classifications.
3. Bottom ash test methods have inconsistently prescribed total concentration values, with regulations only requiring the determination of a handful of toxic substances.
4. Bottom ash leaching test methods have inconsistently prescribed limit values, with regulations only requiring the determination of a handful of toxic substances.
5. Bottom ash leaching test methods are not based on current science and underrepresent real conditions:
    a. They consider short-term leaching only, with some toxic elements mobile after six years of experimentation.
    b. They give spurious results due to pH buffering. This makes the sample appear to be more stable than it actually is.
    c. They fail to consider the influence of humic matter, which is shown to accelerate leaching.
    d. For bound applications they fail to consider the long-term effects of cement carbonation due to atmospheric CO_2 uptake and weathering. This gives a false estimate of stability.
6. There is a likelihood of hazardous bottom ash export to countries with more lenient regulations.
7. The waste incinerator industry fails to mention the hazards associated with bottom ash in its 'fact sheets' and in permit/planning applications.
8. Microplastics are not destroyed by the incineration process, with up to 565 microplastic particles per kg of bottom ash.

9. PCDD/Fs are present in bottom ash in larger volumes than in fly ash and in concentrations of ca. 3/5 that of fly ash. For bottom ash to be used as building aggregate, only one European country assesses for PCDD/F total concentration and no European country assesses for PCDD/Fs in leachate.
10. PBDE concentrations are an order of magnitude higher in bottom ash than in fly ash, and are not destroyed by the incineration process. No European country assesses for PBDEs in bottom ash to be used as a building aggregate, either with total concentration or leachate.
11. PCBs concentrate in bottom ash in quantities almost two orders of magnitude higher than in fly ash (taken by the mean), and they also leach from bottom ash in higher concentrations than fly ash. Only three countries in Europe assess for the total concentration of PCBs in bottom ash for use as a building aggregate, and none assess for PDBs in leachate.
12. PFASs accumulate at three times greater the total concentration in bottom ash than in fly ash. No European country assesses PFASs in bottom ash for use as a building aggregate either by total concentration or leachate.
13. The EU Best Available Techniques for bottom ash processing are outdated and do not represent current scientific knowledge:
    a. Sieving/screening to remove smaller grain size fractions is not satisfactory, with many potentially toxic elements found in larger quantities in larger grain sizes. It leads to a higher risk of toxic dust exposure and airborne toxin dissemination.
    b. Weathering/ageing is not wholly beneficial. It can lead to higher toxin mobility and indirectly increase the hazard of bottom ash by binding more metals within the mineral fraction.
14. The bottom ash treatment industry is still at a fledgling stage and it is currently incapable of removing all metals. The presence of some, such as Al, causes swelling and hydrogen release, along with a possible fire hazard in cement-bound applications over the long term. Even after treatment, Al is present in bottom ash in quantities liable to disrupt the structural integrity of cement-based (blocks and concrete) products over time, creating long-term risks associated with the use of these products.
15. Many independent studies showed that waste incinerators were not operating at a steady state in compliance with the Industrial Emissions Directive. This impacts not only on the capacity of waste incinerators to produce benign bottom ash, but also raises concerns about the efficacy of waste incinerator monitoring and policing.

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Zero Waste Europe is the European network of communities, local leaders, experts, and change agents working towards the elimination of waste in our society. We advocate for sustainable systems and the redesign of our relationship with resources, to accelerate a just transition towards zero waste for the benefit of people and the planet.

GAIA is a global network of more than 800 grassroots groups, NGOs, and individuals. We envision a just, zero waste world built on respect for ecological limits and community rights, where people are free from the burden of toxic pollution, and resources are sustainably conserved, not burned or dumped. We work to catalyze a global shift towards environmental justice by strengthening grassroots social movements that advance solutions to waste and pollution.

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