Environmental impacts of the use of bottom ashes from municipal solid waste incineration: A review

R. V. Silva[1], J. de Brito[2], C. J. Lynn[3] and R. K. Dhir[4,5]

1 CERIS-ICIST, Instituto Superior Técnico, Universidade de Lisboa, Av. Rovisco Pais, 1049-001 Lisboa, Portugal; e-mail: rui.v.silva@tecnico.ulisboa.pt; Corresponding author
2 CERIS-ICIST, Instituto Superior Técnico, Universidade de Lisboa, Av. Rovisco Pais, 1049-001 Lisboa, Portugal; e-mail: jb@civil.ist.utl.pt
3 School of Civil Engineering, University of Birmingham, B15 2TT, UK; email: cjl301@bham.ac.uk
4 School of Civil Engineering, University of Birmingham, B15 2TT, UK; Phone: +44 121 4145075, email: r.k.dhir@bham.ac.uk
5 Applying Concrete Knowledge, 1A Blackened Avenue, Birmingham, B17 8AP, UK; Phone: +44 121 4278108, email: r.k.dhir@bham.ac.uk

Abstract: This paper presents a literature review concerning the performance from an environmental viewpoint of construction related products made with municipal solid waste incinerator bottom ash. It starts with an initial assessment of the bottom ash, and how it performs when used as aggregate substitute in cement-based products, as cement constituent and as raw feed in cement production. Evaluation of the material's environmental performance when used as aggregate replacement in unbound and cement-bound base and subbase layers for road pavement construction, as well as in asphalt concrete layers, is also undertaken. This paper also appraises the behaviour of ceramic-based products, including glass, glass-ceramics, and general ceramics. As a result of the high quantities of potentially leachable contaminants inherent to the bottom ash, the environmental assessment carried out throughout this paper is mostly based on the materials' leaching behaviour, but also based on life cycle assessments and gas emission analyses. The results of several leaching trials, conducted according to various specifications, were reviewed and paralleled with corresponding regulations, with the objective of establishing the products' viability from an environmental point of view.

Keywords: municipal solid wastes; bottom ash; aggregates; cement-based products; road pavement construction; ceramics.

1 Introduction

The worldwide generation of municipal solid waste (MSW) is significantly large; in 2012, the production of this waste was about 1.3 billion tonnes and is expected to increase to 2.2 billion tonnes, by 2025 (Hoornweg and Bhada-Tata, 2012). MSW is comprised of a wide array of constituents, including food, plastics, paper, metals, glass, and textiles, the amount of which varies according to the practices of different cultures, policies and legislation concerning the management of wastes, and on the main economic sectors of different regions (Burnley, 2007, Liu et al., 2006, Wu et al., 2016).

The incineration of MSW with energy recovery is a fundamental stage of the material's life cycle and management as it allows reducing the mass and volume of MSW by 70% and 90%, respectively (Tillman, 1989). For this reason, it is considered as the best cost-effective approach for treating MSW and conserving landfill space area. Of the initial total mass of MSW, most of it is released in the flue gas (about 70%) and a smaller amount turns into residues caught in the air pollution control (APC) systems (Brunner and Rechberger, 2015). The main compounds existing in these emissions include: hydrogen chloride (HCl); nitrogen oxides (NO_x); carbon monoxide (CO); dioxins - polychlorinated dibenzo-*p*-dioxins (PCDD); furans - polychlorinated dibenzofurans (PCDF) (Alonso-Torres et al., 2010). The plant must be designed and operated in such a way that the flue gas resulting from the combustion process must be subjected to a temperature of at least 850 °C for two seconds in order to ensure proper breakdown of toxic organic substances (CEU, 2000). The temperature requirements increase to 1100 °C for at least two seconds, when incinerating hazardous wastes with a content of more than 1% of halogenated organic substances, expressed as chlorine (CEU, 2000). After the incineration process, close to 25% of the initial total mass of MSW are municipal solid waste incinerator bottom ashes (MIBA) (Brunner and Rechberger, 2015). This fraction, however, depends on several variables including the characteristics of the MSW itself (e.g. content of inert materials), the type of furnace (e.g. moving grate, rotary kiln, fluidized bed), the efficiency of the combustion process, among others, which also affect the properties of the resulting MIBA (Chang and Wey, 2006, Collivignarelli et al., 2017). Considering the high quantities of MIBA generated as a result of the combustion of MSW, rather than being looked upon as useless wastes and disposing them in landfills, there have been noteworthy efforts in establishing effective valorisation techniques and using them as substitute for natural resources in construction applications and into the manufacturing of new materials (Reijnders, 2007b). Indeed, even from an economic perspective, they are more appealing when compared with their natural counterparts; in Portugal, for example, in some cases, the MIBA producer does not charge for the product, since most of the revenue from its production comes from selling the recovered metals.

However, fly ashes and bottom ashes from MSW incineration may contain high amounts of hazardous constituents, which may leach out when exposed to e.g. rainwater and can contaminate nearby sensitive recipients, including water bodies, groundwater systems, and, subsequently, fauna and flora (Fuchs et al., 1997, Shih and Ma, 2011a, 2011b, Huang et al., 2017, Huber and Fellner, 2018). For this reason, in the value adding process of MIBA, in addition to the evaluation of their technical feasibility, leachability, ecotoxicity testing and life cycle assessments (LCA) must also be performed simultaneously in order to increase public confidence and acceptance (Breslin et al., 1993). Therefore, this paper seeks to provide an overview of the environmental impacts of different types of construction materials containing MIBA, based on the results of several studies, which were compiled, reorganized and subsequently evaluated. These applications include its use as aggregate or as raw material in the production of cementitious composites, as aggregates in road construction and in the manufacture of ceramic-based products. The majority of the evaluation made throughout this paper was built upon the MIBA-containing materials' leaching behaviour, as it was, undoubtedly, the most popular approach within the literature to assess their environmental performance. Nevertheless, appraisal to the material's environmental impact was also made on the varying gas emissions (e.g. volatilization of heavy metals and organic compounds) as a result of specific manufacturing techniques and based on LCA studies that have compared its use with more conventional scenarios.

2 Methodology

The preparation of this review followed a specific strategy. The initial phase consisted of gathering publications based on various aspects: relevance of the title in terms of environmental impacts of MIBA-containing materials; type of application including MIBA; and existence of significant data for analysis. In the light of the great number of publications, it became necessary to perform an initial appraisal to ascertain which publications were worth pursuing, based on their contents' quality. An analysis was performed for each publication to establish how relevant its contents were (e.g. tests performed, main results, and conclusions) to the theme of this paper. This information was subsequently identified and written in a spreadsheet. Based on this information, a preliminary table of contents was made, which served as a guide for the upcoming investigation. This led to a comprehensive examination of the information regarding the environmental impacts of the use of MIBA in the manufacture of cement-based and ceramic products, and the construction of road pavements.

3 Treatment processes of MIBA

After the process of MSW incineration, MIBA may be subjected to a number of different treatments to reduce the potentially high mobility of hazardous constituents. Such treatment procedures, which depend on the intended application of MIBA, include washing, particle density-based separation, heat treatment (e.g. hydrothermal solidification, vitrification), stabilization with the addition of hydraulic binders, natural weathering, among others (Dhir et al., 2018).

The latter, being the most widely applied treatment process, is given greater emphasis here. The other treatments, in spite of their importance under certain circumstances, are not described in detail here as this was already made in other publications (Dhir et al., 2018) and it is not within the scope of this paper.

By stockpiling the fresh MIBA for a certain period before its use (usually at least three months) will allow the occurrence of biodegradation, carbonation and hydration reactions (Arickx et al., 2010, Arickx et al., 2006, Baciocchi et al., 2010, Dijkstra et al., 2006). The reaction between the alkaline material and the atmospheric CO_2 results in the formation of carbonates (Arickx et al., 2006, Baciocchi et al., 2010, Costa et al., 2007), mainly calcite (Freyssinet et al., 2002). Further hydration reactions result in the material's greater stabilization (Cornelis et al., 2008, Gori et al., 2011, Marchese and Genon, 2009), through the formation of mineral species capable of encapsulating certain toxic constituents, resulting in improved leaching behaviour (Baciocchi et al., 2010, Cornelis et al., 2006, 2012, Shimaoka et al., 2007, Wei et al., 2014, Wei et al., 2011a, 2011b).

Another treatment process, applied to MIBA in only some cases, is exposing them to high temperatures, leading to changes of the mineral and chemical phases' configuration. The output of this process is a less porous and denser material, exhibiting lower ecotoxicity due to the thermal destruction of organic compounds and lower mobility of heavy metals (Chandler et al., 1997, Cheng et al., 2002, Cheng et al., 2007, Kuo et al., 2003, Yang et al., 2003). Nevertheless, despite the high efficacy of thermal processes, these have high energy demands with their own considerably high environmental impacts associated (Gomez et al., 2009, Miyagoshi et al., 2006) and would only make sense if they are already incorporated to the intended application's production process (i.e. ceramic products).

4 Cement-based products

There have been several studies on the solidification/stabilization (S/S) of MIBA with the use of cementitious binding systems (Li et al., 2018), in order to encapsulate hazardous elements and ensure minimum leaching criteria for safe landfill disposal. However, in this paper, emphasis is made on studies that have used processed MIBA into the manufacture of a value-added construction material, namely its use as natural aggregate replacement in cementitious products, as pozzolanic addition, and as raw feed in the production of cement clinker. Table 1 presents the main results based on the leachability behaviour evaluated in those studies.

One should be aware that, even though some of the leaching tests presented in Table 1 have been withdrawn and replaced with up-to-date procedures, they have nonetheless provided concrete evidence at the time of the study and should not be discarded based on that criteria. Furthermore, the evaluation here and throughout the paper is made based on the relative performance of the materials within the same study, which were analysed under the same conditions, and should not to be interpreted as a comparison of results between different testing methods.

Table 1 - Compilation of test methods, mix design and main findings on the leachability behaviour from the literature

Reference	Proportions of MIBA	Leaching test method	Main results
Aggregate replacement			
Dhir et al. (2002)	50% and 25% of MIBA in concrete	NEN-7341 (1995)	Low release rates, which resulted in concentrations below regulatory limits for drinking water
Ginés et al. (2009)	100% MIBA in concrete, cement to aggregate ratio of 0.25 and w/c ratio of 0.64	EN-12457-2 (2002); NEN-7375 (2004)	Granular and monolithic MIBA-containing materials with concentrations comparable to the control; monolithic samples showed Sb above WAC-H
Roethel and Breslin (1995)	55% MIBA, in concrete blocks, 15% cement and 30% sand	Direct rainwater chemical analysis	Rainwater samples showed high Pb concentrations exceeding the limits of public drinking water
Saikia et al. (2008)	25% MIBA + 75% sand, with cement to aggregate ratio of 0.5 and w/c ratio = 0.50-0.60	EN-12457-2 (2002)	Except for Pb and Ba, all leachate concentrations were within WAC-I
Sorlini et al. (2011)	100% and 23% MIBA as aggregate in concrete with 300 kg/m^3 of cement	EN-12457-2 (2002) UNI-10802 (2004)	Higher MIBA content led to increased Pb and Ba release from granular samples; monolithic samples showed significantly lower mobilization of Ba and F
Van den Heede et al. (2016)	Coarse aggregate with 100% MIBA in concrete with cement content of 350 kg/m^3 and w/c ratio of 0.65	NEN-7375 (2004)	Concrete with MIBA showed leachate concentrations similar to those of the control and below detectable limits; Cu and Mo higher, but within limits
Zhang and Zhao (2014)	Coarse washed and unwashed 30% MIBA, in concrete with 320 kg/m^3 cement content and w/c ratio of 0.51	USEPA (1990): TCLP	Washed MIBA mixes showed lower mobility than unwashed MIBA mixes; still, all concentrations were within the than TCLP limits
Cement constituent			
Li et al. (2010)	50% MIBA with w/b ratio of 0.45	USEPA (1990): TCLP	Concentrations in compliance with GB-5085.3 (2007) hazardous waste limits
Li et al. (2012)	30% MIBA with w/b ratio of 0.25 - 0.30	USEPA (1990): TCLP	Concentrations in compliance with GB-5085.3 (2007) hazardous waste limits
Jurič et al. (2006); Kokalj et al. (2005)	15% MIBA with w/b ratio of 0.55 and binder content of 430 kg/m^3	DIN-38414-4 (1984)	Apart from Cr, Ni and Pb, leachate concentrations were within limits for WAC-I
Onori et al. (2011)	40%/20% MIBA with w/b ratio of 0.40	CEN/TS-14429 (2005)	Despite Cr being slightly above the limit, other heavy metal showed progressive immobilization over time
Polettini et al. (2000)	20%/10% MIBA with w/b ratio of 0.35	CEN/TS-14429 (2005)	Lower buffering capacity of binder incorporating MIBA
Qiao et al. (2008)	90% MIBA (untreated or thermally treated) + 10% $Ca(OH)_2$ with w/b ratio of 0.20	NEN-7375 (2004)	Almost all leached metal concentrations, determined inductively coupled plasma atomic emission spectroscopy, were within detection limits
Tang et al. (2016)	Treated MIBA at 30% + 70% cement with a w/b ratio of 0.7	NEN-7383 (2004)	Bound MIBA exhibited higher immobilization when compared with original MIBA; Cu showed lower leachability after heat treatment
Cement clinker production with MIBA as raw feed			
Kikuchi (2001)	40.6%, 30.4% and 27.5% of MSW mixed ash	JIS-K-0102 (1998)	Leachate concentrations within permissible levels
Lam et al. (2010a)	2-8% MIBA	USEPA (1990): TCLP	Slightly higher leachate concentrations than those of the control clinker but within limits for hazardous wastes

4.1 Aggregate replacement

The use of MIBA as aggregate replacement may involve its conversion to a safe and industry-fit aggregate, which normally involves sorting, crushing, grading, pelletisation, thermal treatment and/or binding the material with cement to produce granules. In the latter S/S process, the lower mobility of hazardous elements is attributed to the significantly reduced surface area exposed to a leaching agent and to mineralogical changes, wherein those elements become physically or chemically bound in the matrix. Reasonably dense cement-stabilized MIBA will exhibit enhanced leaching behaviour and thus the results of its evaluation are more representative of the material's performance during its life cycle, in comparison with the assessment made on the same material, but crushed to a smaller particle size (Sorlini et al., 2017). However, at the end of the product's life, it is likely to be crushed into a granular form thereby making it important to ascertain its leachability, since, from an environmental viewpoint, the leached concentrations would be less favourable (Reijnders, 2007a, Sorlini et al., 2017). Indeed, it has been established, by means of an LCA, that the use of MIBA as partial aggregate replacement in the production of cement-based products can be less preferable when compared to its use in road pavement construction, due to the considerable leaching of metals during the recycling phase of the end-of-life cementitious materials (Allegrini et al., 2015).

The pH level is one of the main variables that influences leachability. Bethanis (2007) assessed the acid neutralization capacity (ANC) of MIBA-based artificial lightweight aggregates using a mixture of 60% coal fly ash and 40% MIBA and compared it with that of the commercial counterpart. Cheeseman et al. (2005) also conducted similar testing on rapidly sintered MIBA-based pellets and compared it with the original MIBA. The results showed that the ANC of lightweight cementitious aggregates appeared to decrease more rapidly with the initial acid additions, but less in the MIBA-based aggregate (Bethanis, 2007). The ANC of milled MIBA, although similar to that of the sintered version, presented somewhat higher buffering capacity, which was due to the presence of higher amounts of $CaCO_3$, whereas the sintered MIBA had lower quantities of it as a result of the thermal decomposition (Cheeseman et al., 2005). This caused the inclusion of higher CaO contents in the crystalline silicate and amorphous structures, leading to higher leaching under low pH levels (Bethanis, 2007).

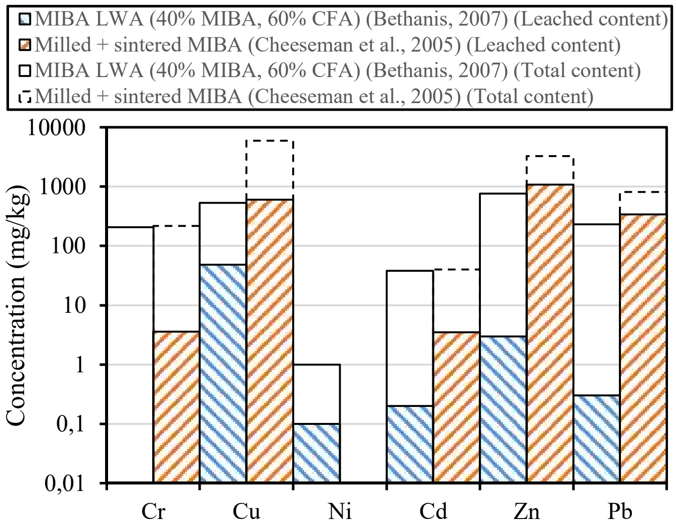

Figure 1 - Leachate concentrations of MIBA-based artificial aggregates (CFA - coal fly ash)

Figure 1 presents the results from the same studies concerning the leachate concentrations of MIBA subjected to a pH level of 3. As anticipated, by subjecting MIBA to an acidic environment, the leachability of the material was higher than what would be expected in real-life circumstances, but was still generally low as only around 10% of the total leachable content was released in the case of lightweight aggregates bound with a mixture of MIBA and coal fly ash (Bethanis, 2007). Furthermore, when compared with sintered MIBA (Cheeseman et al., 2005), the lightweight aggregates consistently showed lower concentrations of elements with comparable total contents, suggesting a greater effectiveness of binding systems to solidify/stabilize a hazardous material. The results of van der Sloot et al. (2001) also suggested that the leaching behaviour of MIBA-based aggregates, produced in a rotary kiln, was very sensitive to lower pH environments. This led them to the conclusion that those aggregates were better suited as natural aggregate replacement in alkaline cement-based products (van der Sloot et al., 2001). Similar conclusions were presented by Wainwright (2002) when studying the behaviour of aggregates fired in a rotary kiln produced with a mix of MIBA (82-90%) and clay (10-18%).

In the study of Cioffi et al. (2011), MIBA were converted to aggregates using different types of hydraulic binders. With the use of a rotary plate granulator, the authors stabilized/solidified with coal fly ash, lime or cement into granules and subsequently analysed the leaching behaviour. The leaching procedure was made in accordance with the guidelines of UNI-10802 (2004), which follows the preparation of eluates specified in EN-12457-2 (2002), and the release of Cr, Cu, Pb and Zn was measured at various periods up to 672 hours. Even though the results showed that cement was more effective at immobilizing those elements, all concentrations were within the allowable releases of heavy metals as per corresponding Italian regulations (Cioffi et al., 2011).

Figure 2 presents the results of leaching tests carried out in accordance with EN-12457-2 (2002) and plots the leachate concentrations from a crushed S/S mix with MIBA as full aggregate replacement and from a control OPC mix (Ginés et al., 2009). All concentrations were compared against those of the European waste acceptance criteria (WAC) landfill guidelines (CEU, 2003). These reference limits were laid down for the acceptance of waste in landfills. However, one should be aware that, normally, it is at the end of the material's life cycle, when it crushed to a finer product and subsequently sent to a landfill, that it is more prone to exhibit greater leachability and not throughout its service life. In Figure 2, even though the use of MIBA as natural aggregate led to increased element mobility, the resulting mix can be categorized as inert, with the concentrations of Pb being borderline. The authors also carried out a leaching test on monolithic samples (NEN-7383, 2004) and observed that, although the concentrations of Cr, Cu, Sb and Mo of MIBA-containing mixes were higher than those of the control OPC samples, all values were inside the range stated in the Dutch Soil Quality Decree (Saveyn et al., 2014). Figure 2 also shows a comparison of the leaching behaviour of washed and unwashed MIBA, with that of natural aggregates (Sorlini et al., 2011). According to the European WAC (CEU, 2003), unwashed MIBA should be disposed in landfills for non-hazardous waste, due to the higher concentrations of Sb and Mo. However, a washing stage significantly improved the leaching behaviour of MIBA, capable of complying with the WAC for inert wastes. Similar results were observed by the same authors for other MIBA in a more recent study (Sorlini et al., 2017), wherein the washed MIBA could be reused as aggregate for the production of concrete as per Italian legislation. As expected, Sorlini et al. (2011) observed that the concentrations of Pb, Br and F^- from ground concrete were consistently higher when compared with corresponding monolithic specimens, yet below the limits specified by the Italian Ministerial Decree for landfill wastes (MD-186, 2006).

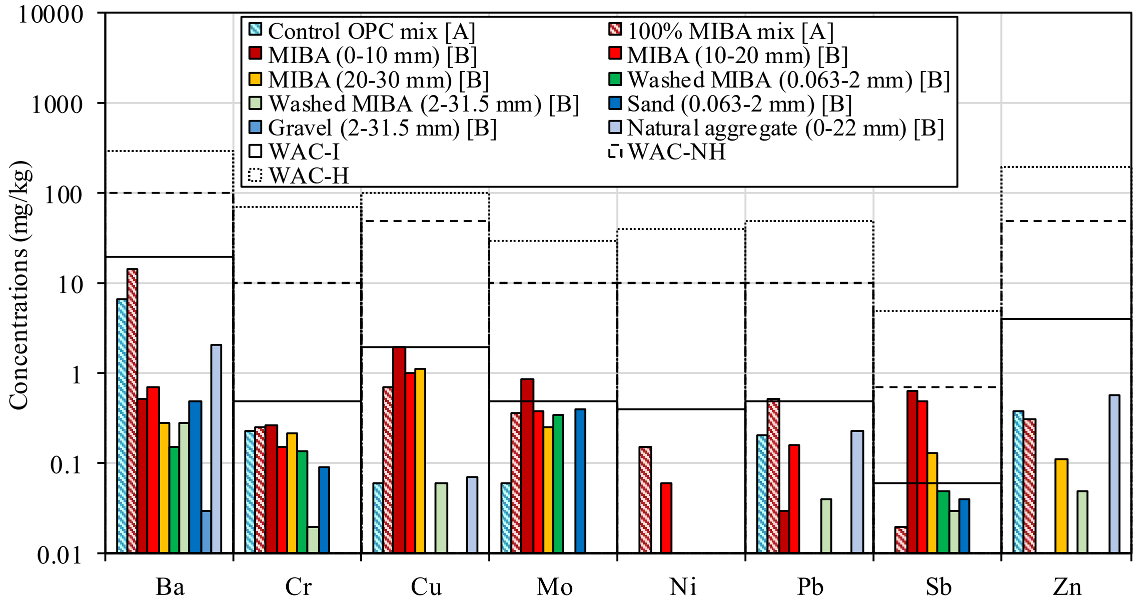

Figure 2 - Leachate concentrations from crushed S/S MIBA-containing mixes (Ginés et al., 2009) and from washed/unwashed MIBA (Sorlini et al., 2011); European waste acceptance criteria for inert (WAC-I), non-hazardous (WAC-NH) and hazardous (WAC-H) wastes for landfills

Del Valle-Zermeno et al. (2013) also analysed the behaviour of 100% MIBA-containing mortars according to EN-12457-2 (2002). The material's granular form showed slightly higher leached concentrations of Pb and Sb than those of the European inert WAC (CEU, 2003). The authors also observed that alkaline materials had enhanced stability of lead-based compounds, which means the leaching behaviour of MIBA can be improved if these are bound in an alkaline environment.

Saikia et al. (2008) used 25% MIBA as sand replacement in mortars and analysed leachability according to EN-12457-2 (2002) guidelines. After having compared with the European WAC (CEU, 2003), the author observed that the mix with 25% MIBA was considered as non-hazardous on the account of Ba and Pb, which were above the inert WAC. However, the control mortar was also considered as non-hazardous, which means, for all practical purposes, that both mixes have equivalent levels of toxicity. The authors also tested monolithic specimens according to NEN-7383 (2004), the results of which showed comparable trends to the previous test method and all element concentrations were within the limits for monolithic materials for new constructions in Flanders, Belgium (Saikia et al., 2008). Similar findings have been observed by other researchers using the same test method and for higher replacement levels (Dhir et al., 2002, Dhir et al., 2011, Van den Heede et al., 2016).

Zhang and Zhao (2014) evaluated the influence of using 30% MIBA, exposed to a washing treatment, on the leaching behaviour of concrete. The leaching test, which followed the TCLP method (USEPA, 1990), showed that mixes made with MIBA subjected to the washing process exhibited lower levels of leachability than mixes made with untreated MIBA. Nevertheless, the heavy metal concentrations of all stabilized samples, as well as those of the untreated unbound MIBA, were already within the limits outlined in (GB-5085.3, 2007) suggesting that those limits may not the most appropriate ones.

Roethel and Breslin (1995) had undertaken a case study on the use of MIBA as replacement for natural aggregate in the manufacture of concrete blocks (15% OPC, 55% MIBA and 30% sand). These units were used in interior and exterior walls in the construction of a boathouse. Evaluations to air quality during a monitoring period of 30 months showed that the concentrations of PCDD/Fs, suspended particulates, volatile mercury and volatile organic compounds inside the boathouse were similar to those in ambient air not exposed to the MIBA-containing bricks. However, analysis to the construction site's surrounding soil showed somewhat high concentrations of heavy metals, which were inconsistent to the leaching test results of MIBA bricks. This expected discrepancy may have been the result of a long-term exposure of the bricks with the surrounding fluctuating water interaction.

In another case study concerning concrete blocks incorporating MIBA, some also contained municipal solid waste incinerated fly ash (MIFA) alongside MIBA (combined total amount of 25% to 50%) as aggregate substitutes (EA, 2002). The results demonstrated the higher levels of toxicity for blocks made with mixed ash (MIBA+MIFA), when compared to blocks made solely with MIBA (117-390 ng I-TEQ/kg and 23 ng I-TEQ/kg, respectively). Nevertheless, it was also observed that the higher levels of the mixed ash blocks would not significantly affect the air quality of a room, though special precautions are required when there is significant dust formation due to e.g. drilling (EA, 2002).

4.2 Cement constituent

MIBA presents some pozzolanicity, the magnitude of which depends on the material's chemical composition and production process. For example, MIBA presenting high glass cullet content is likely to present reasonable pozzolanicity assuming that it adequately processed (Tucker et al., 2018). For this reason, its use as partial cement replacement has also been explored, which also represents yet another solution for the stabilization of hazardous constituents within MIBA (Lin and Lin, 2006). Although the contaminant immobilization mechanism of the previous application can be attributed to the significantly reduced surface area available for leaching, when used as part of the binder, lower mobility may also derive from mineralogical changes.

Researchers analysed the leaching behaviour of cement pastes incorporating 50% and 30%

MIBA (Li et al., 2012, Li et al., 2010) as partial cement replacement in accordance with the TLCP Method 1311 (USEPA, 1990). The leached concentrations of Ba, Cd, Cr, Cu, Ni, Pb and Zn, which were compared to the limits of Chinese National Standards (GB-5085.3, 2007), equivalent to U.S. limits (Liu et al., 2015, USGPO, 2011), were well within boundaries for the identification of hazardous wastes.

Onori et al. (2011) carried out leachability tests based on acid neutralisation capacity (CEN/TS-14429, 2005) on specimens made with MIBA as partial cement replacement. The results showed that the unbound untreated MIBA exhibited leached concentrations of Pb exceeding the WAC for inert waste according to the EU and Italian regulatory limits (CEU, 2003, MD-186, 2006), but complied with the WAC for non-hazardous waste in all other cases. When the 20-40% MIBA were combined with 60-80% cement, increased immobilization had been observed, but the resulting bound material showed values complying with those of the WAC for non-hazardous wastes. However, stabilized specimens showed a notable Cr release when compared to unbound MIBA. Not only was the mobility of Cr from MIBA potentiated by the high alkalinity of cement, but it also exists in relatively high quantities in cement-bound products, probably as a result of the use of coal fly ash though it is also present in the raw materials (Hjelmar et al., 2018).

Qiao et al. (2008) analysed the application of a thermal treatment process of ground MIBA to temperatures up to 800 °C. This allows dehydration of some phases, dehydroxylation of $Ca(OH)_2$, but mostly the decomposition of $CaCO_3$ to CaO thereby increasing its reactivity with cement (Rocca et al., 2013). After having mixed 90% MIBA with 10% $Ca(OH)_2$, the authors observed higher consumption of the latter (20-40%), in comparison with non-treated MIBA (2.5-12.5%). This increased reactivity also resulted in a denser matrix, which improved the material's heavy metal stabilization ability as demonstrated by leaching tests on monolithic samples (Qiao et al., 2008).

Tang et al. (2016) also analysed the influence of a thermal treatment on the leaching behaviour of mortars with 70% cement and 30% MIBA, with a water/binder ratio equal to 0.7. MIBA was subjected types of treatments before its application: milled to particle size <125 µm; MIBA treated at 550 °C and milled to a particle size <125 µm; MIBA treated at 550 °C and milled to <63 µm. The results of the leaching tests (NEN-7383, 2004) showed that the concentrations of Cu, Mo and Sb, from the original unbound MIBA, were significantly higher when compared to the stabilized samples (immobilization of 61-99%). Furthermore, the leached concentrations were all below the limits expressed in the Dutch Soil Quality Decree, which means that the stabilized materials could be used in applications with a high infiltration rate (i.e. 300 mm/year) (Saveyn et al., 2014).

Jurič et al. (2006) and Kokalj et al. (2005) assessed the use of 15% MIBA as partial replacement to cement in the production of concrete and evaluated the leaching behaviour according to DIN-38414-4 (1984), which was withdrawn since then and replaced with EN-12457-4 (2002). The material was categorized as non-hazardous due to Cr, Ni and Pb, which were slightly above the inert WAC (CEU, 2003).

In cementitious systems, evaluation of the acid neutralization capacity (ANC) allows establishing the material's resistance to acid attack, by measuring the solid matrix's ability to maintain high alkalinity, after exposure to an acidic environment. Onori et al. (2011) produced cement pastes with 20% and 40% MIBA as cement replacement and analysed its ANC (Figure 3).

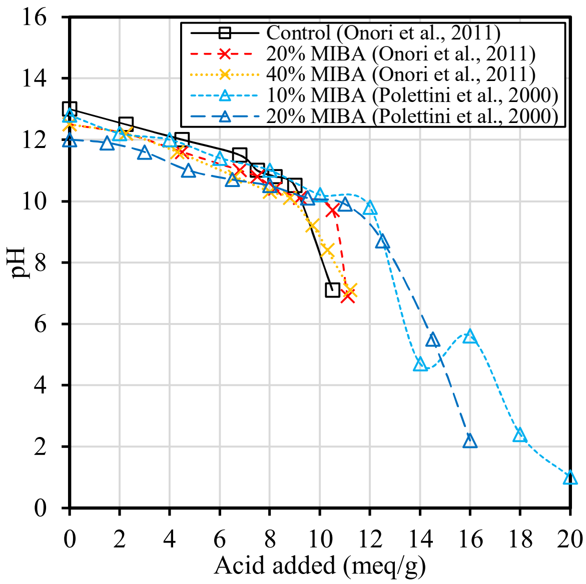

Figure 3 - Acid neutralization capacity of cement pastes with MIBA as partial cement replacement

The results suggested that the use of MIBA as part of the binder caused a slight decrease in buffering capacity, which is indicative of higher heavy metal mobility. Polettini et al. (2000) showed similar findings (Figure 3); for the same quantity of added acid, mixes with 20% MIBA showed slightly lower pH level when compared to mixes with 10% MIBA.

4.3 Cement clinker production with MIBA as raw feed

Since MIBA normally present aluminosilicate composition, its use as raw feed in cement clinker manufacture has been explored. However, environmental related issues can arise from its incorporation, namely additional emissions of volatile hazardous elements released from the kiln, such as Hg, Cd and Pb (Jung et al., 2005, Jung et al., 2004, Jung and Osako, 2007, Reijnders, 2007a), and the possibly greater leaching of hazardous elements from the resulting cement. Nevertheless, one would also have to consider the beneficial outcome of the high temperatures inherent of the cement manufacturing process, which would breakdown toxic organic components, including PCDD/Fs and PAHs, as well as the stabilization of non-volatile heavy metals (Kuo et al., 2003, Yang et al., 2003).

Additionally, Krammart and Tangtermsirikul (2003) also observed reductions in CO_2 emissions when using MIBA as raw feed in clinker manufacture; the use of 5-10% of MIBA, as substitute for raw materials, would save about 25-49 kg of CO_2/tonne of clinker (the production of 1000 kg of clinker may generate slightly over 500 kg of CO_2). In the 28 Member States of the European Union, the annual generation of MIBA was estimated to be close to 16-18 million metric tonnes (Collivignarelli et al., 2017, Lynn et al., 2017) and could answer for part of the considerable demand for raw materials used in the production of cement (the amount of cement produced in EU28 was about 167.2 million metric tonnes in 2015 (CEMBUREAU, 2018)). Margallo et al. (2013, 2014) performed a LCA on the incorporation of 25% MIBA for clinker production. Two scenarios were studied: landfilling of solidified MIBA, including the whole process of MSW incineration and transportation; and Portland cement production, involving the process of incinerating MSW, transport of fresh MIBA and the whole process behind cement production. The results showed that considerable emissions savings could be observed in the latter scenario, including lower consumption of valuable natural resources, among other impacts (i.e. global warming potential, atmospheric acidification, human health, stratospheric ozone depletion and photochemical ozone formation), in comparison with alternative S/S processes.

Concerning the leachability of cement pastes manufactured with MIBA-based clinkers, Lam et al. (2010a) carried out leaching tests, in accordance with the TCLP (USEPA, 1990), on samples made with clinkers with 8% MIBA as part of the raw feed (Table 2). The results showed that the concentrations were within the limits and were only slightly higher when compared to the control clinker. This suggests that heavy metals present in MIBA became immobilized in the cement matrix (Lam et al., 2010b) or were removed during the clinkerization process by volatilization (Shimoda and Yokoyama, 1999).

Table 2 - Leachate concentrations of MIBA-based clinker (values sourced from Lam et al. (2010a))

Heavy metal	Leachate concentrations (ppm)		
	8% MIBA Clinker	Standard OPC clinker	TCLP limit
Ag	-	0,06	50

As	-	0,004	50
Ba	3,841	1,327	1000
Cr	2,402	0,263	50
Hg	-	0,001	1
Pb	3,412	-	50
Tl	0,872	0,056	50
Zn	-	0,065	250

Kikuchi (2001) also reported low leachate concentrations in cement pastes made with clinker containing 27.5-40.6% of MIBA as part of the raw feed. Additionally, the author analysed the exhaust gas during clinker production and observed that the concentration of air pollutants was within permissible levels, suggesting environmental impacts equivalent to those of conventional clinkers.

5 Road pavement construction

The use of MIBA as substitute for natural aggregate in the construction of road pavements is by far the most popular outlet for the material (Poulikakos et al., 2017), in view of these applications' less stringent requirements when compared to the previously mentioned. Still, the main challenge associated to the use of MIBA in this construction application has been the decrease of heavy metal mobility to undersoil and groundwater (Balaguera et al., 2018). Assessments of the material's impacts to the environment have been made on its unbound form, when treated with cement and also when used in hot-mix asphalt.

5.1 Base and subbase layers with unbound MIBA

In comparison to other bound applications, evaluation to the leaching behaviour of unbound MIBA is particularly important, since it is more susceptible of greater leachability due to the higher surface area exposed to water (Abbott et al., 2003). A number of studies were carried out on the leaching behaviour of MIBA-containing road layers, taking into account the influence of different factors (i.e. accelerated ageing, l/s ratio, pH level, microbial attack).

Lynn et al. (2016) carried out a comprehensive review on the leachability of granular MIBA for road pavement construction and in geotechnical applications. A comparison between the amounts of heavy metals obtained from different test methods showed that availability test methods yielded the highest concentrations of elements when compared to batch tests. The former involves acidic conditions to represent a worst-case scenario, which presents a low probability of occurring. The batch tests, however, seek to reproduce the more probable circumstances to which the material would be subjected to and thus provide a representation of the material's behaviour in real life.

Lynn et al. (2016) also compared the leaching behaviour of MIBA with the leaching criteria for the use of waste materials in construction established by some regulating bodies in Denmark (Danish Environmental Protection Agency, 2000), France (French Ministry for Environment, 1994) and Germany (LAGA, 1994), according to the corresponding EN-12457-2 (2002), AFNOR NFX 31-210 (1992) and DIN-38414-4 (1984) batch procedures. It was observed that, in spite of the similar results between the three test methods, significant difference was observed in terms of the allowable limits for waste materials, wherein the German ones were the most stringent and several MIBA samples exhibited leachate concentrations of Cd, Hg and Pb above those limits.

The pH level and liquid to solid (l/s) ratio are key factors that influence MIBA's leaching behaviour (Abbott et al., 2003, Chandler et al., 1997, De Windt et al., 2011, Ecke and Aberg, 2006, Guyonnet et al., 2008, Olsson, 2005). Although MIBA is normally alkaline when fresh, since the material must be subjected to a weathering stage before its use in a more sensitive environment, the carbonation reactions will reduce its pH to a more neutral level. Lynn et al. (2016) extensively studied the influence of the pH level on the mobility of various elements. The authors highlighted the high mobility of Cd and Ni when exposed to acidic conditions and Cr, Cu, Pb, and Zn showed greater release both alkaline and acid conditions. The leached concentrations of Cr, Cu and Pb were lowest when MIBA was subject to a neutral pH level. From a practical point of view, the pH level of a MIBA-containing road pavement layer is likely to progress from alkaline to a neutral level, which would mean the stability of the aforementioned elements, but could result in a greater mobility of Cd and Ni, which increases with decreasing pH level.

The l/s ratio is also a key factor on the leachability of MIBA (Ahmed et al., 2010, Bruder-Hubscher et al., 2001, Chandler et al., 1997, Hjelmar et al., 2007, Izquierdo et al., 2008). Long term testing of full scale road pavement projects built with weathered MIBA as material for unbound subbase layers, include the Dava and Linkoping roads in Sweden (6 and 16 years, respectively), the Ydernæs road in Denmark (12 years), the Herouville road in France (10 years) and the Tagamanent road in Spain (2 years) (Aberg et al., 2006, Bendz et al., 2009, Dabo et al., 2009, De Windt et al., 2011, Di Gianfilippo et al., 2018, Izquierdo et al., 2008, Lidelow and Lagerkvist, 2007). At initially low l/s ratios (~0.5), Cl, Na, and K showed high mobility, when compared to control test sections with natural aggregates (Ahmed et al., 2010). SO_4 behaved differently, with low initial release and higher mobility with increasing cumulative l/s ratios (Bruder-Hubscher et al., 2001, Chandler et al., 1997, Izquierdo et al., 2008). Slower initial dissolution was observed by As, Al, Cd, Ba, Ni and Zn and showed long term high retention rates. Mo, Cr, and Pb though showed higher initial solubility at times, exhibited long term stable dissolution (Lynn et al., 2016).

During the 16-year monitoring period of the Linkoping road, Bendz et al. (2009) observed that the long term accumulated l/s ratio was of 10 l/kg for the MIBA subbase layer, with a variability between 1 l/kg and 50 l/kg. Izquierdo et al. (2008) analysed the long term concentrations of several hazardous elements on the Tagamanent test road section, in Spain. The results, presented in Table 3, suggest that the MIBA-containing road layer presents decreased mobility in most elements exhibited, but, since the quantities of Sb, Cl^-, F^- and SO_4 were above the inert limits according to the EU WAC in landfills (CEU, 2003), it was categorized as a non-hazardous material. Sormunen and Rantsi (2015) also observed that the elements most likely to restrict the use of MIBA for road pavement construction in Finland were Sb and Cl^-.

Table 3 - Cumulative (1-year) releases of hazardous elements (values in bold are above the limits for inert waste

according to European WAC (CEU, 2003); values sourced from Izquierdo et al. (2008)

Heavy metal	Leachate concentrations (mg/kg)	WAC-I (mg/kg)	WAC-NH (mg/kg)	WAC-H (mg/kg)
Al	0.04	0.5	2	25
Ba	0.08	20	100	300
Cr	0.09	0.5	10	70
Cu	1	2	50	100
Mo	0.3	0.5	10	30
Ni	0.1	0.4	10	40
Pb	0.02	0.5	10	50
Sb	**0.09**	0.06	0.7	5
Se	0.05	0.1	0.5	7
Zn	0.3	4	50	200
Cl	**1626**	800	15000	25000
F	**30**	10	150	500
SO_4	**3890**	1000	20000	50000

Some have considered the possibility of using accelerated carbonation to enhance the encapsulation of hazardous elements within MIBA, as a result of the reaction of $Ca(OH)_2$ and other calcium-bearing phases with CO_2, before their use in road construction (Lind et al., 2008, Todorovic and Ecke, 2006). Lin et al. (2015) reported that, apart from the lower mobility of heavy metals, lower release of dissolved organic content may also be observed after accelerated carbonation. Nevertheless, in terms of chloride release, the results of Lin et al. (2015) suggested that this treatment was ineffective and it even increased the mobility of sulphates as a result of the carbonation of $CaSO_4$ and AFt phases.

Sekito et al. (2015) used a high temperature heat treatment based on plasma flame combustion up to 1300 °C and subsequently quenched to stabilize MIBA-based aggregates to be used as material for road base and subbase. The resulting glassy material showed insignificant leached concentrations, none of which exceeded the Japanese criteria.

Stiernström et al. (2014a, 2014b) evaluated the toxicity of leachates from weathered MIBA on the development of larvae. The results suggested the material to present low ecotoxicity thereby validating its safe use in road pavement layers without notable environmental risks.

Phoungthong et al. (2016) suggested that monitoring of impacts to the environment and subsequent protection can be carried out by taking into account the ecotoxicity on fresh water bioluminescent bacteria, since their development is susceptible to the existence of Ba, Cr, Cu, Pb, F^- and toxic organic content, which may leach from MIBA.

The use of a less permeable layer to cover MIBA, such as a cement treated or a bituminous surface layer, would become an effective barrier to rainwater into deeper unbound layers, which is the primary cause of leaching (Bouvet et al., 2007, Triffault-Bouchet et al., 2005). Without such layers, the likelihood of infiltration and groundwater contamination would increase significantly thereby increasing the necessity to develop adequate alternatives to prevent them (Oehmig et al., 2015). Nevertheless, even in a situation with noteworthy rainfall and without impervious surface layers, Bouvet et al. (2007) noticed high accumulation of Pb in the underlying soil, which acted as a barrier of those to more sensitive groundwater systems.

Birgisdóttir et al. (2006, 2007) carried out a LCA making a comparative analysis of two scenarios; one in which MIBA would be sent to a landfill and, in the other, its use in the construction of a subbase layer for a secondary road, both including the transportation of MIBA. The results suggested that, in terms of photochemical ozone formation, global warming potential, acidification and nutrient enrichment, the latter scenario was more positive. However, the authors also observed that, as a result of the underprepared secondary road, in regard to infiltration of contaminated leachate, the long-term accumulation of hazardous elements in groundwater systems would negate the positive impacts of using MIBA. Still, Toller et al. (2009) reported that using MIBA in road pavement construction, despite the potential ecotoxicity, would be more advantageous than landfilling them, since it would prevent other important impacts, including the depletion of natural resources, acidification and climatic change. Furthermore, transportation distances were also highlighted as one of the main variables influencing the results of a LCA's concerning the application of MIBA (Olsson et al., 2006). This means that, even taking into consideration the leachability of the material, it may be significantly more beneficial to use it in the construction of a road than to transport it over long distances to a landfill, or *vice versa*.

Tang et al. (2015) analysed MIBA from two MSW incineration plants during a six-year period. The results of leaching tests showed that the Cu, Sb, Cl and SO_4 concentrations exceeded the regulatory limits for granular materials that can be used in "open" applications, according to the Dutch Soil Quality Decree (Saveyn et al., 2014), where an infiltration rate up to 300 mm/year is expected. However, the results complied with the limits for materials isolated with an impervious barrier, with an estimated infiltration rate up to 6 mm/year.

5.2 Road pavement layers with cement-treated MIBA

Although the leaching behaviour of MIBA in cement bound products was analysed in section 4, since the mix design of cement-treated materials for road construction may involve low amounts of the binder, the leachate concentrations may differ. According to ACI-229 (2005), controlled low strength materials (CLSM) are cementitious materials primarily applied as back-fill, as substitute of compacted fill and subbases and bases in road construction.

Zhen et al. (2012, 2013) evaluated the influence of incorporating MIBA as partial replacement (up to 80%) for calcium sulfoaluminate cement for the production of CLSM. The TCLP leaching test results, plotted in Figure 4, show that among the evaluated heavy metals, the leached concentrations of Cu increased considerably as the replacement level of MIBA increased.

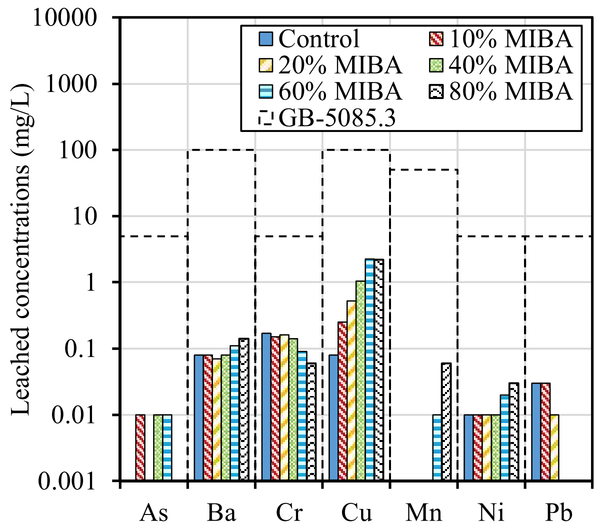

Figure 4 - 24-hour cumulative leaching behaviour of CLSM mortars containing different amounts of MIBA as partial cement replacement (adapted from Zhen et al. (2012))

Similar observations had been made in the study of Cai et al. (2004), wherein the leaching behaviour of cement-stabilized MIBA and coal ash, as replacement for natural aggregate, was evaluated. Nevertheless, Zhen et al. (2012, 2013) reported that all concentrations were well below the levels for hazardous materials as per GB-5085.3 (2007). By means of 3D EEM fluorescence spectroscopy analysis, the authors observed that the low leachability of cement-bound MIBA was associated to the strong binding capacity of AFm and AFt phases, both of which are products of hydration of tricalcium aluminate from cement (Zhen et al., 2012).

Yan et al. (2014) also evaluated the environmental performance of CLSM made with 70-80% MIBA. After having mixed the ash with 20-30% of cement, insignificant element concentrations in the leachate (determined in accordance with the TCLP) were observed in spite of the high total contents of Ba, Cr and Pb in MIBA. The authors suggested that the C-S-H microstructure entrapped the heavy metals thereby reducing their mobility and risk to the environment.

Hansson et al. (2012) carried out percolation leaching tests (CEN/TS-14405, 2004) on low strength mixes made with of 64% MIBA, 5% cement and 31% biofuel fly ash or peat fly ash. The results, plotted in Figure 5, show that, contrary to that observed in some of the previous studies, cement stabilized products may demonstrate higher mobility for specific elements than in comparison to unbound MIBA.

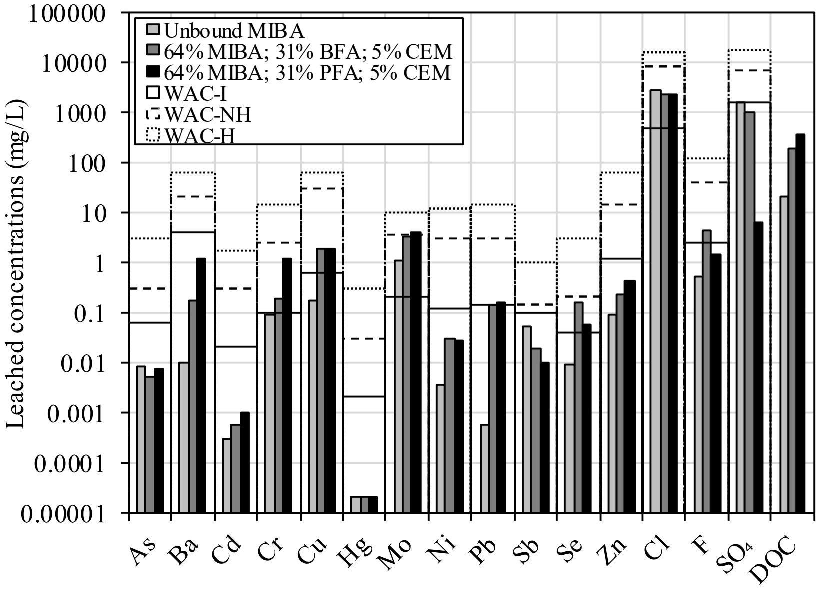

Figure 5 - Element concentration in leachate, at a l/s ratio equal to 0.1, of mixes with 64% MIBA, 31% biofuel or peat fly ash (BFA and PFA, respectively) and 5% cement (adapted from Hansson et al. (2012))

Furthermore, though lower concentrations of Sb and SO_4 were observed, mixing biofuel fly ash or peat fly ash with MIBA also led to higher leached concentrations of Ba, Cr, Cu, Pb, Se and Zn. According to EU criteria (CEU, 2003), S/S mixes would have to be categorized as borderline-hazardous waste on the account of high Mo concentrations, though such had not been observed in unbound MIBA.

Paine et al. (2002) and Dhir et al. (2002) assessed the influence of adding 40%, 70% and 100% of MIBA, as substitute for aggregate, in the leachability of cement treated mixes, designed in accordance with the UK Specification for Highway Works Series 800 (MCHW, 2016). The mixes, which contained cement contents of 2-10%, were subjected to availability testing, the element release of which would allow estimating the leaching behaviour during the road pavement's service life of 100 years. The leached concentrations complied with the EU criteria for drinking water.

Recently, a full-scale road was constructed in order to evaluate the effect of using MIBA in the leaching behaviour of road pavement layers (Toraldo and Saponaro, 2015, Toraldo et al., 2013). After submitting the ash to a 12-hour thermal treatment up to 1200 °C, 20% of it was used as natural aggregate substitute in the granular foundation, 20% in a cement-treated subbase and 10% in binder and base asphalt concrete. The results of batch leaching tests, in accordance with EN-12457-2 (2002), demonstrated the low leachability of the stabilized material when compared with the limits of the Italian Ministerial Decree (MD-186, 2006) for the reutilization of non-hazardous wastes. However, since many of the leached elements from the 10% MIBA-containing S/S material exhibited concentrations similar to or higher than those of the unbound mix with 20% MIBA, it is possible that the key impeding variable to the mobility of those contaminants was the high temperature treatment, rather than the cementitious binding.

5.3 Bitumen-bound MIBA

Studies found assessing the leaching behaviour of bituminous mixes incorporating MIBA have shown unanimous positive findings. Chen et al. (2008) produced hot-mix asphalt with 10-40% MIBA as substitute for aggregate and with 3.5-9.5% bitumen content. The leaching tests, in accordance with the TCLP (USEPA, 1990), showed very low concentrations of heavy metals, many of which below detectable levels. Eighmy et al. (1997) explained that the low leached concentrations of MIBA covered with asphalt are a result of the tortuous and hydrophobic nature of that binding system and of the particle’s surface cover, which isolates the material’s contact with water thereby decreasing infiltration and element mobility.

Huang et al. (2006) reported similar findings after having replaced 25%, 50% and 100% of the natural aggregate fraction of hot-mix asphalt with 4% bitumen content. Apart from the low leachability of MIBA-containing mixes, evaluated as per the TCLP (USEPA, 1990), the leachate concentrations were comparable to those of the control mixes thereby suggesting that the bitumen successfully isolated MIBA from water. Gress et al. (1992) also produced bituminous mixes with 25-100% MIBA as partial replacement for aggregate and 4-12% bitumen content and observed that all hazardous elements' concentrations were within limits.

In a full-scale road test track (Toraldo and Saponaro, 2015, Toraldo et al., 2013), the authors analysed the leaching behaviour of binder and base asphalt concrete mixes, with 4% bitumen content, 10% MIBA, previously subjected to the high temperature treatment. The elements leached from these mixes showed concentrations equivalent to or even lower than those of unbound or cement-stabilized mixes with 20% and 10% MIBA, respectively. Furthermore, all concentrations were within the limits proposed by the Italian Ministerial Decree (MD-186, 2006) suggesting that the risks to the environment are minimal when using up to 20% MIBA in asphalt concrete.

6 Ceramic products

Given the positive experiences on the technical feasibility of using MIBA in ceramic-related products, including tiles, bricks, glass ceramics and glass (Silva et al., 2017), it now becomes especially important to ascertain how these materials would behave in terms of their impact to the environment and thus further boost MIBA's assimilation by the ceramic industry and client's confidence on the materials' safety.

6.1 Glass

There have been several studies on the leaching behaviour of the glassy material resulting from MIBA's vitrification, which involves submitting it to high temperatures of 1400-1500 °C (Andreola et al., 2008, Barberio et al., 2010, Chiou et al., 2009, Karamanov et al., 2014, Kuo et al., 2006, Lam et al., 2010b, Lapa et al., 2006, Lin and Chang, 2006, Xiao et al., 2008). As previously stated, the energy-intensive treatment itself already points towards minimized leachability-related risks to the environment (Bergfeldt et al., 1997, Lapa et al., 2002, Wang et al., 2003), as a result of the significantly reduced mobility of heavy metals that have become entrapped in the glassy matrix (Saffarzadeh et al., 2009). Xiao et al. (2008) produced glass from the vitrification of MIBA and calculated the immobilization efficiency between the concentrations of leached elements from the slag and those from the untreated MIBA. With the exception of Fe, Mo, Ni and Pb, which presented immobilization rates of 73%, 60%, 72% and 80%, respectively, the immobilization rates of all other elements were over 90% (Figure 6). Others have also reported significantly reduced levels of leachability from vitrified MIBA (Chiou et al., 2009, Ecke et al., 2001, Lapa et al., 2006).

In the study of Lin and Chang (2006), vitrified MIBA samples, heated at 1400 °C for 30 min, were characterized in terms of their leachability, according to the TCLP (USEPA, 1990). The concentrations of Cd, Cr, Cu, Pb and Zn from the slag were significantly lower when compared with the untreated MIBA and much lower than the levels imposed by (USGPO, 2011). However, attention was given to the volatilization of some elements, such as Cd and Pb, above 1400 °C, and thus additional research would have to be carried out taking into consideration the gases generated from this process.

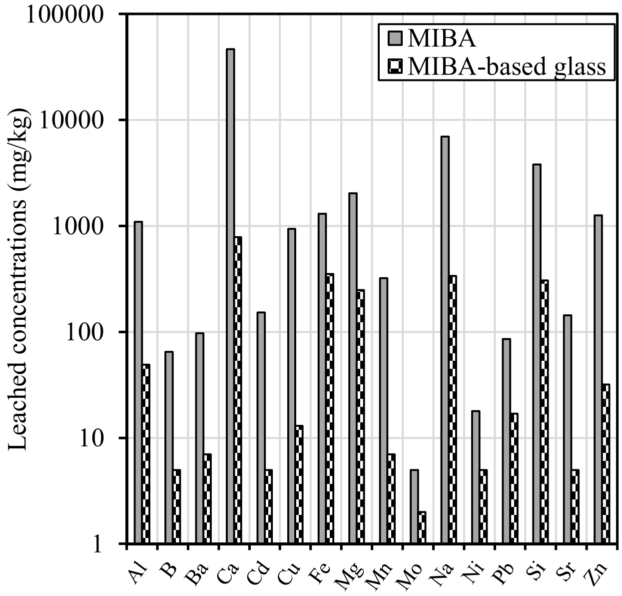

Figure 6 - Vitrification immobilization efficiency of elements leached from vitrified MIBA and untreated MIBA

(values sourced from Xiao et al. (2008))

6.2 Glass-ceramics

Within the specific thermal treatment from which glass-ceramics are derived, Qian et al. (2006) suggested that the production of diopside-based glass-ceramics containing MIBA may show high fixing capacity for heavy metals. Melting the ash, at 1500 °C for 30 min, alongside pondered quantities of MgO and Al_2O_3, and TiO_2 as the nucleating agent, followed by nucleation at 730 °C for 90 min, and crystallization at 880 °C for 10 h, the authors were able to produce a material with insignificant leaching of Cd, Cr and Pb.

Zhang et al. (2015) produced glass-ceramic samples out of a vitrified mixture of 80% oil shale fly ash and 20% MIBA, melted at 1500°C for 1 hour. The leaching test results, as per the TCLP, showed that the element mobility was unimportant when compared with the regulatory limits.

6.3 Ceramics

Cheeseman et al. (2003) sintered 100% MIBA at temperatures of 1080-1115 °C to ascertain the potential of the material for the production of ceramic based products. The sintered ceramics exhibited reduced ANC when compared to the fresh MIBA. However, when tested for their availability via an acidic aqueous solution, all sintered samples exhibited significantly reduced leached concentrations, with insignificant levels of Al, Cu, Pb and Zn at neutral pH levels. These positive results suggest the effective encapsulation of hazardous constituents in glassy and crystalline phases.

Barberio et al. (2010) analysed the effectiveness of using vitrified MIBA as frit for ceramic glazes. After having established the technical feasibility of using the material for this application, the authors carried out an LCA comparing the impacts of using MIBA for frit production with those of the conventional scenario, which is disposal in a landfill. The system boundaries of frit production included transport, recycling of aluminium and iron, production of the glaze frit, recycling of scraps to produce glass, whereas the boundaries of the landfill scenario included transport, landfill and treatment plant construction and operations, production of raw materials and of glaze frit. The results showed that the former scenario was more beneficial as it showed reductions ranging from 65% to 95% in the evaluated impact categories, including abiotic depletion, global warming potential, human toxicity, acidification, among several others.

Rambaldi et al. (2010) studied the possibility of using up to 5% MIBA in the manufacture of silicate-based tiles. Leaching trials, which were conducted in accordance with EN-12457-2 (2002), showed levels corresponding to inert waste as per Italian/European criteria (CEU, 2003).

Schabbach et al. (2012) incorporated 60% MIBA, previously subjected to an upgrading treatment, which included weathering, washing and separation by different particle sizes, in a mix with 40% refractory clay. After grinding the mix to a size below 75 µm and sintering it, the resulting ceramic products' leaching behaviour was evaluated according to EN-12457-2 (2002). The results, plotted in Figure 7, show that the original MIBA was categorized as a non-hazardous waste. After both fractions of MIBA were sintering with refractory clay, a notable decrease in mobility was observed for most elements, wherein ceramic materials made with the finer fraction exhibited greater leachability. Nevertheless, part of the decreased leached concentrations of some of the elements (e.g. Cd, Pb, Cl) was mainly due to their volatilization during the sintering process as previously explained. The ceramic samples made with the coarser MIBA fraction complied with the WAC for non-hazardous wastes on the account of the leached Cu content being slightly over the limit for inert wastes. Furthermore, similar to that observed by Sorlini et al. (2011), for washed and unwashed MIBA, smaller-sized fractions generally exhibited higher leached concentrations than coarser fractions.

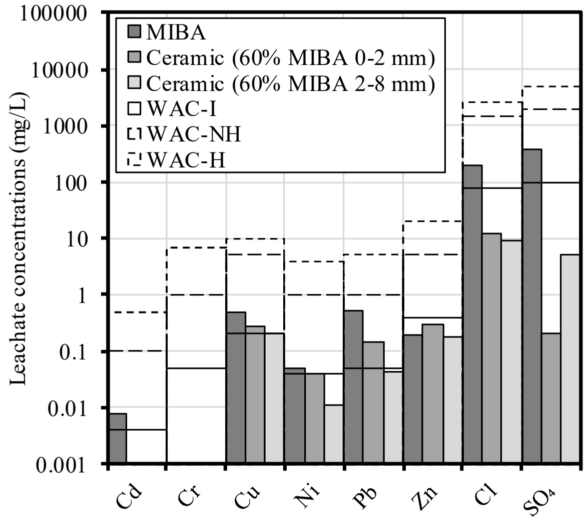

Figure 7 - Leaching behaviour of ceramic materials with MIBA of different sizes (adapted from Schabbach et al. (2012))

7 Conclusions

Environmental contamination is a key factor in the decision making of whether a new material can be incorporated in the construction industry. In spite of the proven technical feasibility of using MIBA in several applications, its environment related performance is a subject that is still being debated and is not widely known. Therefore, this paper seeks to enlighten, not only members of the scientific community, but also stakeholders of waste-to-energy plants and manufacturers of products capable of incorporating MIBA of their improved environmental performance in comparison with conventional counterparts. It is clear that redirecting MIBA from landfills, not only would the corresponding space be saved and contamination to nearby sensitive recipients would be prevented, but its use as raw material in the manufacture of value-added products would delay the depletion of natural resources.

Although the evaluation of the environmental impact of a material can be carried out in a number of ways, most researchers favoured the analysis of the MIBA-containing products' leachability in view of the potential heavy metal contamination; in spite of the application of LCA methodology to better understand the environmental performance of a given product, little emphasis has been given in this regard and future studies should focus on this approach. The following conclusions, which are separated by the use of MIBA in different applications, represent a compilation of the main findings of several studies in the literature.

The use of MIBA in the manufacture of artificial aggregates may result in a low resistance to acidic substances, which suggests that it would attain a lower pH level much faster and thus exhibit greater heavy metal release. To counteract this, if the application of MIBA-based artificial aggregates would involve exposure to an acidic environment, than its use would be preferred as natural aggregate substitute in cementitious systems, rather than using them in an unbound form. The reason for this is that their solidification with cement would maintain the material's alkalinity for longer and thus enhanced leaching behaviour.

Concerning the use of MIBA as substitute for natural aggregates in the production of cementitious composites, the results from several investigations infer that there are no added risks to the environment in comparison with composites made with conventional constituents. The high pH environment of cement-stabilized mixes normally results in a less leachable material, depending on the pH dependence of metal solubility, rather than when placed in acidic settings. However, there are some elements that also present high mobility in an alkaline environment; such critical heavy metals include Cu, Pb, Sb, and Mo. Nevertheless, cement-bound MIBA are likely to exhibit equivalent leached concentrations to conventional products without the ash. From a practical point of view, since both will probably belong to a similar category in terms of their leachability, MIBA-containing concrete and mortars can be used in a way that is similar to that of conventional mixes, assuming that these are not continuously in contact with water that can contaminate water bodies and groundwater systems.

As a result of MIBA's specific chemical composition and reasonably high amount of glassy phases, it can be used as partial replacement to cement, assuming that it is properly treated. The pozzolanic reactions between the amorphous SiO_2 and Al_2O_3 phases of MIBA with the cement's

$Ca(OH)_2$ results in additional products of hydration, especially AFm and AFt phases, which can effectively encapsulate hazardous elements into the cement. Additionally, since this would result in reduced surface area, there would be lower infiltration of water capable of removing those contaminants from the material when compared to the original unbound MIBA.

Regarding the manufacture of cement with the use of MIBA as raw feed component, apart from the resulting clinker's equivalent leached concentrations to those of ordinary Portland cement-based samples, the ash's inclusion in the manufacturing process can result in other environmental benefits, such as decreased CO_2 emissions.

The application of MIBA in base and subbase layers for road pavement construction is the only outlet for the ash in most countries and thus constitutes the subject matter with the greatest amount of research in terms of leaching behaviour. MIBA can be used in its granular form or hydraulically-bound, wherein the former may result in greater leachability when subjected to high rainwater infiltration and thus more likely to contaminate nearby sensitive recipients. Cement-bound MIBA layers, on the other hand, are likely to present enhanced behaviour from an environmental point of view, since heavy metal mobility throughout the road's service life is likely to be restricted due to their physical encapsulation within the cementitious microstructure. Although the use of MIBA in road construction was found to be more advantageous than sending the ash to a landfill, if the material's leaching behaviour is not adequately controlled, the released elements' quantities may be such that the benefits of using them are negated. Therefore, not only would leaching trials be required to ascertain the materials' behaviour, but drainage and rainwater collection systems must also be considered in the road construction so that the water is properly deviated from the MIBA-based layers. Another way of positively influencing the performance of those layers is the application of natural weathering or aging by accelerated carbonation treatments on the original MIBA prior to their use, which produces a more stable material with the formation of $CaCO_3$ and pH reduction to a more neutral level.

Existing findings on the use of MIBA in the construction of asphalt concrete layers were even more encouraging than the previous applications. Leached concentrations from bitumen-bound MIBA have been deemed insignificant, in some cases undetectable, and within regulatory levels for inert wastes. This effective immobilization is prompted by the binder's hydrophobic nature, which prevents water from infiltrating the material and removing its contaminants. However, despite the low environmental risks, after several years of wear, the road's surface layer may become deteriorated to a point that may increase leaching risks and thus well-thought out maintenance operations are necessary to prevent such manifestation.

In the production of ceramic products, the high temperatures involved were found to be very effective at restricting heavy metal release, as a result of the high densification of the material and the formation of new mineralogical species. Moreover, since ceramic products may present high durability, there is a minimum risk of accumulation of heavy metals, especially when used in construction-related applications, wherein most materials are protected rainwater infiltration throughout the structure's life cycle. Finally, apart from the reduced leachability, the heat-treated materials showed high homogeneity and consistent data, which may facilitate future certification of vitrified MIBA or ceramics products containing it and their commercialization.

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Figure captions

Figure 1 - Leachate concentrations of MIBA-based artificial aggregates (CFA - coal fly ash)

Figure 2 - Leachate concentrations from crushed S/S MIBA-containing mixes (Ginés et al., 2009) and from washed/unwashed MIBA (Sorlini et al., 2011); European waste acceptance criteria for inert (WAC-I), non-hazardous (WAC-NH) and hazardous (WAC-H) wastes for landfills

Figure 3 - Acid neutralization capacity of cement pastes with MIBA as partial cement replacement

Figure 4 - 24-hour cumulative leaching behaviour of CLSM mortars containing different amounts of MIBA as partial cement replacement (adapted from Zhen et al. (2012))

Figure 5 - Element concentration in leachate, at a l/s ratio equal to 0.1, of mixes with 64% MIBA, 31% biofuel or peat fly ash (BFA and PFA, respectively) and 5% cement (adapted from Hansson et al. (2012))

Figure 6 - Vitrification immobilization efficiency of elements leached from vitrified MIBA and untreated MIBA (values sourced from Xiao et al. (2008))

Figure 7 - Leaching behaviour of ceramic materials with MIBA of different sizes (adapted from Schabbach et al. (2012))

Table captions

Table 1 - Compilation of test methods, mix design and main findings on the leachability behaviour from the literature

Table 2 - Leachate concentrations of MIBA-based clinker (values sourced from Lam et al. (2010a))

Table 3 - Cumulative (1-year) releases of hazardous elements (values in bold are above the limits for inert waste according to European WAC (CEU, 2003); values sourced from Izquierdo et al. (2008)