



Effect of short-term natural weathering on MSWI and wood waste bottom ash leaching behaviour

M. Gori^{a,*}, B. Bergfeldt^b, G. Pfrang-Stotz^b, J. Reichelt^c, P. Sirini^a

^a Department of Civil and Environmental Engineering, University of Florence, Florence 50139, Italy

^b Karlsruhe Institute of Technology, P.O. Box 3640, D-76021 Karlsruhe, Germany

^c IBR, Obergrömbacher Straße 29, D-76646 Bruchsal, Germany

ARTICLE INFO

Article history:

Received 3 May 2010

Received in revised form 15 February 2011

Accepted 17 February 2011

Available online 5 March 2011

Keywords:

MSWI

Wood waste

Bottom ash

Ageing

Weathering

ABSTRACT

Short term natural weathering was applied on municipal solid waste (MSW) and wood waste incinerator bottom ash (BA). The materials were analysed at different steps of treatment and characterized for chemical and mineralogical composition. Both short and long term leaching behaviour of main elements and heavy metals were investigated as well.

Lead, zinc and copper were the main heavy metals to be released. After 12 weeks of treatment the concentration of leached zinc decreased. Lead concentration was not found to be influenced by pH and decreased only for the biomass samples. Weathering did not have beneficial effects on copper leaching, which was well described by complexation processes with DOC.

The findings from the experimental campaign indicated that weathering reactions improved the mineral stability of the analysed materials but, in contrast with previous works, the treatment was not sufficient to guarantee pH stability and to comply with leaching law limits.

© 2011 Elsevier B.V. All rights reserved.

1. Introduction

In its waste policy and legislation the European Commission has set out a clear hierarchy of waste management options. The hierarchy, declared in the Sixth Environmental Action Plan [1], envisages first waste prevention, then recovery (reuse, material and energy recovery), and finally disposal, that is landfill and incineration without energy recovery. In this regard, diverse studies have been performed in order to evaluate reuse opportunities in civil engineering field for different residual by-products [2–8].

Thermal process residues are important waste streams with regards to utilisation. The most important are MSWI BAs, ashes from coal-fired power stations [9], slags from metallurgy [10,11] and biomass ashes.

Incineration of MSW has an important role in waste management in many industrialised countries. Incineration in modern waste-to-energy plants reduces the volume of MSW by up to 90%, but it also produces by-products such as BA and fly ash (FA), in amount of about 33 wt.% of incinerated waste [12–14]. BA is the most significant residual by-product from the combustion of MSW. It accounts for 85–95% of all the residues produced during combustion [15,16]. In many European countries, especially Germany, The

Netherlands, France and Denmark, a significant amount of MSWI BA is reused as secondary building material, in road sub-bases and in the construction of embankments.

Biomass incineration is a proven technology for heat and power production. Due to growing pressures on global environment and energy security, a significant trend towards the thermal utilisation of biomass has been recognised in Europe within the last years [17–19]. The main advantage of energy production from biomass is the CO₂ neutrality [18–20].

Wood waste is a low-cost biomass fuel and therefore it is very attractive for large-scale biomass combustion plants. Wood waste comprises demolition wood, pallets, fibre boards, residues from the wood processing industry, railway sleepers, pylons, etc. Due to these different sources, waste wood is an extremely inhomogeneous fuel and, therefore, its chemical composition, as well as its content of impurities, can vary significantly [17].

Currently, most of the biomass ash is either disposed of in landfill or applied to soils, thereby recycling nutrients such as Ca, K, Mg and P [6,18,19]. However, considering that the disposal costs of biomass ashes are raising, and that biomass ash volumes are increasing worldwide, a sustainable ash management has to be established [18]. The possible utilisation of biomass FA in civil engineering field has been investigated by several authors [18,21–23]. Studies on utilisation routes for BA from biomass combustion plants are, however, very limited.

For residues to be recycled, their short and long-term behaviour, concerning both environmental and technical aspects, must be

* Corresponding author. Tel.: +39 055 4796317; fax: +39 055 4796494.

E-mail address: manuela.gori@dicea.unifi.it (M. Gori).

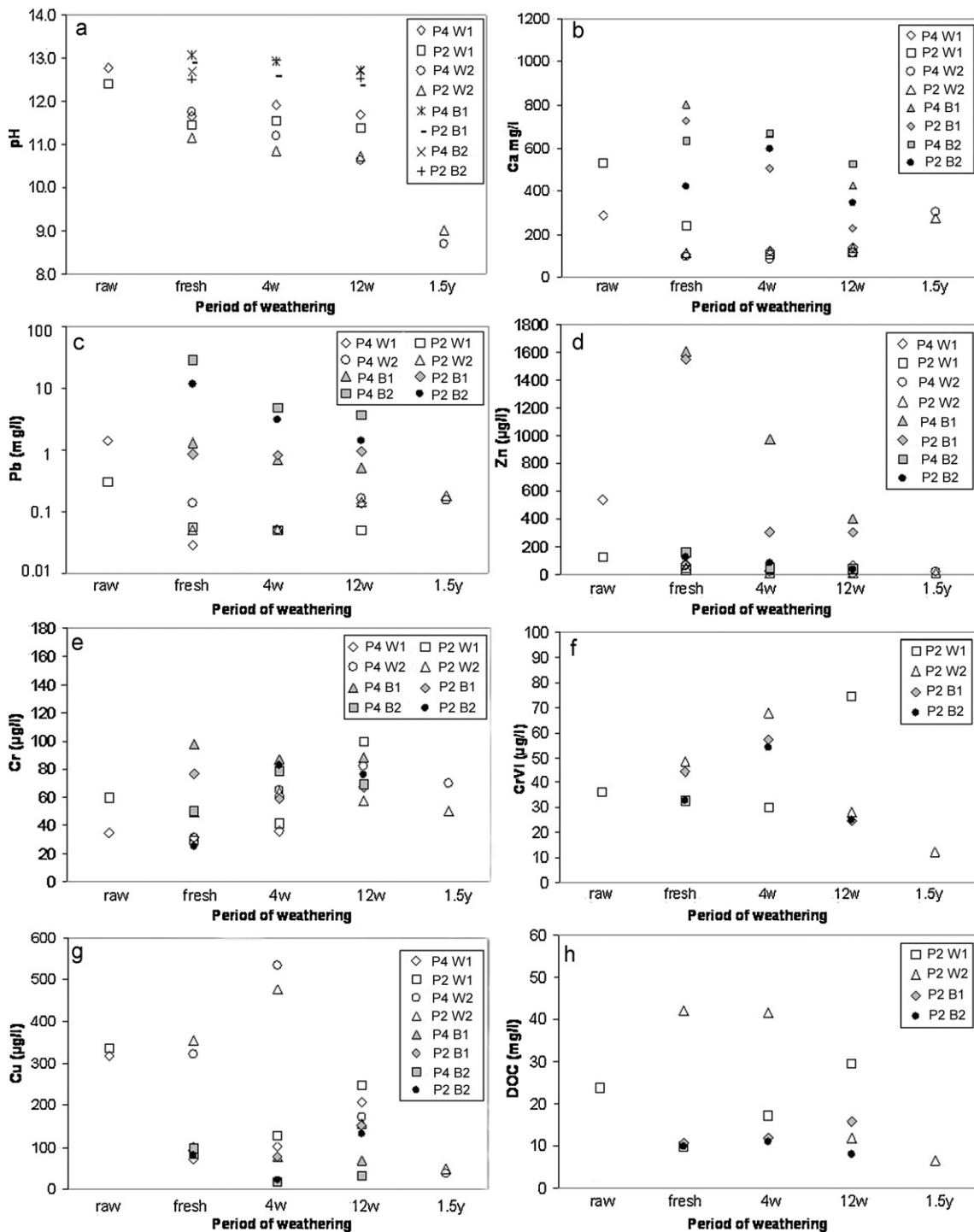


Fig. 1. pH (a) and leaching concentrations of Ca (b), Pb (c), Zn (d), Cr (e), Cr^{VI} (f), Cu (g) and DOC (h) from natural weathered BA samples. Abbreviations: P4, EN 12457-4 leaching test and P2, EN 12457-2 leaching test.

evaluated [24]. In particular, the leaching of contaminants from BA has received a growing attention over the years and many leaching tests have been designed by regulatory agencies in order to comprehend the mechanisms of trace elements mobility and to simulate field leaching scenario [24–31].

In order to comply with limits set by law for leaching, several treatment options are available. In case of thermal process residues, solidification, stabilisation, vitrification, classification by granulometric size particles and ageing or weathering can be applied. The choice of the treatment depends on site specific conditions, utilisation

or disposal objectives and law requirements [29]. When an ash treatment method must be chosen, much attention must be paid to investments and operating costs. In EU countries natural weathering of BA for 6–12 weeks after quenching is the most commonly employed method [29,32,13,33–35].

BA shows a complex mineralogical composition consisting, mainly, of silicates, oxides and carbonates, as well as secondary phases like amorphous glass phases, iron metals and feldspars and trace substances as metals, alloys, sulphates and salts [36]. Most of the high-temperature solids contained in incinerator

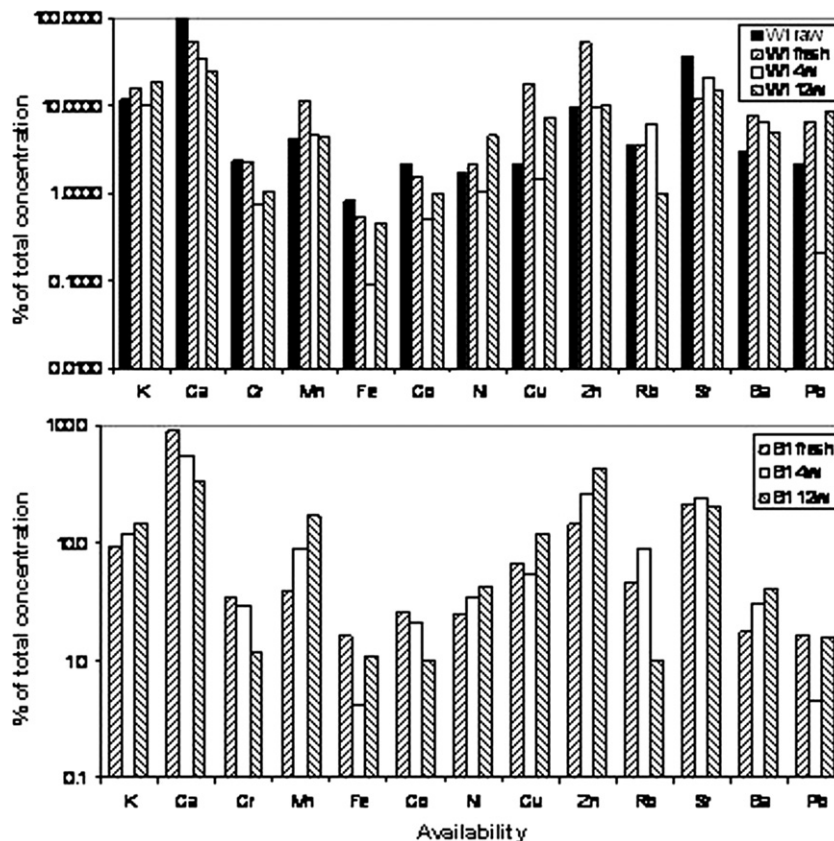


Fig. 2. Availability (NEN 7371) of main elements and metals normalized to total concentration for sample W1 (a) and B1 (b).

Table 4

Comparison of data from EN 12457-4 from MSWI BA 12 weeks from 2004 (values from 10 different MSWI plants [8]) and W1 and W2 12 weeks.

	MSWI BA (2004) Mean	MSWI BA (2004) Minimum	MSWI BA (2004) Maximum	W1	W2
pH	10.9	9.3	11.8	11.7	10.6
C _{Tot} (μg/l)	40	<10	124	124	82
C _{r^{VI}} (μg/l)	35	<10	105	74	28
Ni (μg/l)	<10	<10	<10	55	30
Cu (μg/l)	296	26	694	208	173
Zn (μg/l)	18	<10	44	59	<10
As (μg/l)	<10	<10	<10	<10	<10
Cd (μg/l)	<10	<10	<10	<4	<4
Hg (μg/l)	0.16	<0.1	0.3	<0.2	<0.2
Pb (μg/l)	12	<10	34	128	162
Cl (mg/l)	134	30	241	381	239
SO ₄ ²⁻ (mg/l)	277	81	547	139	348

BA are metastable under atmospheric conditions because they are formed at high temperatures in the combustion chamber and then rapidly cooled via a quench tank. Therefore, under ambient conditions, and especially during the first three months

of weathering, they undergo further mineralogical processes [29,36–38].

Weathering consists of several interrelated processes. Meima and Comans [37] suggest that the main alteration processes con-

Table 5

Comparison of data from EN 12457-4 from industrial by-products [8] with B1 and B2 12 weeks stored (n.a. not analysed).

	Coal fly ash	Electric arc furnace slag	High temperature slag	Recycling material	Boiler sand	B1	B2
pH	12.2	11.2	8.5	11.6	10.0	12.7	12.7
C _{Tot} (μg/l)	381	67	15	25	16	88	69
Ni (μg/l)	n.a.	2	10	5	1	33	<30
Cu (μg/l)	24	29	10	16	27	100	32
Zn (μg/l)	4	3	13	2	5	403	42
As (μg/l)	26	24	3	6	4	<10	<10
Pb (μg/l)	15	19	28	18	17	513	3640
Cl (mg/l)	5.4	0.9	0.5	5	7	44	5
SO ₄ ²⁻ (mg/l)	1050	5	0.6	47	72	47	2

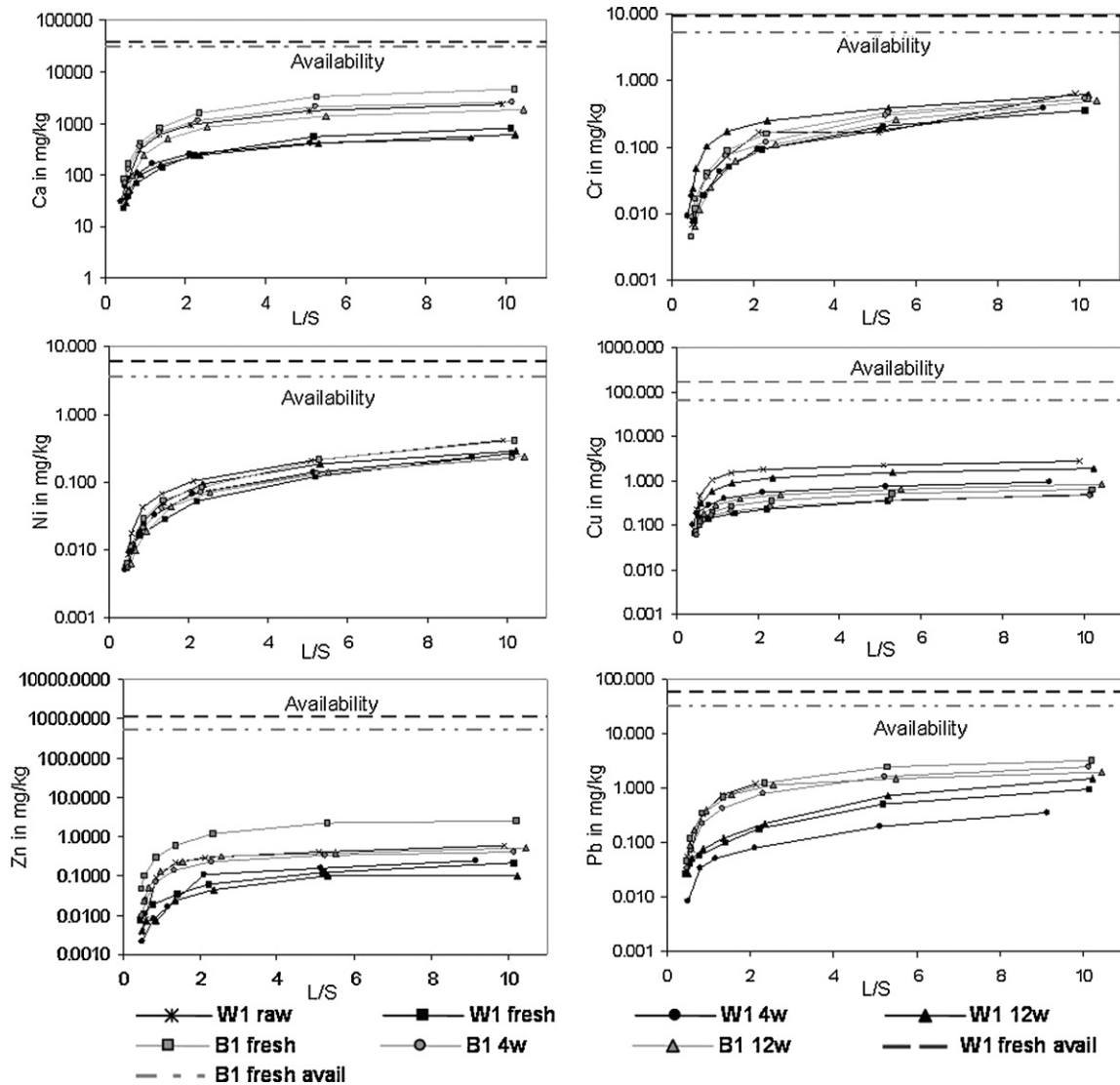


Fig. 3. Availability and elution (NEN 7341) of Ca (a), Cr (b), Ni (c), Zn (d), Cu (e) and Pb (f).

sist of the hydrolysis of cations, carbonation and formation of clay-like minerals from glasses. During the carbonation process, atmospheric CO_2 is absorbed by the initially alkaline BA; the calcite (CaCO_3) thus formed precipitates until the material is in equilibrium with atmospheric CO_2 . pH decreasing causes also $\text{Al}(\text{OH})_3$ and amorphous aluminosilicates to precipitate because Al solubility is strongly reduced with the pH decrease from 10 to 8–8.5 [39].

The chemical and mineralogical changes caused by such processes greatly affect metals solubility as well as their speciation.

Different studies have been performed on MSWI BA weathering processes [16,29,32,37,38,40–44]. However, few data are currently available on the effect of this treatment on wood waste BA.

This paper presents the results of an experimental study carried out to evaluate the effect of short term natural ageing applied to MSW and wood waste incinerator BA. The relationship between pH values and the leaching behaviour of the main trace elements are investigated, paying attention to the mechanisms of weathering and chemical reaction involved.

A previous study conducted in the years 2002–2004 by this research group [8,36] investigated the mineralogical, environmental relevant and structural engineering properties of MSWI BA from 10 different MSWI plants in Germany and of other industrial by-products, such as coal FA and boiler sand from stone coal com-

bustion, electric arc furnace slag and high temperature slags from iron and steel production and recycling material from road destruction. In that study BAs were sampled and stored for 12 weeks to investigate the regional influence on waste input and the influence of technical variations on the BA quality.

It was expected that the quality of the input and the output of MSWI plants would change after the implementation of the European Landfill Directive [45]. Five years after the implementation, leaching data of the 2004 study and data from 2009 were compared. These data are included in this paper to point out whether there are differences between recent and literature results quoted in papers written before 2005 due to changes in legislation or plant parameters. Since BA from biomass combustion plants will be a more and more important residue stream, leaching data were compared, too.

2. Materials and methods

Fresh BA samples were collected from four different grate furnace incineration plants, situated in the south of Germany. Two plants treat MSW and respective samples are indicated as W1 and W2. The other two incineration facilities treat wood waste. In Germany wood waste is classified according to the guideline BGBl. I 2002/3302 into classes A1, A2, A3, A4 and PCB-waste wood. Class A1

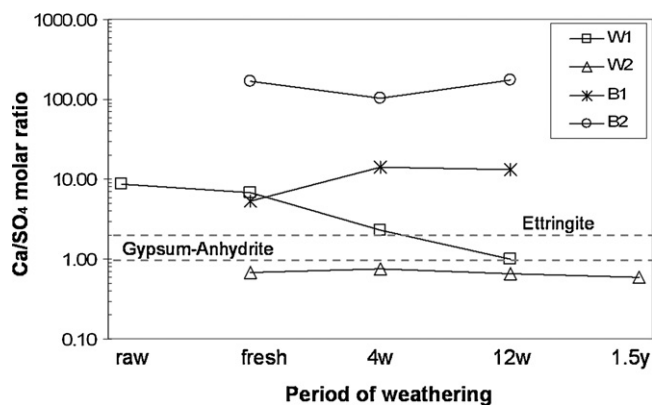


Fig. 4. Kinetics of change of the Ca/SO₄ molar ratio.

is for waste of untreated wood; class A2 is for waste timber which has been treated (e.g. glued, coated or painted) without halogen organic compounds in the coating and without wood preservatives; class A3 is for waste timber with halogen organic compounds in the coating and without wood preservatives; A4 is for waste timber treated with wood preservatives, such as sleepers, poles, which cannot be classified in one of the previous classes. The first biomass incineration facility treats A1 and A11 categories wood waste (samples are indicated as B1), while the other treats A1, A11 and A1V wood waste (samples are indicated as B2). In all considered plants the hot BA is quenched in a water tank, immediately after incineration. Follow-up treatments include the removal of large pieces of unburned materials, the separation of magnetic particles and size reduction by crushing. All these treatments require about three weeks. Subsequently, BA is posed in large storage piles for at least three months before its reuse, in order to allow weathering reactions to take place. Due to heterogeneity of the BA, samples were collected from several points and from different depths (about 20 different points), in an amount of ca. 50 kg, in order to assure the representativeness of the samples. Samples were taken from the new-created pile and then after 4 and 12 weeks. For sample W1 it was possible to take also a raw sample before pre-treatments, while for sample W2 a 1.5 years old weathered sample was taken too. Because of technical problems, the weathering process of sample W2 and B2 was conducted for 4 weeks in the open air and for the other 8 weeks in laboratory, where humidity was monitored and maintained approximately constant through the addition of demineralised water.

BA moisture was determined by drying subsamples at 105 °C for 24 h. To detect elemental composition, samples were milled and digested using a HNO₃/HF mixture in a Teflon bomb heated in a microwave oven.

For each sample the fraction 0.063–0.09 mm was isolated through sieving and then examined by X-ray diffractometry to identify the mineralogical composition.

Concerning the leaching characteristics, the EN 12457-2 and the EN 12457-4 procedures were used as regulatory tests. The two tests differ each other only for the particle size of the tested materials. Italian law refers to EN 12457-2, while German law refers to EN 12457-4 batch test. In order to study the long term leaching behaviour of the considered materials, the availability test (according to NEN 7371) and a column test (according to EN 14405) were carried out. The availability test is a pH controlled extraction test, which allows the estimation of the fraction of the total concentration present, which can be leached when the waste material is exposed to extreme conditions, as in the very long-term. Because of the test conditions, it provides an upper limit to the leaching potential of the analysed materials [46]. Column test, instead, pro-

vides information about the time dependent leaching behaviour: analysis of the eluates at defined ratios between the volume of the eluate and the quantity of the tested material (in the range 0.1–10 g eluate/g tested material) can be considered representative of the elution trend in time [47].

The resulting solutions were filtered and the pH was measured. Filtrates were divided into two samples. One sample was acidified with concentrated HNO₃ and used for the analysis of the following heavy metals in the leachate: Cr, Mn, Fe, Co, Ni, Cu, Zn, Rb, Sr, Ba, Pb and As by Total Reflecting X-ray Fluorescence Analysis (TXFA) and Cd, Cr^{VI}, Hg, Mo, Sb and Se by ICP-OES. The second sample was left untreated and subjected to ion chromatography in order to detect the concentration of chlorides and sulphates. Samples from EN 12457-4 and EN 12457-2 were also analysed for DOC, AOX, COD, phenol index and cyanide according to standard methods [48].

3. Results and discussion

The chemical composition of the examined materials, reported in Tables 1 and 2, indicates that the main trace elements were Ti, Cr, Co, Cu, Zn, Ba, Pb and Ni. Cu, Ni and Cr, because of their lithophilic behaviour [38,49]. Zn and Pb, instead, which are more volatile elements, are usually enriched in the FA and acid gas scrubbing residues [49].

The significant variations in metal concentration occurring for different metals within the same BA samples were attributed to the high heterogeneity of the material.

The effects of natural weathering were monitored for each sample during the entire duration of the experiment by means of the leaching tests. Fig. 1a–h shows the experimental results for each leachate sample. In the following discussion mineral solubility was calculated on the basis of thermodynamic stability constants.

The results of the EN 12457-2 and EN 12457-4 compliance tests showed that all the analysed samples were alkaline, with a pH value ranging between 11.2 and 11.8 for the waste BA and 12.5 and 13.1 for the biomass BA.

The initial alkaline pH of BA can be imputed to the presence of alkaline and alkaline earth elements and metal oxides, which form hydroxides during hydrolysis. The presence of Ca(OH)₂ in BA is due to calcination reactions involving calcite, gypsum (CaSO₄·2H₂O) or anhydrite (CaSO₄) to generate CaO, CO₂ or SO₂. Later, during quenching, the majority of the CaO is hydrolysed to the corresponding hydroxide [19,29,37,38,40]. XRD analysis data are reported in Table 3. According to several authors [37,50] calcite, gypsum and anhydrite were detected in fresh quenched BA, showing that the calcination process was not complete. Nevertheless, these minerals have very low solubility at a pH greater than 12 [29].

Concerning the W1 and W2 fresh samples, if the solution pH were controlled by Ca(OH)₂, the pH would be approximately 12.3. Since the solutions pH were lower (between 11.2 and 11.8), the solutions were undersaturated with respect to the aforementioned phase [51]. The W1 raw sample, instead, had a pH of 12.4–12.8 and the eluates could be slightly oversaturated with respect to Ca(OH)₂. These pH values, however, could be due to the presence of NaOH and KOH.

After 12 weeks, the W1 sample still had a pH between 11.4 and 11.7 and the leachates were undersaturated in portlandite. This results, coupled with data from XRD analysis (Table 3), suggest that no significant amounts of Ca(OH)₂ were present in BA.

The pH value of the W2 sample after 12 weeks ranged between 10.64 and 10.73. According to geochemical modelling from Poletini and Pomi [38], a pure system containing the minerals gibbsite (Al(OH)₃), gypsum and ettringite has at the equilibrium a pH value of about 10.33, which is close to the value detected in the present study. After 1.5 years of treatment, a pH value equal to 8.7–9.0 was

detected. This value could be reasonably imputed to the presence of calcite (CaCO_3). The completeness of the weathering process is typically characterized by pH 8.5, when the precipitation of calcite occurs and the material is in equilibrium with atmospheric CO_2 [37,38,52].

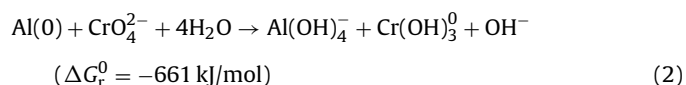
Wood waste samples had very high initial pH (between 12.9 and 13.1 for sample B1 and between 12.5 and 12.7 for sample B2). The high pH values and the high concentration of Ca in the leachates from both the aforementioned samples confirmed the assumption that the pH was controlled by the $\text{Ca}(\text{OH})_2$ solubility. Moreover, because of the Ca content and the Ca hydroxide solubility within the detected pH range, the eluates were oversaturated. Even if XRD analysis did not highlight the presence of NaOH and KOH, traces of these alkaline species could as well have contributed to the high final pH values. The pH values of samples B1 and B2 were still very high at the end of the ageing period.

The solubility of lead was quite constant for both the waste BA samples, despite the pH decrease. By contrast, a significant decrease of Pb was detected for both the wood waste BA samples, in particular the B2 sample, which was characterized by a pH value almost constant during the whole weathering period. The relative independence of the Pb leaching from pH led to exclude solubility control by a pure mineral phase, rather indicating the occurrence of sorption mechanisms [38,41,53]. Consistently with this assumption, the dissolved Pb concentrations observed in all the samples showed an oversaturation with respect to lead hydroxide ($\text{Pb}(\text{OH})_2$), and an undersaturation with respect to anglesite (PbSO_4), cersusite (PbCO_3) and hydrocersusite ($\text{Pb}_3(\text{CO}_3)_2(\text{OH})_2$). Similar findings have been reported by other authors [29,41,54,55]. Meima and Comans [56] as well as Poletini and Pomi [38] assumed that the low Pb leaching for naturally aged bottom ash could be explained by the strong affinity of Pb towards surface complexation onto Fe and Al (hydr)oxides. Dijkstra et al. [57] found that Pb behaviour was well described by surface complexation to Fe and Al-(hydr)oxides up to pH 10. Above this pH value the concentration of Pb corresponded better to those predicted by the solubility of pure $\text{Pb}(\text{OH})_2$.

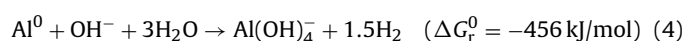
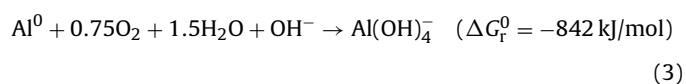
Cr (under the form of Cr^{VI}) tends to behave as an oxyanion in alkaline conditions [34]. According to Sabbas et al. [33] and Cornelis et al. [58], oxyanion forming elements exhibit similarities in their leaching trend, with a leaching minimum found around pH 12 and sometimes also at pH < 6. As shown in Fig. 1, Cr^{VI} was the prevalent species. Cr^{III} compounds, in fact, are sparingly soluble and hence, predominantly Cr^{VI} is generally found in BA leachates due to their oxidation and high pH characteristics [53,58,59].

According to Van Gerven et al. [34], the formation of carbonates with the proceeding of the weathering treatment should reduce the leaching of Cr [34]. Moreover, different authors indicated that Al^{O} present in the residues can control Cr leaching by reducing Cr^{VI} [59–61].

Cr present in BA as Cr^{VI} , once released to solution (Eq. (1)), can be reduced to Cr^{III} by Al^{O} (Eq. (2)):



But Al^{O} can be oxidized also by oxygen (Eq. (3)) and water (Eq. (4)):



All these three Al^{O} oxidation reactions have a very favourable Gibb's free energy. As calculated according to Stumm and Morgan [62], however, oxidation of Al^{O} by O_2 may be more favourable than that by Cr^{VI} , while oxidation with water may be the less favourable. As a consequence, during stabilisation and sample handling Al^{O} reduction capacity is diminished due to oxidation by O_2 , with a creation of a surface layer of Al-oxides on the Al^{O} particles [59–61]. This could be one of the factors causing Cr leaching to increase as a result of weathering.

The enhancement of Cr leaching with weathering observed in this study confirmed the results of previous researches [34,63–65].

Zinc concentration trend showed a continuous decrease in all the samples. All leachates resulted to be strongly undersaturated with respect to zincite (ZnO) and Zn hydroxide ($\text{Zn}(\text{OH})_2$). Geochemical modelling from Meima and Comans [41] indicated that almost all Zn-minerals in the database were too soluble to control Zn leaching. According to Meima and Comans [41,56], Dijkstra et al. [54,57] and Poletini and Pomi [38] the leaching of Zn could be explained with either surface precipitation or surface complexation mechanism of Zn onto BA phases, including Fe and Al (hydr)oxides.

According to several authors [29,32,37,41,54], the leaching of Cu from fresh MSWI BA does not occur via a concentration-limiting mechanism. Furthermore, the dissolved Cu in leachates is strongly influenced by the complexation with dissolved humic and fulvic acids, the major reactive components of DOC in MSWI BA leachates. The high degree of complexation of Cu with DOC is caused by the higher affinity of Cu for specific binding to humic substances, if compared to Ni, Zn and Cd [54,55]. As regards the different Cu species, oversaturation for copper hydroxide ($\text{Cu}(\text{OH})_2$) and undersaturation for copper carbonate (CuCO_3) were observed for the MSWI BAs. On the contrary, wood waste BAs showed Cu concentrations varying between undersaturation values or values close to $\text{Cu}(\text{OH})_2$ solubility; Cu content was anyway almost constant for the entire weathering process. The Cu concentration trend was always similar to that of DOC, thus confirming that the Cu solubility could be explained by the aforementioned complexation processes with DOC. The leachability decrease of Cu and DOC observed for samples W2 and B2 could be explained by the adsorption of natural humic substances and Cu complexes onto neo-formed aluminium (hydro)oxides [16].

Fig. 2 shows the concentration of major elements of W1 and B1 samples at different weathering stages. The experimental results were not easy to interpret, also because the metals concentration detected in the eluates from the availability test were often very near to the instrument detection limit. The diagrams show that only the availability of Ca, Cr, Co and Rb decreased during the weathering treatment. This could be explained by the presence of more stable minerals. In W1 sample Ni and Cu increased, while for sample B1 the mobility of Ni, Cu, Mn, Zn and Ba was enhanced. Significant changes were not observed for the other analysed elements.

Fig. 3 reports the results of column and availability tests for Ca, Cr, Ni, Zn, Cu and Pb for samples W1 and B1; concerning the availability test, only the results of the fresh materials are reported.

Column tests data showed an almost stable Ca leaching. The curves slope, which was almost constant, confirmed that, even for the aged samples, leaching of Ca was not complete at a L/S of 10. A similar behaviour was observed in the other samples.

As shown in Fig. 4, a significant difference was observed for the Ca/SO_4 leaching ratio. For all the samples, except the W2 one, a high Ca/SO_4 molar ratio was detected at the beginning of weathering if compared to gypsum and ettringite, thus indicating that calcium was provided primarily by non-sulphated species [42]. After 4 weeks, a molar ratio of about 2 was observed in sample W1, showing a preferential release of calcium and sulphates by ettringite. The molar ratio value was about 1 after 12 weeks, showing that leaching was mainly controlled by sulphates, in particular anhydrite and

gypsum. Sample W2 maintained a stable Ca/SO₄ ratio of about 1 for the whole ageing period, probably because of a significant presence of anhydrite. On the contrary, wood BA samples, were always characterised by high Ca/SO₄ ratio values, probably due to the high content of portlandite, even after 12 week.

For the other analysed elements, the column test confirmed the EN 12457-2 and EN 12457-4 leaching tests results: only Zn seemed to be significantly affected by the weathering treatment. An overall slight decrease of Zn in the leachates was observed as well as a tendency of the aged material to first complete the leaching.

As regards the comparison of the data from this study with that of 2004, Table 4 shows that the leaching results from samples W1 and W2 were mainly in the range of the older BAs. Differences could be seen for Pb, which was found in significantly higher concentration in the leaching solutions from W1 and W2. Also higher concentrations could be found for Cl and Ni. Due to the fact that at present all MSW has to be treated by thermal processes, the authors expected higher input streams in the MSWI plants and therefore problems to keep the quality of the residues on a high level. It can be stated from our results that this was true for Pb, Cl and Ni solubility.

Table 5 reports the comparison of the leaching data from biomass BAs with that from other residual by-products investigated in 2004. On the first view the difference of pH value was significant. The high solubility of Pb from B1 and B2 would cause problems for utilisation even after 12 weeks storage. Also rather high were the concentrations of Cr, Cu and Zn, especially in B1 leachates.

4. Conclusions

The present study showed the effect of short-term natural weathering treatment on the mineralogical and chemical properties of MSW and biomass BA, on the mineral phases, pH value, and leaching characteristics.

Lead, zinc and copper were the main heavy metals to be released. After 12 weeks of weathering treatment, the concentration of leached Zn fell dramatically, while only in the biomass samples a Pb decrease was observed. Pb concentration was not found to be influenced by the pH values of the BA. Leachates were oversaturated with respect to Pb hydroxide and undersaturated if compared to anglesite, cerrusite and hydrocerrusite. Pb solubility trends could be imputed to sorption processes. Cu leaching was well described by complexation processes with DOC. For the other contaminants no beneficial effects in the leachate composition were observed. Availability and column leaching tests confirmed the above mentioned results.

The experimental results showed that the weathering reaction improved the mineral stability even if, in contrast with previous works, the applied 12 weeks storage resulted insufficient to guarantee the complete stability of the material and to comply with leaching law limits.

Acknowledgements

Authors would like to thank Monika Schleinkefer and Patrick Hammer from KIT for the precious work in analysis with ion chromatography and TXFA.

References

- [1] EPC, The European Parliament and the Council of the EU, Decision no 1666/2002/EC of the European Parliament and of the Council of 22 July 2002 laying down the Sixth Community Environment Action Programme. Official Journal of the European Communities, 10.9.2002.
- [2] K.M. Skarzynska, Reuse of coal mining wastes in civil engineering. Part 1. Properties of minestone, Waste Manage. 15 (1995) 3–42.
- [3] K.M. Skarzynska, Reuse of coal mining wastes in civil engineering. Part 2. Utilization of minestone, Waste Manage. 15 (1995) 83–126.
- [4] J.O.V. Tränkler, I. Walker, M. Dohmann, Environmental impact of demolition waste—an overview on 10 years of research and experience, Waste Manage. 16 (1996) 21–26.
- [5] V.W.Y. Tam, C.M. Tam, A review on the viable technology for construction waste recycling, Resour. Conserv. Recy. 47 (2006) 209–221.
- [6] L. Reijnders, Disposal, uses and treatments of combustion ashes: a review, Resour. Conserv. Recy. 43 (2005) 313–336.
- [7] G. Pfrang-Stotz, J. Reichelt, Chemical composition and leachability of municipal solid waste incineration (MSWI) bottom ash, in: Proceeding International Conference on Incineration and Thermal Treatment Technologies, May 12–16, Oakland, California, 1997.
- [8] G. Pfrang-Stotz, J. Reichelt, Einfluss geänderter Stoffströme in der Abfallwirtschaft auf die zukünftige Qualität und die Verwertungsmöglichkeiten von Müllverbrennungsschlacken, Wissenschaftliche Berichte FZKA 7025 (2005) 301–307.
- [9] European Association for Use of the By-Products of Coal-Fired Power-Station (ECOBA), <http://www.ecoba.com>.
- [10] L. Tiruta-Barna, E. Benedetto, Y. Perrodin, Environmental impact and risk assessment of mineral wastes reuse strategies: review and critical analysis of approaches and applications, Resour. Conserv. Recy. 50 (2007) 351–379.
- [11] The European Slag Association (EUROSLAG). Legal status of slags. Position Paper, January 2006. <http://www.euroslag.org>.
- [12] C.R. Cheeseman, A. Makinde, S. Bethanis, Properties of lightweight aggregate produced by rapid sintering of incinerator bottom ash, Resour. Conserv. Recy. 43 (2005) 147–162.
- [13] J.M. Chimenos, A.I. Fernandez, L. Miralles, J.R. Rosell, A. Navarro Ezquerria, Change of mechanical properties during short-term natural weathering of MSWI bottom ash, Environ. Sci. Technol. 39 (2005) 7725–7730.
- [14] M. Ferraris, M. Salvo, A. Ventrella, L. Buzzi, M. Veglia, Use of vitrified MSWI bottom ashes for concrete production, Waste Manage. 29 (2009) 1041–1047.
- [15] E. Rendek, G. Ducom, P. Germain, Influence of organic matter on municipal solid waste incinerator bottom ash carbonation, Chemosphere 64 (2006) 1212–1218.
- [16] M. Rosende, M. Miró, V. Cerdà, The potential of downscaled dynamic column extraction for fast and reliable assessment of natural weathering effects of municipal solid waste incineration bottom ash, Anal. Chim. Acta 619 (2008) 192–201.
- [17] S. Van Loo, J. Koppejan, The Handbook of Biomass Combustion and Co-firing, Earthscan, UK, 2008.
- [18] R. Rajamma, R.J. Ball, L.A.C. Tarelho, G.C. Allen, J.A. Labrincha, V.M. Ferreira, Characterisation and use of biomass fly ash in cement-based materials, J. Hazard. Mater. 172 (2009) 1049–1060.
- [19] B.-M. Steenari, L.G. Karlsson, O. Lindqvist, Evaluation of the leaching characteristics of wood ash and the influence of ash agglomeration, Biomass Bioenergy 16 (1999) 119–136.
- [20] M. Narodoslawsky, I. Obernberger, From waste to raw material—the route from biomass to wood ash for cadmium and other heavy metals, J. Hazard. Mater. 50 (1996) 157–168.
- [21] E. Tkaczewska, J. Małolepszy, Hydration of coal-biomass fly ash cement, Constr. Build. Mater. 23 (2009) 2694–2700.
- [22] S. Wang, E. Llamazos, L. Baxter, F. Fonseca, Durability of biomass fly ash concrete: freezing and thawing and rapid chloride permeability tests, Fuel 87 (2008) 359–364.
- [23] S. Wang, A. Miller, E. Llamazos, F. Fonseca, L. Baxter, Biomass fly ash in concrete: mixture proportioning and mechanical properties, Fuel 87 (2008) 365–371.
- [24] H.A. van der Sloot, L. Heasman, P. Quevauviller, Harmonisation of leaching/extraction tests, Stud. Environ. Sci. 70, Elsevier Science, Amsterdam (1997).
- [25] H.A. van der Sloot, D.S. Kosson, O. Hjelm, Characteristics, treatment and utilization of residues from municipal waste incineration, Waste Manage. 21 (2001) 753–765.
- [26] H.A. van der Sloot, R.N.J. Comans, J.J. Dijkstra, Leaching methods for soil, sludge and treated biowaste, Horizontal Desk Study no. 23, August 2003.
- [27] H.A. van der Sloot, A. Van Zomeren, J.C.L. Meussen, P. Seignette, R. Bleijerveld, Test method selection, validation against field data and predictive modelling for impact evaluation of stabilised waste disposal, J. Hazard. Mater. 141 (2007) 354–369.
- [28] V. Cappuyns, R. Swennen, The application of pHstat leaching test to assess the pH-dependent release of trace metals from soils, sediments and waste materials, J. Hazard. Mater. 158 (2008) 185–195.
- [29] J.M. Chimenos, A.I. Fernández, R. Nadal, F. Espiell, Short term natural weathering of MSWI bottom ash, J. Hazard. Mater. B79 (2000) 287–299.
- [30] D.S. Kosson, H.A. van der Sloot, F. Sanchez, A.C. Garrabrants, An integrated framework for evaluating leaching in waste management and utilisation of secondary materials, Environ. Eng. Sci. 19 (2002) 159–203.
- [31] A.T. Ahmed, H.A. Khalid, A.A. Ahmed, D. Chen, A lysimeter experimental study and numerical characterisation of the leaching of incinerator bottom ash waste, Waste Manage. 30 (2010) 1536–1543.
- [32] J.M. Chimenos, A.I. Fernández, L. Miralles, M. Segarra, F. Espiell, Short term natural weathering of MSWI bottom ash as a function of particle size, Waste Manage. 23 (2003) 887–895.
- [33] T. Sabbas, A. Poletini, R. Pomi, T. Astrup, O. Hjelm, P. Mostbauer, G. Cappai, G. Magel, S. Salhofer, C. Speiser, S. Heuss-Assbichler, R. Klein, P. Lechner, Management of municipal solid waste incineration residues, Waste Manage. 23 (2003) 61–68.

- [34] T. Van Gerven, E. Van Keer, S. Arick, M. Jaspers, G. Wauters, C. Vandecasteele, Carbonation of MSWI-bottom ash to decrease heavy metal leaching, in view of recycling, *Waste Manage.* 25 (2005) 291–300.
- [35] S. Arickx, V. De Borger, T. Van Gerven, C. Vandecasteele, Effect of carbonation on the leaching of carbon and copper from MSWI bottom ash, *Waste Manage.* 30 (2010) 1296–1302.
- [36] G. Pfrang-Stotz, J. Reichelt, Technical properties and Environmental compatibility of MSWI bottom ash and other industrial by-products used as recycled materials in road construction, in: *Proceedings IT'02 Conference*, New Orleans, Louisiana, 2002.
- [37] J.A. Meima, R.N.J. Comans, Geochemical modeling of weathering reactions in municipal solid waste incinerator bottom ash, *Environ. Sci. Technol.* 31 (1997) 1269–1276.
- [38] A. Polettini, R. Pomi, The leaching behaviour of incinerator bottom ash as affected by accelerated ageing, *J. Hazard. Mater.* B113 (2004) 209–215.
- [39] J.A. Meima, R.D. van der Weijden, T.T. Eighmy, R.N.J. Comans, Carbonation processes in municipal solid waste incinerator bottom ash and their effect on the leaching of copper and molybdenum, *Appl. Geochem.* 17 (2002) 1503–1513.
- [40] P. Piantone, F. Bodènan, L. Chatelet-Snidaro, Mineralogical study of secondary mineral phases from weathered MSWI bottom ash: implications for the modelling and trapping of heavy metals, *Appl. Geochem.* 19 (2004) 1891–1904.
- [41] J.A. Meima, R.N.J. Comans, The leaching of trace elements from municipal solid waste incinerator bottom ash at different stages of weathering, *Appl. Geochem.* 14 (1999) 159–171.
- [42] P. Freyssinet, P. Piantone, M. Azaroual, Y. Itard, B. Clozel-Leloup, D. Guyonnet, J.C. Baubron, Chemical changes and leachate mass balance of municipal solid waste bottom ash submitted to weathering, *Waste Manage.* 22 (2002) 159–172.
- [43] J.A. Meima, R.N.J. Comans, Overview of geochemical processes controlling leaching characteristics of MSWI bottom ash, *Stud. Environ. Sci.* 71 (1997) 447–457.
- [44] C. Zevenbergen, R.N.J. Comans, Geochemical factors controlling the mobilization of major elements during weathering of MSWI bottom ash, *Stud. Environ. Sci.* 60 (1994) 179–194.
- [45] Council Directive 1999/31/EC of 26 April 1999 on the Landfill of Waste, *Official Journal of the European Communities*, 1999.
- [46] EA NEN 7371:2004. The Maximum Availability Leaching Test. Version 1.0, April 2005. <http://www.environment-agency.gov.uk/commondata/acrobat/ea_nen_7371_2004_1026067.pdf>.
- [47] B. Bergfeldt, V. Schmidt, A. Selinger, H. Seifert, J. Vehlou, Investigation of sintering processes in bottom ash to promote the reuse in civil construction (Part 2)—long term behaviour, in: *Proceedings of the WASCON'97, Waste Materials in Construction: Putting Theory into Practice*, Maastricht, NL, 1997, pp. 51–58.
- [48] APHA, *The Standard Methods for the Examination of Water and Wastewater*, 20th ed., American Public Health Association, Washington, DC, USA, 1998.
- [49] O. Hjelmar, Disposal strategies for municipal solid waste incineration residues, *J. Hazard. Mater.* 47 (1996) 345–368.
- [50] C.S. Kirby, J.D. Rimstidt, Mineralogy and surface properties of municipal solid waste ash, *Environ. Sci. Technol.* 27 (1993) 652–660.
- [51] C.A. Johnson, S. Brandenberger, P. Baccini, Acid neutralization capacity of municipal waste incineration bottom ash, *Environ. Sci. Technol.* 29 (1995) 142–147.
- [52] E. Rendek, G. Ducom, P. Germain, Influence of waste input and combustion technology on MSWI bottom ash quality, *Waste Manage.* 27 (2007) 1403–1407.
- [53] M. Kersten, H.C. Moor, C.A. Johnson, Speciation of trace metals in leachate from a MSWI bottom ash landfill, *Appl. Geochem.* 12 (1997) 675–683.
- [54] J.J. Dijkstra, H.A. van der Sloot, R.N.J. Comans, The leaching of major and trace elements from MSWI bottom ash as a function of pH and time, *Appl. Geochem.* 21 (2006) 335–351.
- [55] C.A. Johnson, M. Kersten, F. Ziegler, H.C. Moor, Leaching behaviour and solubility-controlling solid phases of heavy metals in municipal solid waste incinerator ash, *Waste Manage.* 16 (1996) 129–134.
- [56] J.A. Meima, R.N.J. Comans, Application of surface complexation/precipitation modeling to contaminant leaching from weathered municipal solid waste incineration bottom ash, *Environ. Sci. Technol.* 32 (1998) 688–693.
- [57] J.J. Dijkstra, J.C.L. Meeussen, H.A. van der Sloot, R.N.J. Comans, A consistent geochemical modelling approach for the leaching and reactive transport of major and trace elements in MSWI bottom ash, *Appl. Geochem.* 23 (2008) 1544–1562.
- [58] G. Cornelis, C.A. Johnson, T. Van Gerven, C. Vandecasteele, Leaching mechanisms of oxyanionic metalloid and metal species in alkaline solid wastes: a review, *Appl. Geochem.* 23 (2008) 955–976.
- [59] T. Astrup, C. Rosenblad, S. Trapp, T.H. Christensen, Chromium release from waste incineration Air-Pollution-Control residues, *Environ. Sci. Technol.* 39 (2005) 3321–3329.
- [60] Z. Abbas, B.-M. Steenari, O. Lindqvist, A study of Cr(VI) in ashes from fluidized bed combustion of municipal solid waste: leaching, secondary reactions and the applicability of some speciation methods, *Waste Manage.* 21 (2001) 725–739.
- [61] Z. Cai, D. Chen, K. Lundtorp, T.H. Christensen, Evidence of Al–Cr interactions affecting Cr-leaching from waste incineration ashes, *Waste Manage.* 23 (2003) 89–95.
- [62] W. Stumm, J. Morgan, *Aquatic Chemical Equilibria and Rates in Natural Waters*, 3rd ed., John Wiley, New York, 1996.
- [63] B.D. Bone, K. Knox, A. Picken, H.D. Robinson, The effect of carbonation on leachate quality from landfilled municipal solid waste (MSW) incinerator residues, in: T.H. Christensen, R. Cossu, R. Stegmann (Eds.), *Proceedings Sardinia 2003, 9th International Waste Management and Landfill Symposium*, 2003 (on CD ROM).
- [64] A. Polettini, R. Pomi, M. Ragaglia, Accelerated ageing as a tool for sustainable disposal of incinerator bottom ash, in: R. Cossu, R. Stegmann (Eds.), *Proceedings Sardinia 2005, 10th International Waste Management and Landfill Symposium*, 2005 (on CD ROM).
- [65] R. Baciocchi, G. Costa, E. Lategano, C. Marini, A. Polettini, R. Pomi, P. Postorino, S. Rocca, Accelerated carbonation of different size fractions of bottom ash from RDF incineration, *Waste Manage.* 30 (2010) 1310–1317.