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Heavy Metals Leaching of MSWI Bottom Ash: Effect of Short-term Natural Weathering

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Abstract. Municipal solid wastes incineration (MSWI) is an important part of the waste management systems in many European countries. Incineration process generates two main by-products: fly ash (FA) and bottom ash (BA). Bottom ash is composed of a variety of oxides, heavy metals and salts. Landfilling of untreated (fresh) BA can cause soil, surface and ground water contamination problems, because atmospheric precipitation in BA formed leachate, which contains of various materials. In this study investigates the influence of natural weathering to heavy metals leaching from BA. Leaching tests of bottom ash were carried out in 6 months, leachate samples were taken at 12 times (every 2 weeks). Heavy metals (Pb, Cu) concentrations were determined by atomic adsorption of spectral analysis method. The research shown, that the highest lead and copper concentration was determinated in 2 weeks weathered bottom ash leachate, respectively was 0.613 mg l^{-1} and 0.068 mg l^{-1}. In both cases the concentration of Pb (0.010–0.052 mg l^{-1}) and Cu (0.010-0.018 mg l^{-1}) became almost stable after 12 weeks. Can be concluded, that short-term (3 months) natural weathering is sufficient time for stabilise MSWI bottom ash and heavy metals (Cu and Pb) leaching. After 18–24 weeks heavy metals concentrations stabilized and remained almost constant.

Keywords: municipal solid waste incineration bottom ash, natural weathering, heavy metals, leaching.

Conference topic: Environmental protection.

Introduction

Incineration is one of the many technologies for managing municipal solid waste. Municipal solid waste incineration enables to reduce the mass of original waste by up to 70% (volume by up to 90%) and recovery energy (approximately 200 kWh ton-[1]) (Arickx *et al.* 2006; Su *et al.* 2013). MSWI produces two mains type of by-products: bottom (BA) and fly (FA) ashes (in amount of about 33% of the incinerated waste (Qiao *et al.* 2008; Ferraris *et al.* 2009; Gori *et al.* 2011). BA accounts for 85–95% of all the residues produced during MSWI (Rednek *et al.* 2006; Rosende *et al.* 2008). These ashes consist of inorganic matter (stone, ceramic, glass), ferrous and non-ferrous metals and unburned organic matter (plastic, fibre, wood etc.) (Allegrini *et al.* 2014; Del Valle-Zermeño *et al.* 2014a; Thomé-Kozmiensky, Thiel 2015). Unlike FA, BA is classified as non-hazardous waste by the European Waste Catalogue.

Ash composition can be expected to vary facility to facility (Siddique 2010; Zekkos *et al.* 2013). The main reason of that is constantly changing composition of waste, whitch are incinerated. Comparison of the chemical composition of BA in different countries incinerators are given in Table 1.

The research (Hyks *et al.* 2009; Bayuseno, Schmahl 2010; Rambaldi *et al.* 2010; Wei *et al.* 2011; Tang, Steenari 2016) in different countries showed that bottom ash contains of large quantities of heavy metals: zinc (903–7732 mg kg^{-1}), copper (1041–7743 mg kg^{-1}), lead (687–4552 mg kg^{-1}). Concentrations of cadmium and cobalt in the bottom ash were small (1–92 mg kg^{-1}).

MSWI bottom ash are unstable under atmospheric conditions, because municipal solid waste is incinerated in high temperatures and MSWI BA thereafter cooled rapidly. Environmental precipitation (snow and rain) and various chemical reactions influence bottom ash, therefore leachate is formed, which contains high amounts of various salts and heavy metals (Bouvet *et al.* 2007). Forteza *et al.* (2004), Valle-Zermeño *et al.* (2014) ir Rambaldi *et al.* (2010) analysis heavy metals concentrations in MSWI bottom ash eluate (Table 2).

The Table 2 shows that the highest concentration are lead (Pb) and copper (Cu), which respectively are 0.079–1.698 mg l^{-1} and 0.480–0.898 mg l^{-1}. High concentration of zinc (Zn) (0.818 mg l^{-1}) were found in one of tree samples. It was concluded that heavy metals concentrations in eluate are directly dependent on heavy metal amount in MSWI bottom ash.

Are several technologies (Fig. 1), such as vitrification (Toraldo *et al.* 2013; Ferraris *et al.* 2009), water washing (Kuo *et al.* 2013), full metal separation (Olsson *et al.* 2009) and weathering (natural and accelerated) (Arickx *et al.* 2006; Gori *et al.* 2011; Santos *et al.* 2013; Valle-Zermeño *et al.* 2013; Valle-Zermeño *et al.* 2015), for MSWI bottom ash stabilization.

Table 1. Amounts of major chemical elements in MSWI bottom ash in various countries

Country	Amount, wt%								Reference
	SiO_2	CaO	Fe_2O_3	Na_2O	Al_2O_3	MgO	K_2O	Other	
Spain	43.30	16.90	14.10	7.58	5.80	2.22	1.11	8.99	Valle-Zermeño *et al.* 2014b
Italy	33.70	35.00	5.37	2.27	13.31	4.62	1.66	4.07	Rambaldi *et al.* 2010
Germany	55.70	11.90	8.80	1.40	14.10	2.70	1.20	4.20	Müller and Rübner, 2006
France	47.82	15.99	6.23	6.34	8.63	2.38	n.d.	12.61	Rednek *et al.* 2007
Nether-lands	54.23	13.45	13.83	2.81	7.86	1.81	0.88	5.13	Tang *et al.* 2015
Slovenia	24.00	39.00	2.70	0.90	14.80	1.70	0.20	16.70	Jurič *et al.* 2006
Lithuania	54.94	16.12	7.82	5.12	4.88	3.28	1.21	4.64	Authors
Sweden	37.00	15.00	15.00	0.28	13.00	0.25	0.14	19.33	Lidelöw, Lager-kvist 2007
Sweden	37.00	15.00	15.00	0.28	13.00	0.25	0.14	19.33	Lidelöw, Lager-kvist 2007
Japan	31.93	33.40	5.97	2.53	16.67	3.33	2.22	3.97	Wei *et al.* 2011
China	59.59	7.58	5.50	1.32	18.61	1.32	2.29	3.79	Li *et al.* 2012
Taiwan	50.30	15.27	7.72	1.30	16.43	n.d.	2.14	6.84	Cheng 2012
USA	23.64	23.84	17.05	1.70	14.25	1.85	0.42	17.27	Saffarzadeh *et al.* 2011

Table 2. Amounts of major chemical elements in MSWI bottom ash in various countries

Country	Concentration in eluate, mg l^{-1}						Reference
	Cr	Zn	Cd	Ni	Pb	Cu	
Spain	<0.075	<0.200	<0.003	<0.040	1.698	0.898	Forteza *et al.* 2004
Spain	0.390	0.818	0.043	0.060	0.079	0.989	Valle-Zermeño *et al.* 2014
Italy	<0.001	0.073	0.008	<0.003	0.530	0.480	Rambaldi *et al.* 2010

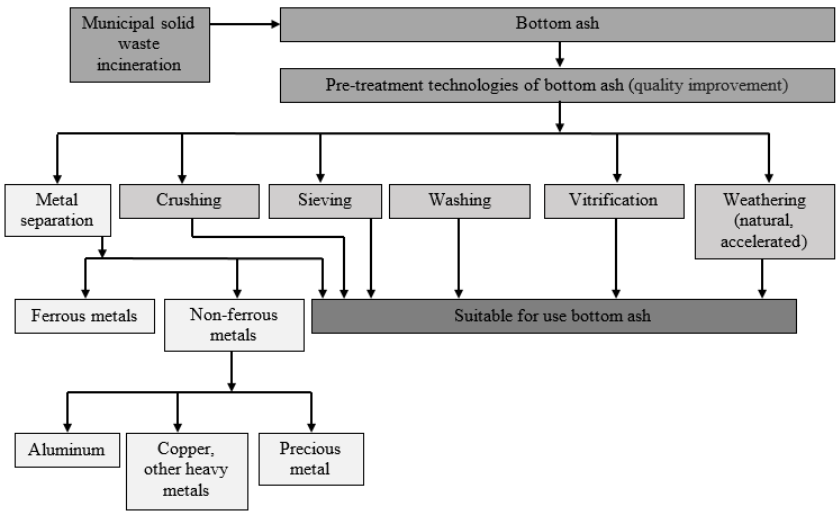

Fig. 1. Scheme of MSWI bottom ash pre-treatment technologies (Santos *et al.* 2013; Thomé-Kozmiensky, Thiel 2015)

Bottom ash pre-treatment technologies requires additional costs. Natural weathering is the most cost-effective method of MSWI bottom ash treatment. In 1–6 months period, some of the chemical and mineralogical characteristics of the BA changing. The main reaction are: oxidation of some metals (e.g. aluminium, copper), neutralisation of pH, dissolution and precipitation of the hydroxides and salts, carbonation (Meima, Comans 1999; Chimenos *et al.* 2003; Polettini, Pomi 2004). Carbonation reaction (Eq. 1) has been recognized to be an important step in weathering process

(Rednek *et al.* 2006). This process is described as an at leat two-stage mechanism. First CO_2 is absorbed in water, witch is in bottom ash porous. After that, it reacts with some components of MSWI bottom ash to produce carbonated species. The main reaction is the portlandite ($Ca(OH)_2$) carbonation (aqueous medium is mandatory), which can be represented by the following equation:

$$Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O. \quad (1)$$

The CO_2 dissolution in water causes pH to decrease and calcite to precipitate. Main positive effect of process is decreasing leaching of heavy metals (Cu, Pb, Cd, Zn) (Saffarzadeh *et al.* 2011; Yao *et al.* 2010; Yao *et al.* 2012). Metals are trapped in the new forms od minerals. Calcite is the predominant newly formed mineral during bottom ash maturation, combined with aluminium hydroxides and various sulfates (Piantone *et al.* 2004). On weathering process, CO_2 source is the atmospheric CO_2 (from air) and water – atmospheric precipitation.

This study aim is determine changes of concentrations of heavy metals (Pb, Cu) in short-term (1–6 months) weathering MSWI bottom ash filtrate, in unstable weather conditions.

Materials and Methods

Fresh bottom ash sample was taken from waste-to-energy plant storage tank in Klaipėda city, Lithuania. Plant operating a moving-grate furnace (850–1050 °C). Feed stream (treatment capacity of 816 t d^{-1}) mainly consists of household waste after sorting, with a small input of biofuel and non-hazardous industrial waste. The sampling procedure was carried out through quartering. The total amount of bottom ash collected was about 40 kg. Then, the bottom ash sample was mingled, homogenized.

For leaching of heavy metals (Cu, Pb) from MSWI bottom ash experiments, during weathering, was created a laboratory stand (Fig. 2), which is consisted of two main parts: bottom ash and leachate storage containers. Plastic containers with each other separated by a perforated plate and dense mesh. In order to protected leachate from bottom ash particles. Top part of laboratory stand (Fig. 2a) was filled with fresh bottom ash (~28 kg).

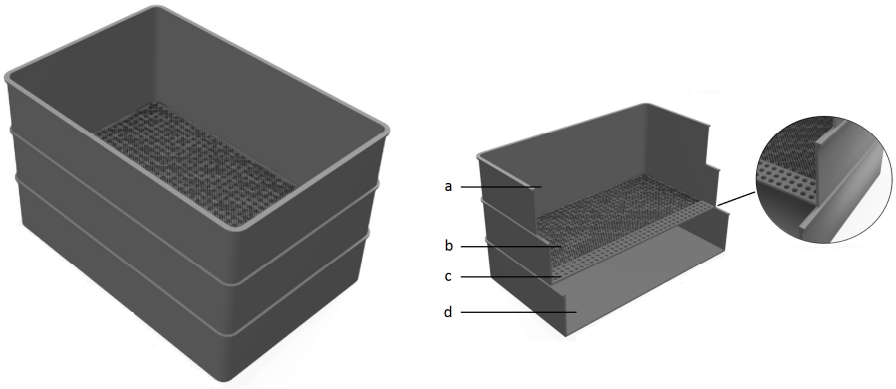

Fig. 2. Laboratory stand: a) MSWI bottom ash storage container; b) a perforated plastic plate; c) dense plastic mesh for fine particles detention; d) the filtrate collection container

6 months (December–May) bottom ash was exposed to atmospheric precipitation (rain, snow) and atmospheric carbon dioxide (CO_2). The formed and through bottom ash percolated leachate accumulated in the bottom container (Fig. 2d). Samples (leachate) for analysis takes 12 times (every 2 weeks). The samples are filtered using "Frisenette Qualitative Filter Paper Grade 201" filter paper. Heavy metals (Pb, Cu) concentrations were determined using atomic adsorption spectral analysis method.

The data of analysis to systematize and evaluate statistical methods. 15 values of copper (Cu) and lead (Pb) obtained from each sample (12 samples) of leachate. Estimated arithmetic mean of individual measurements, the following formula:

$$\overline{x} = \frac{\sum_{i=1}^{n} x_i}{n}, \quad (2)$$

where: $\overline{x}$ – the arithmetic mean of the individual measurements; x_i – individual measurements results; n – number of measurements.

After that, the dispersion is calculated (s^2):

$$s^2 = \frac{\sum_{i=1}^{n}(x_i - \overline{x})^2}{n-1}, \quad (3)$$

where: s^2 – dispersion; $\overline{x}$ – the arithmetic mean of the individual measurements; x_i – individual measurements results; n – number of measurements.

The average standard deviation is found by the formula:

$$s = \sqrt{\frac{\sum_{i=1}^{n}(x_i - \overline{x})^2}{n-1}}, \quad (4)$$

where: s – standard deviation; $\overline{x}$ – the arithmetic mean of the individual measurements; x_i – individual measurements results; n – number of measurements.

Random measurement errors, relative random and systematic errors was calculated. After that, relative error of measuring established:

$$\partial_t = \sqrt{\partial_r^2 + \partial^2}, \quad (5)$$

where ∂_t – relative error of measuring; ∂_r – relative random error; ∂ – systematic error.

If the relative error is less than 5%, it can be said that the results are credible.

Results and discussion

First established MSWI bottom ash leachate pH values (Fig. 3), which was 12.75–7.45. pH values in leachate formed after 2–10 weeks natural weathering process was almost stable 11.80–12.57. The highest pH value (12.57) established in sample taken after 6 weeks.

Striking pH decrease (from 12.15 to 7.75) is observed after 12 weeks weathering process. It can be assumed that after 3 months period begin to form ettringite (Bayuseno, Schmahl 2010). This chemical process can be described by the following formula:

$$6Ca^{2+} + 2Al^{3+} + 38H_2O \rightarrow 12H^+ + Ca_6Al_2(SO_4)4(OH)_{12} \times 26H_2O. \quad (6)$$

As the reaction (formula 6) product, formed a hydrogen ions (H^+), witch causing leachate acidification process (decrease of pH values). After 12–24 weeks MSWI bottom ash leachate pH values becoming stable (7.48–7.75).

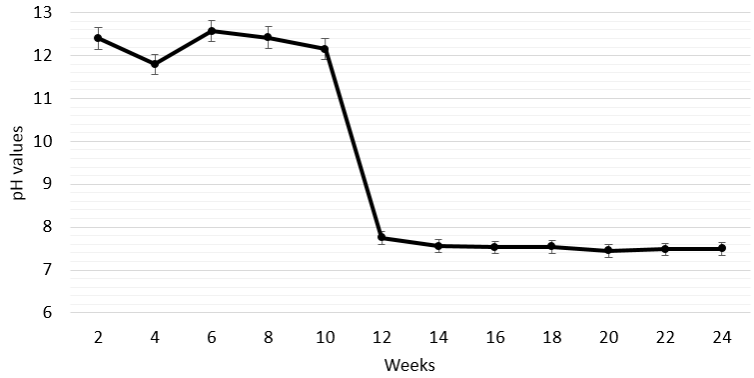

Fig. 3. pH values of MSWI bottom ash leachate after 2–24 weeks natural weathering process

In Fig. 4 is showed the lead (Pb) concentration in leachate. It was determinated that the concentration of lead in the samples are 0.010–0.613 mg l^{-1}. Highest concentrations was found in samples after 2 and 10 weeks weathering process, respectively 0.613 mg l^{-1} and 0.235 mg l^{-1}. After 12 weeks concentration of lead was quite small (0.010–0.052 mg l^{-1}) and almost stable. Comparing the concentrations of lead after 2 and 12 weeks, shows that the lead content in the eluate decreased 26.65 times.

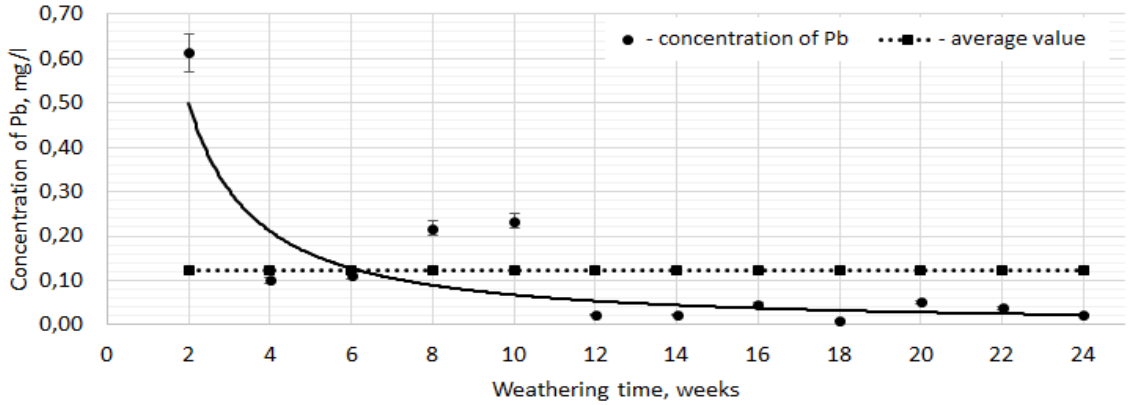

Fig. 4. Correlation between lead (Pb) concentration in MSWI bottom ash leachate and MSWI bottom ash weathering time

Fig. 5 demonstrates leachate values of copper after 2–24 weeks natural weathering process. Copper concentrations range in MSWI bottom ash leachate is 0.010–0.068 mg l^{-1}. The highest concentration (0.068 mg l^{-1}) of Cu was found in sample after 2 weeks of wathering. After 4 weeks Cu concentration decreased almost 2 times, and was 0.037 mg l^{-1}. Fig. 5 data shows that significant decreace of Cu concentration is in sample taken after 12 weeks (0.016 mg l^{-1}). After 12–24 weeks weathering process Cu concentratios have a low range of change (0.010–0.018 mg l^{-1}). It can be observed that the leached concentrations of Pb and deacreace dramatically after a very short period (2 weeks) of natural weathering. In contrast, the Cu concentrations decrace only slowly during the period studied (after 8 weeks).

According to the literature (Chimenos *et al.* 2000; Gori *et al.* 2011), Cu has a strong affinity with with dissolved humic and fulvic acids, to form organometallic complexes (Chimenos *et al.* 2003; Dijkstra *et al.* 2006). It can be assumed that dissolved organic carbon in MSWI bottom ash causes the Cu release. The low concentration of Cu decrease may be due to the partial oxidation of organic substance (by oxygen) and the furthermore formation of insoluble copper oxide at this pH. Pb concentration in MSWI bottom ash leachate decreaces much faster the pH values for the same period (Fig. 3). It can be assumed that the lower lead oxides solubility may be a result of sorption processes and formation of more stable mineral phases (Chimenos *et al.* 2000; Dijkstra *et al.* 2006; Gori *et al*. 2011).

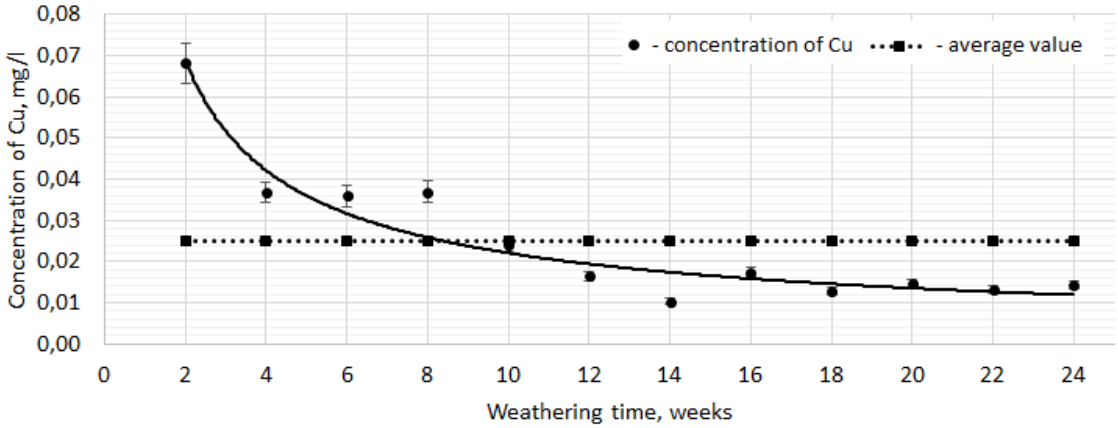

Fig. 5. Correlation between copper (Cu) concentration in MSWI bottom ash leachate and MSWI bottom ash weathering time

Can be concluded, that short-term (3 months) natural weathering is sufficient time for stabilise MSWI bottom ash and heavy metals (Cu and Pb) leaching.

Conclusions

1. Short-term (1–6 months) natural weathering of MSWI bottom ash experiments showed that leachate pH is almost stable (11.8–12.57) in 2–10 weeks period. Significant changes of pH (form 12.15 to 7.75) observed after 12 weeks weathering. Can be concluded that after 3 months period begin form sulphates. As the reaction product, formed a hydrogen ions (H^+), witch causing leachate acidification process.
2. Analysis of leachate samples showed that Pb concentrations in MSWI bottom ash are 0.010–0.613 mg l^{-1}. Highest concentrations ware found in samples after 2 (0.613 mg l^{-1}) and 10 (0.235 mg l^{-1}) weeks weathering process. After 12–24 weeks concentration of lead was small (0.010– 0.052 mg l^{-1}) and almost stable.

3. Copper concentrations range in MSWI bottom ash leachate is 0.010–0.068 mg l^{-1}. The highest concentration (0.068 mg l^{-1}) of Cu was found in sample after 2 weeks of wathering. Significant decreace of Cu concentration seen in sample taken after 12 weeks (0.016 mg l^{-1}).
4. The low decrease of concentration of Cu may be due to the partial oxidation of organic substance and the further formation of insoluble copper oxide. Lower lead oxides solubility may be a result of sorption processes and formation of more stable mineral phases.
5. Short-term natural weatering (till 6 months) is sufficient time for stabilise MSWI bottom ash and heavy metals (Cu and Pb) leaching.

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