COMPARISON OF SOLIDIFICATION/STABILIZATION OF FLY ASH AND AIR POLLUTION CONTROL RESIDUES FROM MUNICIPAL SOLID WASTE INCINERATORS WITH AND WITHOUT CEMENT ADDITION

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Abstract

Solidification/stabilization (S/S) of fly ash and air pollution control residues from MSWI was studied, with and without the use of cement. It appeared that metal and oxyanion leaching from these stabilised residues was comparable after 75 days of curing. Leaching of chloride and sulfate did not decrease by the addition of cement during S/S treatment. In an LCA perspective the environmental impact of the S/S treated mixtures with cement in the toxicity related impact categories was higher than the impact of the S/S treated mixtures without cement. Moreover, the production of cement for application in S/S also causes an impact in the impact categories climate change and terrestrial acidification. For environmental considerations in an LCA perspective S/S without cement should be preferred.

Keywords

Solidification/Stabilization – Fly ash – Air Pollution Control Residues – Landfill – Municipal Solid Waste Incineration

1. Introduction

Air pollution control residues from municipal solid waste incineration (MSWI) are hazardous waste materials for which landfilling is the most appropriate treatment. Solidification and stabilization (S/S) using cement as a binder is worldwide the most often applied pre-landfill treatment method for air pollution control residues from MSWI [1-3]. This method not only involves a conversion of the residues into a monolithic or granular material, ensuring easy handling and transportation to landfill sites [2], but also immobilises toxic pollutants by physical encapsulation, chemical incorporation and/or adsorption [2,4,5].

Although common practice, the use of cement for S/S of MSWI residues has some major disadvantages. Typically, around 300-400 kg of cement is used per tonne of residue treated [6,7]. Along with the added water, this causes a significant increase of the volume and mass to be landfilled. Moreover, the production of cement has a large environmental impact. For instance, when solidified/stabilised fly ash is landfilled, almost the entire impact on global warming is related to the production of the cement used, which corresponds to 820 kg CO₂-eq. per tonne of cement [8].

Similarly to cement clinkers, MSWI fly ash contains a high concentration of Ca, Si and Al. Furthermore, when exposed to temperatures higher than 850 °C in a post-combustion chamber, the reactions occurring in ash particles are similar to those in a cement kiln, resulting in boiler ash and fly ash with cementitious properties. A large fraction of most air pollution control residues is unreacted hydrated lime, which is also known as a binding agent. As these MSWI residues as such have cementitious properties [9,10], it is likely that S/S of these materials can also be achieved by only adding water, thereby significantly reducing the environmental impact of the treatment. This paper compares the performance and environmental impact of S/S

treatment of fly ash and air pollution control residue from a MSWI in Flanders, with and without addition of cement. The leaching behavior is assessed and compared to the waste acceptance criteria (WAC) for hazardous waste landfills (EU Council decision 2003/33/EC [11]). Although in reality the leachate from a landfill is collected and treated, in this paper we use the data from the leaching tests as input for the environmental impact assessment. The mechanical strength of the residues S/S treated without cement may be lower than that of residues S/S treated with cement. However, since the present paper examines S/S as a treatment to comply with leaching criteria for landfilling, for which only a limited mechanical strength is required, this aspect was not assessed.

2. Materials and Methods

2.1 Installation and samples

Fly ash (FA) and air pollution control (APC) residues were sampled from a MSWI installation in Eeklo, Belgium, during steady-state operation. The installation comprises two grate furnace incinerators, with a combined capacity of approximately 100,000 tonne per year (each line accounting for 7 tonne h⁻¹). The flue gas cleaning section consists of an electrostatic precipitator, a semi-dry scrubber with lime injection (70 kg h⁻¹ for each line), activated carbon (7 kg h⁻¹ for each line) and sodium bicarbonate injection (dosing by continuous measurements of acid gas emissions), a baghouse filter and a catalytic deNO_x system. The APC residue collected in the baghouse filter is approximately 175 kg h⁻¹ for each line. The ash from the boiler and from the electrostatic precipitator is collected together as FA. [12]

2.2 S/S treatment

200 g of FA was mixed with 60 ml of double deionised (Millipore Milli-Q) water and stirred until a paste was obtained. At the same time, samples composed of 200 g of FA or APC residue, 60 g of cement (type CEM II) and 78 ml of water were prepared.

In a modern MSWI installation, the ratio of generated FA to APC residue is approximately 30/70 on a wt/wt basis [7]. Therefore, 200 g of a 30/70 (wt/wt) mixture of FA and APC residue was mixed with 60 ml of water (w), and treated similarly as the previous samples. For comparison, a sample with cement was prepared: 60 g of cement (CEM) mixed with 200 g of the 30/70 mixture of FA and APC residue, to which 78 ml of water was added.

Approximately 60 g of each prepared sample was poured into separate cups. Reference samples of untreated FA and APC residue were kept in a closed recipient throughout the test.

The resulting 6 types of S/S treated mixtures (FA+w, APC+w, FA+CEM+w, APC+CEM+w, FA+APC+w, FA+APC+CEM+w) were left to cure in ambient air at 25 °C for 35 days. After this period, each mixture was ground and homogenised (with respect to the degree of carbonation, as this may have a consequence on the pH and hence the leaching of heavy metals). The ground mixtures were subsequently dried at 40 °C for 5 days, before performing the leaching tests.

2.3 Leaching tests

All leaching tests were performed in duplicate or triplicate. After 40 days of curing-drying (35 days at 25 °C and 5 days at 40 °C), 20 g samples from each of the 6 S/S treated mixtures and from the reference FA and APC residues were leached at a liquid/solid ratio (L/S) of 2, and 5 g samples from the 6 S/S treated mixtures were leached at an L/S of 10, in 24 h batch leaching tests with double deionised water at 25 °C using a shaking device (Gerhardt Laboshake, 160 rpm). The remaining part of the ground mixtures was left to cure in ambient air at 25 °C for another 35 days, bringing their total curing-drying time to 75 days. After this period, 25 g samples of each mixture were leached at an L/S of 2 and 5 g samples of each mixture were leached at an L/S of 10.

The leachates were filtered over a 0.45 µm membrane filter (Chromafil), whereafter the concentrations of regulated metals were measured by inductively coupled plasma mass spectrometry (ICP-MS, Thermo Xi series), and the chloride and sulfate concentrations by ion chromatography (IC, Dionex 2000). Se concentrations are not reported, because they were below the detection limit in most samples. For the cement containing samples, the leaching values (in mg/kg DM) are converted to the original mass of the MSWI residues, to avoid the dilution effect from cement.

A pH dependent leaching test was performed on the non-treated FA and APC residue, and on the S/S treated FA and APC residue with and without cement addition after 40 days of curing. The pH was adjusted with various amounts of HNO₃ or NaOH in a batch leaching test with an L/S of 10 as described above.

2.4 Environmental impact assessment

The environmental impact of landfilling the MSWI residues is assessed, using the midpoint impact factors, hierarchic perspective, of the ReCiPe environmental impact assessment method version 1.08 [13]. The functional unit is landfilling 1 tonne of a mixture of S/S treated FA and APC residue (30/70 wt/wt ratio), for a period corresponding to an L/S of 2 for short term impacts (roughly 100 years) and an L/S of 10 for long term impacts. The mass of cement itself is excluded from the functional unit, because only the landfilling of the MSWI residues with different technologies is compared. Landfill construction and operation are excluded from the assessment. The heavy metal emissions from the landfill are estimated based on the batch leaching results with an L/S of 2 for short term leaching and with an L/S of 10 for long term leaching. The leached metal amounts, estimated by the batch leaching tests, are multiplied by the impact factor of the respective metals (from the database ReCiPe environmental impact assessment method version 1.08, midpoint, hierarchic perspective [13]), resulting in the total

impact of the leachate, for instance expressed as kg 1,4-dichlorobenzene equivalents per tonne of landfilled S/S treated residue for impact categories related to toxicity.

It is assumed that the leachate is emitted to the environment, resulting in an overestimation of the real environmental impact, as the leachate from landfills is generally collected and purified in a waste water treatment plant. The environmental impact of cement production is taken from the LCA database Ecoinvent v2. Blast furnace slag (BFS) cement was selected for the assessment, as it is very unlikely that Portland cement is used for low-end applications such as S/S treatment. We assume that 300 kg of cement is used per tonne of residue.

For e.g. the impact category “human toxicity”, the impact is calculated analogously to Vermeulen et al. [14] as:

$$HT = \sum_i HTP_i \cdot m_{em,i} + I_{HT,CEM} \cdot m_{CEM}$$

The impact in the category human toxicity (HT, expressed as kg 1,4-dichlorobenzene equivalents per tonne of landfilled S/S treated residue) is thus calculated as the sum of the direct and the indirect contribution. The direct contribution is a summation of the emitted mass of all relevant pollutants i ($m_{em,i}$), each multiplied by their respective human toxicity potential (HTP_i). The indirect contribution is calculated by summing the mass of cement added to the MSWI residue during S/S treatment (m_{CEM}), multiplied by the impact factor of the production of cement for the impact category “human toxicity” ($I_{HT,CEM}$). Because the S/S treatment without cement has only a small influence on the landfilled mass and volume, and hence on the impact of the infrastructure and transport, which are already low compared to the direct impact and the impact of cement production [14], the changes of these indirect impacts were not taken into account. For other considered impact categories the environmental impact was calculated similarly to the impact in the category “human toxicity”.

The resulting environmental impact for both scenarios, i.e. S/S treatment with and without the use of cement, is normalised based on the annual emissions per European inhabitant in 2000, as reported in ReCiPe version 1.08.

3. Results and discussion

3.1 Heavy metal and oxyanion leaching

Tables 1 and 2 show the metal leaching of the FA and APC residues, untreated, S/S with only water, and S/S with cement and water, for the regulated liquid/solid ratios of 2 (Table 1) and 10 (Table 2). For both leaching tests the WAC for hazardous waste landfills from EU Council decision 2003/33/EC [11] are shown, together with the WAC for non-hazardous waste landfills.

Untreated FA complies with the WAC for hazardous waste landfills and could be landfilled without prior treatment. This is not the case for the untreated APC residue, since the Pb leaching exceeds the WAC significantly, so that treatment, e.g. S/S, is necessary before landfilling. Contrarily to our expectations, S/S treatment with curing for 40 days did not reduce Pb leaching from the APC residue to below the WAC, even with the use of cement (Table 1 and 2). In fact, Pb leaching remained at least one order of magnitude higher than the WAC for hazardous waste landfills. However, in the APC residue samples to which cement was added during the S/S treatment, the Pb leaching decreased more than in the APC residue samples without cement addition. Table 1 and 2 proves that for Pb S/S treatment not only decreases leaching due to the precipitation of $\text{Pb}(\text{OH})_2$, as the leachate pH of the different materials is similar. Instead, the strongly decreased Pb leaching may be attributed to the formation of double hydroxides $\text{Ca}(\text{OH})_2 \cdot x\text{M}(\text{OH})_z \cdot y\text{H}_2\text{O}$, such as $\text{CaPb}_2(\text{OH})_6$ or $\text{CaPb}(\text{OH})_4$ [2,15]. In addition, according to Chen et al. [2], heavy metals may sorb on calcium silicate gel (C-S-H), formed when cement reacts with water, may physically sorb to charged hydrated cement particles and Pb^{2+} may replace Ca^{2+} in crystal lattices.

For the FA samples the addition of cement had no positive effect on the Pb leaching, which was already low. In contrast with the APC residue samples, the Pb leaching is higher in the FA samples to which cement was added, probably due to the higher pH, although it remained below the WAC for non-hazardous waste landfills.

Table 1 Average leaching of heavy metals for a L/S of 2 of untreated FA and APC residue, of both residues S/S with only water and of both residues S/S with cement and water (duplicates, 40 days). The underlined values exceed the WAC for non-hazardous landfills, those in bold exceed the WAC for hazardous landfills.

<u>L/S = 2</u> [mg/kg]	pH	As	Ba	Cd	Cr	Cu	Mo	Ni	Pb	Sb	Zn
FA	10.6	0.16	0.45	0.02	2.21	0.05	4.68	0.03	1.65	<u>0.41</u>	0.24
FA+w	10.3	0.04	0.25	0.02	0.42	0.07	2.77	0.01	0.75	0.19	0.24
FA+CEM+w	12.5	0.13	0.22	0.04	<u>4.58</u>	0.10	<u>19.9</u>	0.01	4.25	-	2.79
APC	11.7	<u>1.01</u>	<u>88.7</u>	0.03	0.19	7.20	1.66	0.54	2130	0.18	<u>37.4</u>
APC+w	11.8	<u>1.04</u>	<u>33.0</u>	0.02	0.21	2.93	1.31	0.47	1109	0.10	<u>51.0</u>
APC+CEM+w	12.0	<u>1.30</u>	<u>53.2</u>	0.03	0.37	0.73	0.90	0.54	291	-	8.76
WAC hazardous		6	100	3	25	50	20	20	25	2	90
WAC non-hazardous		0.40	30	0.60	4	25	5	5	5	0.2	25

Table 2 Average leaching of heavy metals for a L/S of 10 of untreated FA and APC residue, of both residues S/S with only water and of both residues S/S with cement and water (triplicates, 40 days). The underlined values exceed the WAC for non-hazardous landfills, those in bold exceed the WAC for hazardous landfills.

<u>L/S = 10</u> [mg/kg]	pH	As	Ba	Cd	Cr	Cu	Mo	Ni	Pb	Sb	Zn
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FA	10.3	0.17	1.72	0.06	2.73	0.26	2.16	0.04	3.00	0.85	0.74
FA+w	10.0	0.05	1.03	0.04	3.15	0.75	2.79	4.14	2.60	0.43	0.89
FA+CEM+w	12.2	0.06	0.98	0.05	3.83	0.39	<u>17.1</u>	0.48	4.48	-	8.17
APC	12.3	1.39	87.5	0.03	0.41	10.4	3.95	0.10	<u>1042</u>	0.12	<u>69.1</u>
APC+w	12.4	1.27	35.9	0.04	0.67	3.77	2.52	0.34	<u>389</u>	0.05	<u>63.3</u>
APC+CEM+w	12.5	1.46	56.0	0.01	0.59	1.27	1.95	0.85	<u>191</u>	-	28.0
WAC hazardous		25	300	5	70	100	30	40	50	5	200
WAC non-hazardous		2	100	1	10	50	10	10	10	0.7	50

The beneficial effect of S/S treatment observed for the leaching of heavy metals, such as Pb and Cu, does not occur for the leaching of oxyanion forming elements, such as Cr and Mo. On the contrary, S/S treatment of the FA with cement increased the leaching of these elements, especially in the case of Mo where the WAC for non-hazardous waste landfills was exceeded after treatment. This effect is caused by the high pH of the S/S treated FA after cement addition.

The pH dependent leaching of the oxyanion forming elements (shown for Mo in Fig. 1a, for the other oxyanion forming elements as supplementary information, online resource) confirms the increase of Mo leaching for high leachate pH (Table 1). The pH dependent leaching behavior of Cr is similar to that of Mo and is not shown. For the S/S treated residues, leaching of Cr, As and Mo is low until approximately pH 12, probably caused by adsorption to portlandite, and incorporation in ettringite at pH 11-12 [16]. The reduced leaching of Sb can be related to the formation of calcium antimonite [17]. Addition of cement to these residues results in higher leachate pH values (Table 1 and 2). At a pH higher than 12, the Mo and Cr leaching from the FA increases strongly (Fig. 1a).

Fig. 1 pH dependent leaching of (a) Mo from FA, untreated and S/S treated with only water, and with cement and water and (b) Pb from APC residue, untreated and S/S treated with only water, and with cement and water. The WAC for hazardous waste landfills is shown as a dotted line, the WAC for non-hazardous landfills as a solid line.

The pH dependent leaching of the heavy metals Pb, Cu, Zn, and Ni is given in the supplementary information (online resource). Only the pH dependent leaching of Pb from APC residue (untreated, S/S treated without cement and S/S treated with cement) is shown in Fig.

1b, as only the Pb leaching from this residue still exceeds the WAC for hazardous waste landfills after 40 days. Pb, Cu, Zn and Ni all show an amphoteric solubility with minimum leached concentrations at different pH values. In general the leaching of these heavy metals in the pH range relevant for FA and APC residue (9-13) is lower for the residues S/S treated with cement.

Fig. 1b shows that the Pb leaching from APC residue S/S treated without cement is lower than the Pb leaching from untreated APC residue, for a similar pH. Moreover, in the most relevant range (pH 10-13), the Pb leaching from APC residue S/S treated with cement is even lower than the leaching from APC residue S/S treated without cement. This confirms S/S treatment not only reduces leaching by $\text{Pb}(\text{OH})_2$ precipitation, but also by precipitation of $\text{CaPb}_2(\text{OH})_6$ or $\text{CaPb}(\text{OH})_4$, adsorption or occlusion by C-S-H [2]. In addition, Fig. 1b shows that the stabilization with cement performs better than without cement, because more C-S-H can be formed.

The FA and APC residue mixture, S/S without cement, has a very high Pb leaching after 40 days (Tables 3 and 4), which is comparable to the Pb leaching from the separate APC residue S/S without cement (Tables 1 and 2). The Pb leaching from the FA and APC residue mixture S/S with cement (Tables 3 and 4) is also comparable to the Pb leaching from the pure APC residue S/S with cement (Tables 1 and 2).

However, after 75 days of curing, the Pb leaching from FA and APC residue mixture S/S with and without cement decreased with a factor 100 or more (Tables 3 and 4), and the samples were in full compliance (for all of the regulated heavy metals) with the WAC for both hazardous and non-hazardous waste landfills. The reduction of the Pb leaching as a function of time is mainly caused by a pH reduction (Fig. 1b) due to carbonation, assisted by an increased capacity of the C-S-H gel of retaining heavy metal cations and heavy metal hydroxyl ions due to carbonation of C_3S [2].

Table 3 Average leaching of heavy metals for a L/S of 2 of FA and APC residue S/S with only water and of both residues S/S with cement and water (duplicates, 40 and 75 days). The underlined values are higher than the WAC for non-hazardous landfills, values in bold higher than the WAC for hazardous landfills.

<u>L/S = 2</u> [mg/kg]	t [d]	pH	As	Ba	Cd	Cr	Cu	Mo	Ni	Pb	Sb	Zn
FA+APC+w	40	12.0	1.04	20.6	0.02	0.38	1.66	2.51	0.37	<u>636</u>	0.02	24.8
FA+APC+w	75	10.8	-	6.76	0.10	0.53	1.66	-	0.44	0.66	0.14	-
FA+APC+CEM+w	40	12.1	1.06	14.9	-	0.54	0.20	1.93	0.48	<u>134</u>	-	4.53
FA+APC+CEM+w	75	11.6	-	4.90	0.02	0.87	1.02	-	0.49	1.08	0.11	-
WAC hazardous			6	100	3	25	50	20	20	25	2	90
WAC non-hazardous			0.40	30	0.60	4	25	5	5	5	0.2	25

Table 4 Average leaching of heavy metals for a L/S of 10 of FA and APC residue S/S with only water and of both residues S/S with cement and water (triplicates, 40 and 75 days). The underlined values are higher than the WAC for non-hazardous landfills, values in bold higher than the WAC for hazardous landfills.

<u>L/S = 10</u> [mg/kg]	t [d]	pH	As	Ba	Cd	Cr	Cu	Mo	Ni	Pb	Sb	Zn
FA+APC+w	40	12.5	1.10	25.2	0.07	1.51	2.87	5.26	1.61	<u>389</u>	0.01	70.0
FA+APC+w	75	10.9	1.11	7.6	0.07	1.17	3.09	6.86	0.62	1.64	0.16	18.7
FA+APC+CEM+w	40	12.5	0.98	18.0	0.01	1.18	0.17	4.89	0.59	<u>116</u>	0.01	13.3
FA+APC+CEM+w	75	11.6	1.15	6.05	0.03	2.03	1.67	3.61	0.62	1.10	0.06	2.35
WAC hazardous			25	300	5	70	100	30	40	50	5	200
WAC non-hazardous			2	100	1	10	50	10	10	10	0.7	50

3.2 Chloride and sulfate leaching

Leaching of chloride and sulfate ($L/S = 2$) from the 6 S/S residues, and the 2 reference untreated residues, is shown in Fig. 2. S/S treatment, with or without the use of cement, does not reduce the leaching of chloride and sulfate significantly. The WAC for hazardous and non-hazardous waste landfills for chloride and sulfate are indicated in Fig. 2. The FA contains more sulfate than chloride, because most alkali and alkaline earth sulfates are more stable and less volatile than their chlorides. As a result, more chloride than sulfate leaches from the APC residue. The sulfate leaching from APC residue of the considered installation (using both lime and sodium bicarbonate) is expected to be higher than from APC residue of an installation using only lime, because of the high solubility of sodium sulfate salts compared to calcium sulfate salts. The sulfate limits are exceeded for all FA samples, the chloride limits are exceeded for all APC residue samples.

For granular waste material, the WAC in EU Council decision 2003/33/EC [11] specify leaching standards for chloride and sulfate, shown in Fig. 2. However, when the waste is landfilled as monolithic material, national legislations might not include limits for chloride or sulfate leaching, as is the case in e.g. Flanders (VLAREM II 5.2.4 [18]), because EU Council decision 2003/33/EC [11] mentions that it is up to the member states to set standards for monolithic waste. Lampris et al. [19] showed based on monolithic leaching tests that the physical immobilization of chloride in S/S treated APC residues is very low. Whether or not the S/S treated waste is landfilled as monolithic or granular material, Fig. 2 illustrates that chloride and sulfate leaching from waste S/S without cement is similar to leaching from waste S/S with cement. The effect on soil and water salinity will therefore also be similar.

Fig. 2 Leaching of chloride and sulfate ($L/S = 2$) from untreated FA and APC residue, S/S treated FA and APC residue with only water and from both residues S/S treated with water and cement (duplicates). The solid line gives the WAC for non-hazardous waste landfills for both chloride and sulfate, the dashed and dotted line the WAC for hazardous waste landfills for chloride and sulfate.

3.3 Environmental impact

The human toxicity potential (Human Tox.), calculated using the leaching values of the S/S FA and APC mixtures after 75 days given in Tables 3 and 4 (for a L/S of 2 and 10), comparing the

mixtures S/S with only water and the mixtures S/S with cement and water is shown in Fig. 3. After 75 days of curing, the S/S treated FA and APC mixtures comply with the WAC for hazardous waste landfills, allowing landfilling, meaning that longer curing times are not applied in practice. The impact of leachate from the mixtures S/S with cement is slightly lower than the impact of leachate from the mixtures S/S treated without cement, which is mainly due to the higher Ba, Mo and Zn leaching from the samples without cement. However, the production of the blast furnace slag cement used in the S/S treatment significantly increases the impact on human toxicity of landfilling these residues. As a result, considering the uncertainties on the leachate concentrations, the impact on human toxicity of both treatment methods is similar, especially for an L/S of 10.

Fig. 3 Impact in the impact category Human Toxicity of the landfilled FA and APC residue mixtures S/S with or without cement after 75 days of curing, leached at L/S of 2 and 10, caused by leachate emissions and by emissions from cement production.

In other toxicity related impact categories, fresh water ecotoxicity (Freshw. Ecotox.) and terrestrial ecotoxicity (Terr. Ecotox.), the impact of the residues S/S with cement is clearly higher than the impact of the residues S/S without cement, as shown in Fig. 4. The use of cement for S/S treatment of MSWI residues also causes an environmental impact in the impact categories climate change and terrestrial acidification (Terr. Acid.), due to the production of BFS cement (Fig. 4). The impact in the category photochemical oxidant formation (Photochem. Ox.) is negligible. The magnitude of the environmental impact of landfilling S/S MSWI residues strongly depends on the type of cement used, Portland cement causing a higher impact than BFS cement, which is partly composed of a secondary raw material.

Fig. 4 Normalised environmental impact, in % of the yearly emissions per European inhabitant in 2000, of S/S treated FA and APC residue mixtures with and without cement, leached with a L/S of 2 and 10.

The environmental impact calculated here, which is based on batch leaching experiments, is in many cases an overestimation of the real impact. In modern landfills, leachate is collected and physico-chemically treated, resulting in lower metal concentrations in the discharge water. Moreover, leaching from landfilled waste in monolithic form might be lower than from granular material, due to the reduced contact with water, leading to lower emissions. However, the purpose of this paper was to evaluate the difference between the environmental impact of S/S treatment with cement and without cement, not to accurately determine the environmental impact of landfilling MSWI residues. The exact environmental impact of landfilling FA and APC residue depends strongly on site specific parameters, such as water infiltration, the presence of synthetic liners, leachate pH and treatment etc., and should hence be determined site specifically.

Because of the aforementioned reasons, the environmental impact of the heavy metal leaching from the S/S treated FA and APC residue shown in Fig. 4, estimated by batch leaching tests, should be considered an upper limit indication for the environmental impact of modern landfill leachate. This implies that the relative contribution of the indirect impact of the production of cement, shown in Fig. 4, to the total environmental impact is underestimated.

4. Conclusions

Lab scale experiments showed that solidification/stabilization significantly reduces the leaching of heavy metals from fly ash and (lime + bicarbonate based) air pollution control residue of a modern MSWI. Especially the leaching of Pb from untreated APC residue, highly exceeding the WAC for hazardous waste landfills, was reduced below this WAC for a mixture of fly ash and APC residue after 75 days of curing. It was shown that Pb stabilization occurs by precipitation of $\text{CaPb}_2(\text{OH})_6$ or $\text{CaPb}(\text{OH})_4$, or occlusion by C-S-H, so not only by precipitation

of $\text{Pb}(\text{OH})_2$. Carbonation of the S/S treated material, resulting in a lower pH than the initial materials, was needed to comply with the WAC for Pb.

Although the use of cement for solidification/stabilization is common practice, it is unnecessary, given the cementitious properties of fly ash and air pollution control residues, for the following reasons: (i) after a sufficient amount of time the leaching values of heavy metals are similar for these MSWI residues solidified/stabilised with and without cement and below the WAC for non-hazardous waste landfills, (ii) the environmental impact of the production of cement in the impact categories climate change, human toxicity, fresh water and terrestrial ecotoxicity and terrestrial acidification can be avoided, (iii) cement has a positive economic value and increases the mass and volume of the landfilled material.

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