

EFFECT OF DEDICATED ADDITIVES AND PRETREATMENT ON LEAD AND CHLORIDE LEACHING FROM WASTE-TO-ENERGY FLY ASH AND AIR POLLUTION CONTROL RESIDUES

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ABSTRACT: Most of the fly ash (FA) and air pollution control residue (APC residue) of modern waste-to-energy (WtE) plants do not meet the Waste Acceptance Criteria (WAC) for hazardous waste landfills set by the EU Council decision 2003/33/EC, primarily because the leaching of lead (Pb) and/or chlorides (Cl⁻) is too high. In this work, the effect of selected additives on Pb leaching from APC residue was investigated. Addition of 10% by mass of FeCl₃ had the highest effect on the Pb leaching from APC residue, which decreased immediately from 1694 mg.kg_{DM}⁻¹ to 9.7 mg.kg_{DM}⁻¹ and remained almost constant upon further curing. Addition of 10% by mass of Na₂CO₃ or Fe₂(SO₄)₃ immediately decreased the Pb leaching by about 50% but further curing of the S/S material was needed to reach the WAC. The effect of all these additives surpassed that of the effect of the pH change they caused. A second series of leaching experiments showed that addition of cement to mixed FA and APC residue reduced the Cl⁻ leaching by about 35%, which was not sufficient to reach the WAC. Additional experiments showed that washing of residues prior to landfilling can reduce the Cl⁻ leaching of the washed residues by about 90% to values below the WAC. Addition of Na₂CO₃, Fe₂(SO₄)₃, FeSO₄ or H₃PO₄ to APC residue prevented co-elution of Pb during the washing step and assured that the Pb leaching of the washed residues was well below the WAC. An economic evaluation taking into account additive and landfilling cost showed that addition of 0.2 Mg Na₂CO₃, 0.25 Mg FeSO₄ or 0.1 Mg H₃PO₄ per Mg of APC residue were cheaper options than adding 0.4 Mg cement.

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1. INTRODUCTION

Most of the fly ash (FA) and air pollution control residue (APC residue) generated in modern waste-to-energy (WtE) plants do not meet the Waste Acceptance Criteria for hazardous waste landfills (WAC) set by the EU Council decision 2003/33/EC, primarily because the leaching of lead (Pb) is too high, but also the leaching of chlorides (Cl⁻) can be problematic. This non-compliance necessitates pre-landfill treatment of these residues and solidification/stabilization (S/S) using cement as a binder is the most often applied method worldwide (Polletinti et al. 2001; Quinna et al., 2008). Adding cement and water to the residues converts them to easily transportable and manageable products, and also immobilises regulated elements by physical encapsulation, chemical incorporation and/or adsorption (Chen et al., 2009, Billen et al., 2015). The major disadvantage of using cement for S/S of municipal solid waste incineration (MSWI) residues is the increase in mass of the final product: around 300–400 kg of cement is used per Mg of residue treated (Van Gerven et al. 2005). Moreover, the production of cement is very energy intensive, has a high environmental impact and adds to the treatment cost.

Previous research by Billen et al. (2015) showed that, because of their cementitious properties, WtE residues can be solidified/stabilised (S/S) by adding only water. However, the WAC for hazardous waste landfills for Pb, whose leaching behaviour as function of pH is amphoteric, was only met after a curing time of 75 days, corresponding to the time needed to decrease the pH of the S/S residue to below 11.5 by natural carbonation. In practice, operators of WtE plants often don't have the space to store the S/S residues on-site for such a period of time. Therefore this work studies the effect of adding selected compounds to WtE residues on the curing time needed to reduce the Pb leaching below the regulatory limit value.

Billen et al. (2015) also showed that adding cement up to a "wt cement/ wt APC residue" ratio of 0.3 had no notable effect on the Cl⁻ leaching from the APC residues considered in their work, which remained above the WAC for hazardous waste landfills for granular waste materials. Therefore this work also studies the effect of adding higher amounts of cement on the Cl⁻ leaching from WtE residues. Furthermore, the possibility of washing the residues to remove the Cl⁻ prior to landfilling is investigated. However, during the washing process also the more soluble heavy metals, more specifically Pb, will leach from the residue into the washing water, impeding extensive treatment before the washing water can be discharged. Therefore, also the effect of selected additives on the co-elution of Pb was investigated.

The overall aim of this work is to establish a method for S/S of WtE residues without cement that is workable for plant operators and guarantees compliance with the WAC for hazardous waste landfills.

2. MATERIALS AND METHODS

2.1 Pb and Cl⁻ leaching experiments

2.1.1 FA and APC-residue

The FA and APC residue used in the Pb leaching experiments was obtained from WtE plant located in Belgium (plant A), incinerating about 400 000 Mg of MSW per year in three grate furnace lines. In each line, the flue gas is passed through a boiler for energy recovery. Flue gas treatment consists of a spray dryer followed by a fabric filter in a baghouse and a wet scrubbing installation. A more detailed description of the plant can be found in Vandecasteele et al. (2007). The FA and APC residue used in the experiments were sampled from the hoppers below the steam boiler and below the fabric filter, respectively. Preliminary leaching tests showed that the Pb leaching from FA and APC residue was 13.6 and 1 694 mg.kg_{DM}⁻¹, respectively, whereas the WAC for Pb leaching corresponds to 50 mg.kg_{DM}⁻¹. Because only the Pb leaching from the APC residue exceeded the WAC, this material was used in further experiments and is referred to as APC residue A.

For the chloride leaching experiments, a 1/3 mixture of FA and APC residue, further referred to as mixed ash (MA), was prepared. The MA was sampled from a 32 thermal MW WtE plant located in Poland (plant B), combusting 120 000 Mg MSW per year of about 7.5 MJ.kg⁻¹. Flue gas treatment consists of conditioned dry lime injection followed by a fabric filter in a baghouse. The FA was sampled from the hoppers below the convective passes of the boiler whereas the APC residue was sampled from the residue silo. The chloride leaching from untreated MA was 155 200 mg.kg_{DM}⁻¹.

2.1.2 Additives used in the Pb leaching experiments

Table 1 gives an overview of the different compounds that were added to the APC residue. Na₂CO₃ was selected because it releases carbonates upon dissolution, which can precipitate with different heavy metal ions, including Pb²⁺, as poorly soluble carbonates.

FeCl₃ and Fe₂(SO₄)₃ yield poorly soluble Fe(OH)₃ upon dissolution, incorporating heavy metals in the gelatinous precipitate, and therefore reducing heavy metal mobility (sweeping floc mechanism). Furthermore, these Fe(III) salts reduce the pH of APC residue of around 12.0 towards the point of lowest Pb leaching i.e. around a pH of 10.0 (Van Caneghem et al., 2016).

KH₂PO₄ was selected as an additive because the phosphates and hydrogen phosphates released upon its dissolution can form poorly soluble metal phosphates (Crannell et al., 2000; Geysen et al., 2006).

All additives used were analytical grade.

Table 1: Overview of the amount of additives added to APC residue A in the Pb leaching experiments.

Additive	Amount added to APC residue (wt%)
Na ₂ CO ₃	1%, 10%
FeCl ₃	10%
Fe ₂ (SO ₄) ₃	10%
KH ₂ PO ₄	10%

2.1.3 Sample preparation, leaching test and leachate analysis

For the Pb leaching experiments, blocks of solidified material were prepared: the compounds listed in Table 1 were each added in the indicated amount(s) to dried APC residue of plant A. About 22 ml of double deionized water (resistivity > 18.2 MΩ*cm) was then added per 100g of residue/additive mixture so that a paste-like structure was obtained. The paste was

subsequently poured in jars and after 8, 14, 19, 28, 49, 98 and 119 days, the hardened and cured material was submitted to a batch leaching tests according to EN 12457-4: 100ml of double deionized water was added to 10 g of crushed material with particle size < 4 mm and put in a shaking device (160 rpm) at room temperature for 24 hours.

Also for the chloride leaching tests, blocks of solidified material were prepared: Portland cement (BEAMIX cement 800) was added to MA of plant B in ratios (wt cement/ wt MA): 0.5, 0.75, 1.0, 1.5, 2.0, 3.0. After homogenisation of the material, about 25 ml to 40 ml of double deionized water was added per 100 g of cement/MA mixture to obtain a paste-like structure. The paste was then poured in moulds of $35 \cdot 10^{-3} \text{ m} \times 35 \cdot 10^{-3} \text{ m} \times 35 \cdot 10^{-3} \text{ m}$ (volume of about $43 \cdot 10^{-3} \text{ l}$). After 2 days, the samples were demoulded and left for further curing at room temperature. After a curing time of 14, 28, 42 and 84 days, the material was crushed and submitted to a batch leaching test as described for the Pb leaching experiments.

APC residue A was also submitted to a pH dependent leaching test in which increasing amounts of nitric acid (HNO_3) were added to the leaching water in different shaking jars in order to reduce the pH stepwise from 12.0, which was the intrinsic pH of the residue, to 7.1. After 24 hours of shaking, the leachate was filtered over a $0.45 \mu\text{m}$ glassfibre filter. The Pb concentration in the leachate was determined with a Thermo-Elemental X-series ICP-MS. The chloride concentration was determined by titration with AgNO_3 and K_2CrO_4 as an indicator (Mohr method).

2.2 Washing experiments

A second series of experiments aimed at removing Cl^- by washing prior to S/S and landfilling.

2.2.1 APC-residue

The APC residue used in the washing experiments was obtained from WtE plant C located in Belgium, incinerating about 100 000 Mg of MSW per year in two grate furnace lines. In each line, the flue gas is passed through a boiler for energy recovery. Flue gas treatment consists of a combination of an electrostatic precipitator (ESP), a spray dryer in which lime milk is injected and a baghouse filter. The APC residue used in the washing experiments consists of a mixture of ash collected in the spray dryer and the bag house filter and is further referred to as APC residue C. Preliminary leaching tests showed that the Pb and Cl^- leaching from untreated APC residue C equalled $1\,530 \text{ mg.kg}_{\text{DM}}^{-1}$ and $166\,000 \text{ mg.kg}_{\text{DM}}^{-1}$, respectively.

2.2.2 Additives used in the washing experiments

To limit the leaching of Pb from APC residue C during washing, Na_2CO_3 , $\text{Fe}_2(\text{SO}_4)_3$ and H_3PO_4 were added in the amounts indicated in Table 2. These values correspond to 0.28 g of CO_3^{2-} , 0.5 g of Fe^{3+} and 0.4 g of PO_4^{3-} , which are the active, relevant ions for stabilization, added to samples of 5 g of APC residue each. The amounts were selected by preliminary optimization of the washing procedure for APC residue C (De Smet, 2014), using the amounts yielding the lowest Pb concentration in the washing water. FeSO_4 was also used as a source of Fe^{2+} , as suggested in the Ferrox process (Sorensen and Koch, 2001; Lundtorp et al., 2002), but no improved effect was observed in comparison to $\text{Fe}_2(\text{SO}_4)_3$ (Fe^{3+}) and therefore these results are not shown in this paper.

Table 2: Overview of the amount of additives added to APC residue C in the washing experiments.

Additive	Amount added to APC residue (wt%)
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Na ₂ CO ₃	17
Fe ₂ (SO ₄) ₃	26
Na ₂ CO ₃ + Fe ₂ (SO ₄) ₃	13 + 23
H ₃ PO ₄	8

2.2.3 Washing experiments and water analysis

APC residue C was mixed with the amount of additives indicated in Table 2. The mixed material was then put in a jar and double deionized water was added in an L/S ratio of 10. Subsequently the jars were shaken at 160 rpm during 10 minutes since preliminary tests (De Smet, 2014) showed that equilibrium concentrations of Cl⁻ in the leachate were obtained after this amount of time.

The content of the jars was then filtered over a 8-12 µm filter (Fisherbrand qualitative filter paper) under light vacuum to obtain a liquid fraction i.e. the washing water, and a dried solid fraction. In the washing water, the Pb concentration was determined with ICP-MS as described in Section 2.2.3 and the Cl⁻ concentration was determined with a Dionex IC 2000 ion chromatograph. The solid fraction was submitted to a leaching test as described in Section 2.2.3 and the Pb and Cl⁻ concentration in the leachate were determined with ICP-MS and IC, respectively.

3. RESULTS AND DISCUSSION

3.1 Pb leaching experiments

3.1.1 Effect of pH and additives on Pb leaching

The open circles in Figure 1 represent the leaching of Pb from APC residue A without additives as a function of the pH of the leachate obtained after 24 hours of shaking. The results for the additive/residue mixtures in Figure 1 were obtained after immediate submission of the material to a batch leaching test, so without prior curing of the mixture. Figure 1 shows clearly that of all the additives, FeCl₃ had the highest effect on the Pb leaching that decreased from 1694 mg.kg_{DM}⁻¹ (APC residue A without additives) to 9.7 mg.kg_{DM}⁻¹, which is well below the WAC of 50 mg.kg_{DM}⁻¹. The addition of 10% FeCl₃ caused the pH of APC residue A to decrease from 12.0 to 11.1, decreasing the leaching of Pb as shown by the results of the pH dependent leaching test in Figure 1. Figure 1 however also clearly shows that the effect of 10% FeCl₃ addition surpassed this "pH effect", because the Pb leaching, represented by a black rhomb in Figure 1, is well below the Pb leaching from untreated APC residue at the same pH of 11.1, represented by the open circles. Probably the Pb leaching was further reduced by sweep flocculation with the gelatinous Fe(OH)₃ formed upon dissolution of FeCl₃ and by adsorption of Pb-ions to the formed Fe₂O₃. The addition of 10% KH₂PO₄ also immediately decreased the Pb leaching, which can be explained by the precipitation of poorly soluble Pb₃(PO₄)₂ but the WAC was however not met. The effect of Na₂CO₃ was less pronounced; 10% addition resulted in an immediate Pb leaching reduction of about 50%, but the leaching concentration remained above WAC. Na₂CO₃ can lead to formation of poorly soluble PbCO₃, but simultaneously increases the pH, increasing the solubility of both Pb(OH)₃ and the formed PbCO₃. Although the reaction mechanisms for Fe₂(SO₄)₃ are the same as for FeCl₃, the effect on the Pb leaching of adding 10% Fe₂(SO₄)₃ was less pronounced than of

adding 10% FeCl_3 . This can partly be explained by the less pronounced pH reduction when $\text{Fe}_2(\text{SO}_4)_3$ was added and partly by the fact that the same amounts of both Fe-compounds were added and FeCl_3 is relatively richer in Fe^{3+} than $\text{Fe}_2(\text{SO}_4)_3$ (1 g of FeCl_3 contains about 0.34 g of Fe^{3+} whereas 1 g of $\text{Fe}_2(\text{SO}_4)_3$ contains about 0.28 g of Fe^{3+}). The addition of Fe-sulphates during washing of MSWI residues is also known as the Ferrox process and was, amongst others, described by Lundtorp et al. (2001). Comparison of the obtained Pb leaching concentrations with the results of the pH dependent leaching test in Figure 1 shows that the effect of all additives surpassed that of the effect of the pH change they caused.

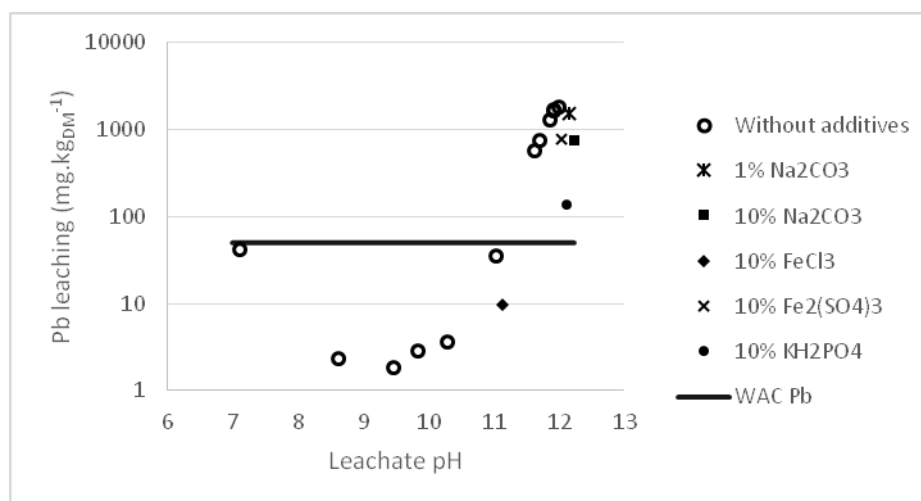


Figure 1. Pb leaching ($\text{mg.kg}_{\text{DM}}^{-1}$) from APC residue A with and without additives as a function of the pH of the leachate after 24h.

Figure 2 shows the leaching of Pb from S/S APC residue A with and without additives as a function of the curing time. If no additives were added to the APC residue, the Pb leaching decreased, but remained above the WAC, even after 119 days of curing. This decrease can be explained by natural carbonation: $\text{Ca}(\text{OH})_2$ present in the residue reacts with CO_2 from the air, decreasing the pH and the solubility of Pb species as shown by the pH dependant leaching curve in Figure 1. Furthermore, the formed carbonates (CO_3^{2-}) can react with Pb ions to form poorly soluble PbCO_3 . Also for the APC residue to which Na_2CO_3 or $\text{Fe}_2(\text{SO}_4)_3$ was added, the Pb leaching decreased over time. The decrease was however more pronounced than for the APC residue without additives, indicating that it was not only the result of carbonation reactions, but probably also of other physico-chemical phenomena initiated by the additives. In the APC residue with 1% Na_2CO_3 and 10% $\text{Fe}_2(\text{SO}_4)_3$, the Pb leaching did not reach the WAC, even after 119 days of curing time and for the APC residue with 10% Na_2CO_3 , the WAC was only met after this period of time. Although addition of 10% KH_2PO_4 had a strong initial effect, the Pb leaching decreased little further over time and it took 100 days to reach the WAC. The Pb leaching from the APC residue with 10% FeCl_3 was below the limit immediately and remained below the WAC. The apparent slight increase in the Pb leaching measured after 8 and 14 days are probably due to inhomogeneities in the samples rather than to chemical and/or physical interactions. The WAC also includes leaching limit values for the anions Cl^- and SO_4^{2-} and, as already stated in Section 1, in APC residue particularly the leaching of Cl^- is often close to or above the limit. In this view, especially the addition of FeCl_3 further increasing the Cl^- leaching can result in non-compliance of the residue.

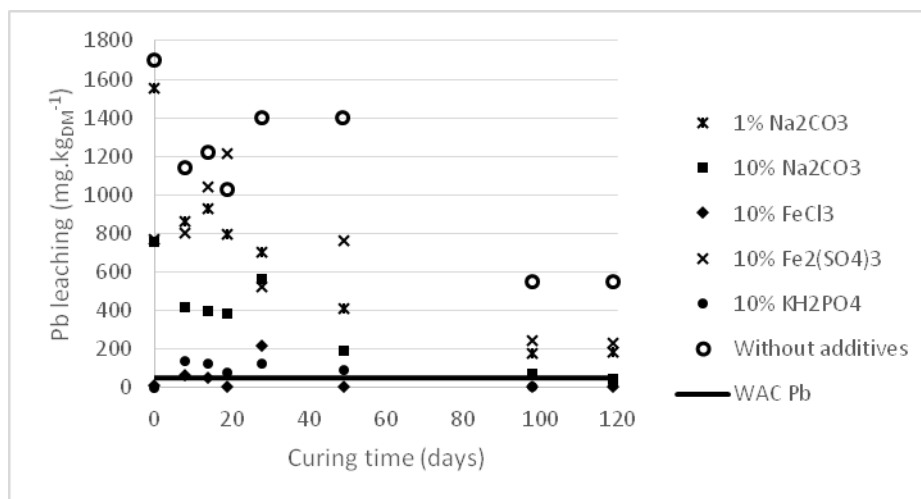


Figure 2. Pb leaching ($\text{mg.kg}_{\text{DM}}^{-1}$) from S/S APC residue A with and without additives as a function of curing time.

3.2 Cl⁻ leaching experiments

Figure 3 (a) gives the leaching from S/S MA and cement/MA mixtures as a function of curing time. The chloride leaching of S/S MA was about $155\,000\text{ mg.kg}_{\text{DM}}^{-1}$ and could only be reduced below the WAC for hazardous waste landfills of $25\,000\text{ mg.kg}_{\text{DM}}^{-1}$ by adding 3 times the MA weight of cement. Since the chloride concentration in cement is negligible compared to the chloride concentration in MA, cement “dilutes” the chlorides in the MA. Taking into account this “dilution effect” (Figure 3 (b)), cement only reduced the chloride leaching by about 35%, which can, given the high solubility of the present chloride species, most likely be attributed to physical encapsulation of the MA particles. Curing did not further decrease the chloride leaching, but slightly increased chloride mobility instead (Figure 3 (a) and (b)).

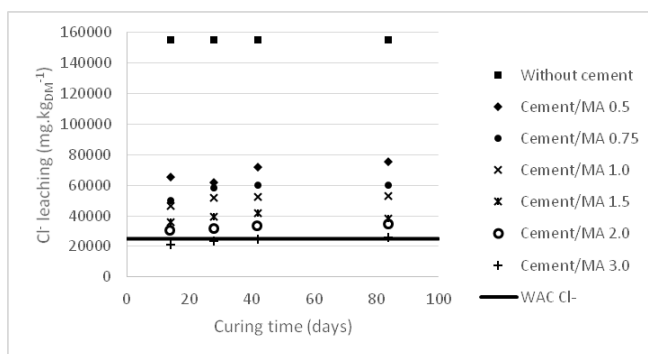


Figure 3 (a). Cl⁻ leaching ($\text{mg.kg}_{\text{DM}}^{-1}$) from S/S MA with and without cement addition as a function of curing time.

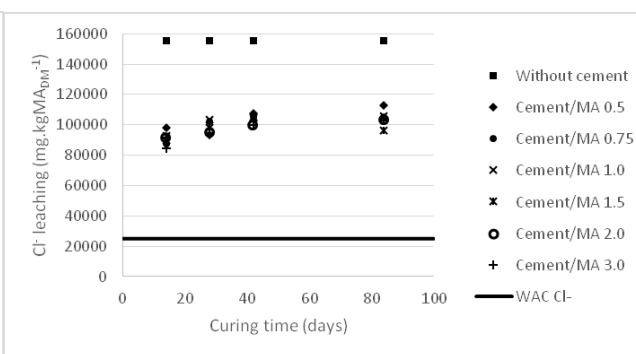


Figure 3 (b). Cl⁻ leaching ($\text{mg.kgMA}_{\text{DM}}^{-1}$) from S/S MA with and without cement addition as a function of curing time. The Cl⁻ leaching is expressed per kg of MA in the MA/cement mixture, this way correcting for the dilution effect of the added cement.

It should be noted that the WAC used as a standard for acceptance on hazardous waste landfills in this work apply for granular, non-monolithic waste. In practice however, FA and APC residue are mixed with water and or cement and poured in e.g. big bags to form big monolithic blocks that are then landfilled. EU Council decision 2003/33/EC states that each

member states shall set own criteria for monolithic waste to provide the same level of environmental protection given by the WAC for granular waste. In Flanders as an example, WAC for monolithic waste are stated in the environmental permit of the landfill operator and leaching of regulated components is to be determined by means of a diffusion test instead of a batch leaching test. Furthermore, EU Council decision 2003/33/EC provides that the WAC for TDS can be used alternatively to the WAC for SO_4^{2-} and Cl^- so that flue gas cleaning residues with Cl^- leaching above 25 000 can still comply. All these aspects have to be considered for each specific residue when establishing an optimal treatment.

3.3 Washing experiments

When APC residue C without additives was washed as described in Section 2.2.3, determination of the Cl^- concentration in the washing water confirmed that all leachable chlorides were dissolved in the washing water. The washing water however contained about 190 mg.l^{-1} of Pb, indicating that during the washing also Pb was leached from the APC residue. The concentration of Cr, Cd and Sb in the washing water was below 1 mg.l^{-1} , whereas the concentration of Cu and Zn was about 6 mg.l^{-1} , indicating that Pb co-elution was indeed most problematic. To reduce the co-elution of primarily Pb, the compounds listed in Table 2 were chosen based on the results from section 3.1 and were added to APC residue C after which the mixtures were washed as described in Section 2.2.3.

Table 3 gives the Cl^- and Pb leaching upon washing and from the obtained washed residues for the different APC residue/additive combinations.

Table 3: Pb and Cl^- ($\text{mg.kg}_{\text{DM}}^{-1}$) leaching upon washing and from the obtained washed residues for the different APC residue/additive combinations. N.d. means not detectable, i.e. below the detection limit of $0.1 \text{ mg.kg}_{\text{DM}}^{-1}$.

Leaching upon washing ($\text{mg.kg}_{\text{DM}}^{-1}$)	Pb	Cl^-
Without additives	785	129 000
17% Na_2CO_3	49	141 000
26% $\text{Fe}_2(\text{SO}_4)_3$	0.2	155 000
13% Na_2CO_3 + 23% $\text{Fe}_2(\text{SO}_4)_3$	0.3	133 000
8% H_3PO_4	4.1	134 000
Leaching from washed residue ($\text{mg.kg}_{\text{DM}}^{-1}$)		
Without additives	16.4	12 300
17% Na_2CO_3	1.3	15 900
26% $\text{Fe}_2(\text{SO}_4)_3$	n.d.	16 600
13% Na_2CO_3 + 23% $\text{Fe}_2(\text{SO}_4)_3$	n.d.	14 700
8% H_3PO_4	n.d.	15 100
<i>WAC hazardous waste landfill</i>	<i>50</i>	<i>25 000</i>
<i>WAC non-hazardous waste landfill</i>	<i>10</i>	<i>15 000</i>

The Cl^- leaching from the washed residue equalled around $15 000 \text{ mg.kg}_{\text{DM}}^{-1}$, which is well below the WAC for hazardous waste landfills. This Cl^- leaching is due to the Cl^- containing washing water that is not removed during the residue filtration and drying step. Indeed, upon solid/liquid separation as described in Section 2.2.3, a solid residue of washed ash with a dry matter content of around 62.5% by mass was obtained.

If no additives were added to the APC residue, the Pb leaching upon washing is high, as

expected. This is unwanted, as it transmits the Pb problem from a solid to a liquid waste, while the latter preferably is only contaminated with Cl⁻. The results in Table 3 show that all additives strongly reduce the co-elution of Pb, which can be explained by the stabilisation mechanisms described in Section 3.1. The best results were obtained with Fe₂(SO₄)₃; the effect of FeCl₃ was not tested because of the unwanted increase of Cl⁻ the addition causes. The obtained washed residue complied with the WAC for hazardous waste landfills and the Pb leaching from APC residue with additives was even below the limit value for non-hazardous waste landfills. As explained, the Cl⁻ leaching is caused by washing water that is still present in the washed residues and can be reduced by a better dewatering of these residues, or by performing a continuous instead of a batch extraction.

For each of the considered additives, an economical evaluation was done and compared to the treatment cost of Portland cement, which is currently the best available technology (BAT). Two cost factors were considered: the additive cost and the landfilling cost. The additive cost was calculated by multiplying the average bulk price of an additive with the minimum amount of the additive needed to sufficiently influence Pb leaching behaviour (Alibaba, 2014). These amounts were considered to be per Mg of APC residue: 0.2 Mg Na₂CO₃, 0.25 Mg FeSO₄, 0.35 Mg Fe₂(SO₄)₃, 0.1 Mg H₃PO₄ and 0.4 Mg cement. FeSO₄ was considered as an alternative to Fe₂(SO₄)₃, because it is much cheaper and it was proven that it can have the same effect as Fe₂(SO₄)₃ provided the washing water is sufficiently aerated to oxidize Fe²⁺ to Fe³⁺ (De Smet, 2014). To calculate the landfilling cost of the mixture, the average landfilling price in Flanders for category 2 landfills, which is € 120 per Mg, was used (OVAM, 2012).

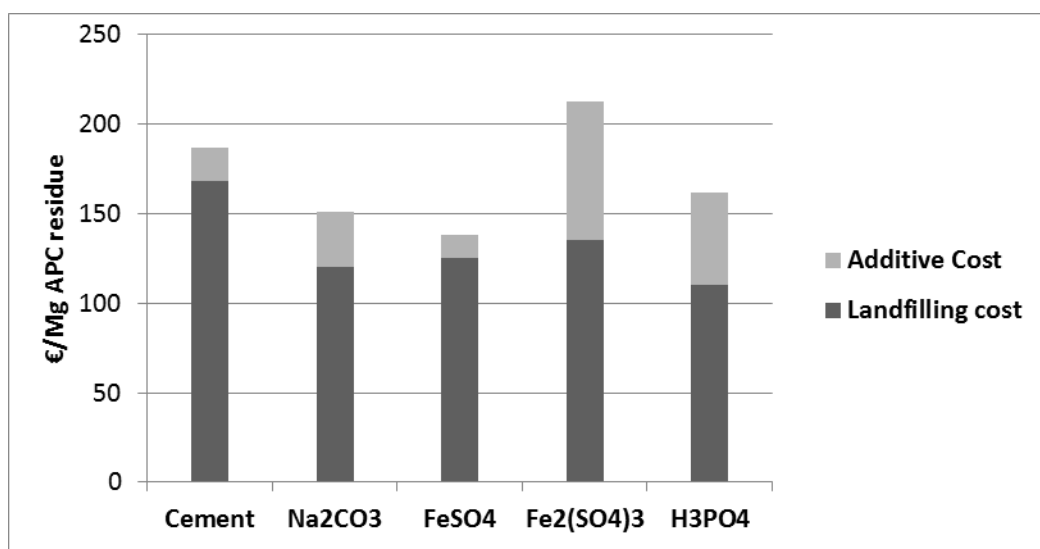


Figure 4. Additive and landfill cost (€) to treat 1 Mg of APC residue. (The added amounts correspond to 0.2 Mg Na₂CO₃, 0.25 Mg FeSO₄, 0.35 Mg Fe₂(SO₄)₃, 0.1 Mg H₃PO₄ and 0.4 Mg cement per Mg of APC residue).

Figure 4 shows that the landfilling cost of the additive alternatives is lower than for the BAT treatment with cement. For FeSO₄, also the additive cost is lower than for cement, so overall addition of 0.2 Mg of FeSO₄ per Mg of APC residue is the cheapest option. Although the cost of adding 0.2 Mg Na₂CO₃ or 0.1 Mg H₃PO₄ is higher than the cost of 0.4 Mg cement, also for these additives the overall treatment cost remains lower. Treatment with 0.35 Mg Fe₂(SO₄)₃ is the only treatment option that is more expensive than 0.4 Mg cement addition.

5. CONCLUSIONS

The results of the experiments discussed in this work, provide the basis for tailored S/S of WtE residues.

If only the Pb leaching from the WtE residue exceeds the WAC, addition of FeCl_3 is sufficient to immediately decrease the Pb leaching and assure compliance with the WAC for hazardous waste landfills. The exact amount of FeCl_3 that has to be added depends on the initial Pb leaching value and pH of the residue. For the APC residue considered in this work, 10 % of FeCl_3 was needed to reduce the Pb leaching from $1694 \text{ mg.kg}_{\text{DM}}^{-1}$ to $9.7 \text{ mg.kg}_{\text{DM}}^{-1}$.

If only the leaching of Cl^- exceeds the WAC, washing of the residues with subsequent mechanical drying before landfilling is the best treatment option since the effect of cement addition on the Cl^- leaching is very limited. To illustrate this, in the case of the representative APC residue considered in this work, a cement/APC residue ratio as high as 3 was needed to comply with the WAC, which is not workable.

If both the Pb and Cl^- leaching exceed the WAC, addition of Na_2CO_3 , FeSO_4 , $\text{Fe}_2(\text{SO}_4)_3$ or H_3PO_4 will ensure that during the washing step Pb is not co-eluted and that the washed residue complies with the WAC. Again, the exact amount of additives needed to reach the WAC depends on the initial Pb and Cl^- leaching of the WtE residue. An economic evaluation based on additive and landfilling cost showed that addition of 0.2 Mg Na_2CO_3 , 0.25 Mg FeSO_4 or 0.1 Mg H_3PO_4 per Mg of APC residue were cheaper options than adding 0.4 Mg cement.

REFERENCES

Alibaba. Retrieved 20/05/2014. <http://www.alibaba.com/>

Billen P., Verbinnen B., De Smet M., Dockx G., Ronsse S., Villani K., De Greef J., Van Caneghem J., Vandecasteele C. (2015). Comparison of solidification/stabilization of fly ash and air pollution control residues from municipal solid waste incinerators with and without cement addition. *J. Mater. Cycles Waste Manag.*, vol. 17, 229-236

Crannell B.S., Eighmy T.T, Krzanowski J.E., Dykstra Eusden J., Shaw E. L., Francis C.A. (2000). Heavy metal stabilization in municipal solid waste combustion bottom ash using soluble phosphate. *Waste Manage.*, vol. 20, 135-148

De Smet M. (2014). Advanced treatment of air pollution control residues from Waste-to-Energy: metal immobilization and chloride removal. Master thesis KU Leuven.

Geysen D., Vandecasteele C., Jaspers M., Brouwers E., Wauters G. (2006). Effect of improving flue gas cleaning on characteristics and immobilisation of APC residues from MSW incineration. *J. Haz. Mat.*, vol. B128, 27-38.

Lundtorp K., Jensen D., Sorensen M., Christensen E. (2002). Treatment of waste incinerator air-pollution-control residues with FeSO_4 : Concept and product characterization. *Waste Manag.* Vol. 20, 69-79.

OVAM. (2012). Tarieven en capaciteiten voor storten en verbranden actualisatie tot 2012.

Quina M.J., Bordado J.C., Quinta-Ferreira R.M. (2008). Treatment and use of air pollution control residues from MSW incineration: an overview. *Waste Manag.*, vol. 28, n.11, 2097–2121.

Polettini A., Pomi R., Sirini P., Testa F. (2001). Properties of Portland cement—stabilised MSWI fly ashes. *J. Hazard. Mater.*, vol. B88, 123–138.

Sorensen M.A., Koch C.B. (2001). Thermally induced transformations of Fe oxide-stabilized residues from waste incineration. *Mineralogical Mag.*, vol. 65, n. 5, 635-643.

Van Caneghem J., Verbinnen B., Cornelis C., de Wijs J., Mulder R., Billen P., Vandecasteele C. (2016). Immobilization of antimony in waste-to-energy bottom ash by addition of calcium and iron containing additives. *Waste Manag.*, in press.
<http://dx.doi.org/10.1016/j.wasman.2016.05.007>

Vandecasteele C., Wauters G., Arickx S., Jaspers M., Van Gerven T. (2007). Integrated municipal solid waste treatment using a grate furnace incinerator: The Indaver case. *Waste Manag.* Vol. 27, 1366-1375.

Van Gerven T., Geysen D., Stoffels L., Jaspers M., Wauters G., Vandecasteele C. (2005). Management of incinerator residues in Flanders (Belgium) and in neighbouring countries. A comparison. *Waste Manag.*, vol. 25, n.1, 75–87.