



On-site treatment and landfilling of MSWI air pollution control residues

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Abstract

Air pollution control (APC) residues from municipal solid waste incineration (MSWI) are difficult to landfill due to substantial leaching of trace metals. An on-site pretreatment prior to landfilling of APC-residues was investigated in terms of bench-scale experiments with a semidry APC-residue and a fly ash. The treatment involved mixing of the residues with a ferrous sulphate solution and subsequent oxidation of the suspension. Afterwards, the suspension was spread on a dedicated landfill section and allowed to drain by gravity through the drainage system of the landfill. The wastewater from the process, collected through the drainage system, contained large concentrations of salts (Cl: 14–30 g/l, Na: 4–9 g/l, K: 5–11 g/l, Ca: 2–12 g/l) but low concentrations of trace metals (e.g. Pb: 14–100 µg/l, Cd: <2–7 µg/l). The treated residues left in the landfills were later subject to leaching by simulated rainfall. The leachate contained low concentrations of trace metals (Pb: <120 µg/l, Cd: <2 µg/l, Cr: <485 µg/l). The leachate concentrations from the treated APC-residues were substantially reduced compared to concentrations in leachate from untreated APC-residues. Particularly in the early stages of the leaching, concentrations of trace metals were reduced by up to four orders of magnitude.

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1. Introduction

Air pollution control (APC) residues from municipal solid waste incineration (MSWI) is produced in large amounts in all countries, where MSWI is an important part of the waste

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management system. For example, in Denmark approximately 70 000 ton APC-residues are produced annually. The residues are considered hazardous waste in several countries and are difficult to dispose off due to the leaching of high concentrations of salts and trace metals, especially Pb [1,2]. In the future, this problem is likely to increase within the EU as incineration of waste probably will increase, due to the implementation of the new EU-directive on landfilling of waste adopted on 26 April 1999 [3,4] prescribing a significant reduction in biodegradable waste going to landfills. Pretreatment of the APC-residues before disposal will be needed [3,4]. Methods to reduce leaching from APC-residues include separation processes, solidification, chemical stabilization, encapsulation, sintering, and vitrification [1,5].

A new treatment process called the Ferrox-process has proven to be able to reduce the leaching of Pb, Cd, and other trace metals substantially [6]. This process builds on the well-established geochemical knowledge that many iron oxides are able to bind substantial amounts of trace metals [7–9]. The Ferrox-process is carried out by mixing the APC-residue with a ferrous sulphate-solution and subsequently aerating the suspension. The aeration oxidizes the precipitated ferrihydroxide to ferrihydrite. Afterwards, the suspension is separated into treated residue and wastewater. By this treatment salts are washed out whereas the trace metals are retained in the solids, thereby leading to less leaching of salts and metals from the treated APC-residue. The wastewater separated from the solids has a high content of salts and a low content of trace metals. The ferrous sulphate used in the process is a by-product in titanium production and is therefore available at low cost.

This paper describes, in terms of bench-scale experiments, how the Ferrox-process can be performed as a simple on-site treatment prior to landfilling of the APC-residues. The process is carried out as a single stage process in a mixing/aeration reactor at the landfill and the suspension is discharged onto the landfill cell, using the drainage system of the landfill to separate the wastewater from the solids.

2. Materials and methods

The experiments involved two different APC-residues from two Danish MSWI plants: a semidry gas cleaning residue containing fly ash and lime (SD) and a fly ash from an electrostatic precipitator (FA). Typical contents of selected elements in these residues are given in Table 1.

APC-residue (800 g) was mixed with a ferrous-sulphate solution (4 l) corresponding to 50 g Fe²⁺/kg semidry APC-residue and 25 g Fe²⁺/kg fly ash. The amount of acidic iron sulphate added was limited by the easily-available alkalinity of the materials in order to maintain pH high for easy precipitation of iron as ferrihydroxide. After mixing the APC-residue with the ferrous-sulphate solution, the suspension was oxidized for 24 h with atmospheric air. The suspension changed colour from black to reddish brown after 3 h of aeration. After the aeration, the suspension was transferred gently to a model landfill with a surface area of 0.2 m². The bottom of the landfill contained four different layers of sand and gravel with a drainage system in the bottom layer as shown in Fig. 1. The drainage layer was prewashed with deionized water.

Table 1
Typical content of selected elements in semidry APC-residue (SD) and fly ash (FA)

	SD	FA
Cl	169	109
Ca	271	183
K	24	46
Na	26	38
S	30	26
Si	0.7	1.2
Sr	0.4	0.3
Al	28	44
Ba	0.64	0.81
Cd	0.15	0.21
Co	0.01	0.02
Cr	0.11	0.23
Cu	0.79	1.5
Fe	5.6	12
Hg	0.025	0.0015
Mo	0.012	0.034
Ni	0.032	0.066
Pb	3.98	6.37
Zn	14.7	17.0

Units are g/kg.

On the model landfill, the water of the suspension percolates off through the drainage system (called wastewater) and the moist solids are left on top of the sand/gravel layers. Fig. 2 illustrates the on-site Ferrox-process. This procedure was repeated 25 times over a period of 2 months. Hence, 25 layers of treated residue were built up on top

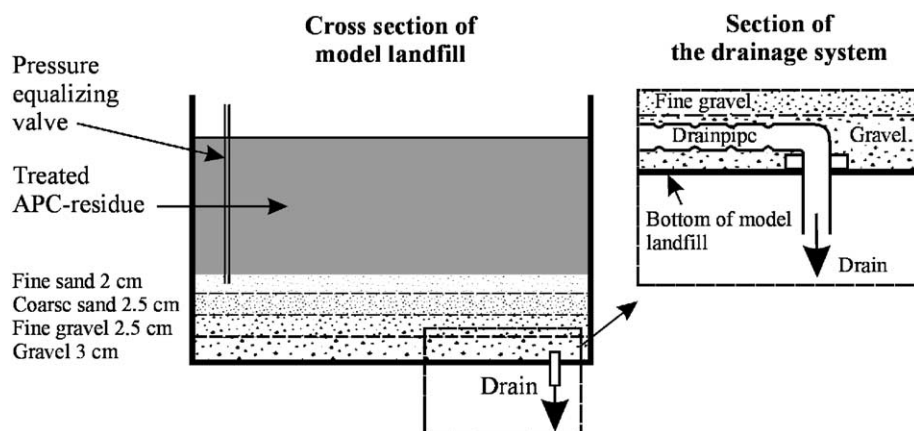


Fig. 1. Structure of a model landfill. The numbers show the thickness of the various layer. The drainage system sand/gravel consisted of material of different particle size: fine sand, 0.13–0.50 mm; coarse sand, 0.63–1.40 mm; fine gravel, 1.25–2.24 mm; and gravel, 2.00–3.55 mm.

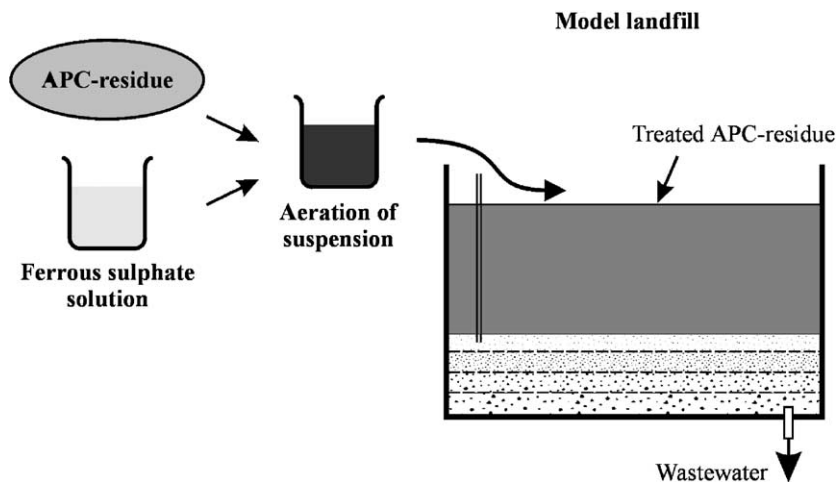


Fig. 2. Overview of the on-site pretreatment and landfilling of APC-residues.

of each other yielding a total thickness of 13–15 cm. The wastewater was collected and sampled 25 times, once for each new layer. The material was compacted only by gravity.

After establishment of the two model landfills, one for each type of treated APC-residue, they rested for 2 months, before leaching by simulated rainfall started. The surface of the model landfills was sprayed with deionized water at pH 4 (adjusted with concentrated HNO_3) in amounts corresponding to a liquid to solid ratio (L/S) of 0.25 l/kg twice a week. The leachate was collected through the drainage system and sampled.

Parallel to the leaching of model landfills containing treated residues, model landfills containing untreated residues were established. This was done by humidifying the two untreated residues, corresponding to a moisture content of 20% for SD and 25% for FA. This equals the humidifying done at the full scale MSWI prior to landfilling of the residues. The moist residue was then disposed off on a model landfill, identical to the one shown in Fig. 1, in batches equal to 800 g of dry matter. This was done 25 times over a period of three months. After another 2 months, the model landfills with the untreated residues were leached by simulated rainfall in the same way as the model landfills with treated residues.

Samples were filtered through a 0.4 μm polypropylene filter and preserved with concentrated HNO_3 (pH between 1 and 2). Furthermore, selected samples of wastewater and leachate from the model landfills containing treated residues were also taken and not filtered in order to investigate the role of large colloids as carriers of metals. All samples were measured with respect to pH (measured in a flow cell at the outlet of the drainage system), specific conductivity, Pb, Cd, and Cr. Pb, Cd, and Cr were analysed by atomic absorption spectroscopy, using graphite furnace or flame techniques. In addition selected samples were analysed for a broader spectrum of parameters by inductively coupled plasma–optical emission spectroscopy (ICP–AES), inductively coupled plasma–mass spectroscopy (ICP–SMS);

Sector, and atomic fluorescence (AFS). The analysis was performed by SGAB Analytica AB, Luleaa, Sweden and is accredited by Swedac (accepted in a range of countries in Europe and in the US) for all elements except Cl and S.

3. Results and discussion

3.1. Wastewater

During active operation of the on-site pretreatment and landfilling of the residues, the only waste stream leaving the landfill is the wastewater removed through the landfill drainage system. The content of the wastewater, therefore, is of special interest during the period of active operation.

Fig. 3 shows pH and specific conductivity in the collected wastewater as a function of the number of layers landfilled. Initially, the conductivity was low due to dilution with water left from the washing of the drainage system, and the buffer capacity of the drainage layers lowered pH somewhat. Specific conductivity and pH reached a relatively stable level after the first five layers were landfilled. Table 2 shows typical contents of selected elements in the collected wastewater after stable levels were reached.

The content of trace metals in the wastewater collected through the landfill drainage system was similar to the content in wastewater sampled in the reactor at the end of the aeration period as previously reported by [6]. This was also the case for elements such as Cl, Na, K, and Ca. This suggests that filtration through the landfilled, treated APC-residue did not significantly affect the composition of the wastewater. The pH was 7.0–8.1 (semidry APC-residue) and 6.6–9.8 (fly ash) in the wastewater collected through the landfill drainage system suggesting that spreading of the suspension on top of the landfill has allowed for partial carbonatization (uptake of carbon dioxide from the air) and development of pH values just above neutral. The pH of the wastewater after aeration, but prior to carbonatization, is in the range 10.8–11.0 [6].

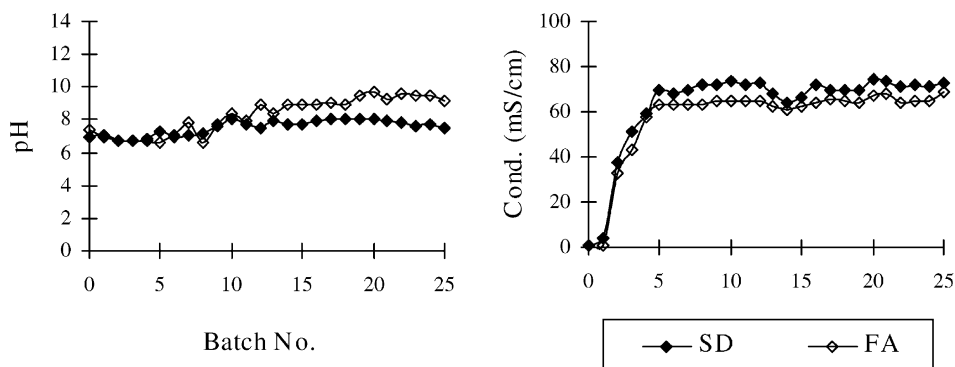


Fig. 3. pH and specific conductivity in the wastewater as a function of the number of batches of treated residues landfilled. SD is a semidry APC-residue and FA is a fly ash.

Table 2
Composition of wastewater from model landfills containing treated residues

	SD	FA
pH	7.0–8.1	6.6–9.8
Specific conductivity (mS/cm)	64–75	61–69
Cl (g/l)	28–30	14–24
Ca (g/l)	11–12	2.1–2.3
K (g/l)	4.6–5.0	9.5–11
Na (g/l)	4.1–4.7	7.5–8.6
S (g/l)	0.36–0.39	1.0–1.1
Al ($\mu\text{g/l}$)	45–114	66–240
Ba ($\mu\text{g/l}$)	3500–5500	375–670
Cd ($\mu\text{g/l}$)	<2–3.7	<2–7.1
Co ($\mu\text{g/l}$)	<34	<5
Cr ($\mu\text{g/l}$)	<60	700–1100
Cu ($\mu\text{g/l}$)	<40–56	<40–90
Fe ($\mu\text{g/l}$)	<100–200	<100
Hg ($\mu\text{g/l}$)	0.63–20	0.23–3.4
Mo ($\mu\text{g/l}$)	910–1020	2380
Ni ($\mu\text{g/l}$)	<380	<53
Pb ($\mu\text{g/l}$)	14–86	19–100
Zn ($\mu\text{g/l}$)	36–290	<25–216

SD is a semidry APC-residue and FA is a fly ash.

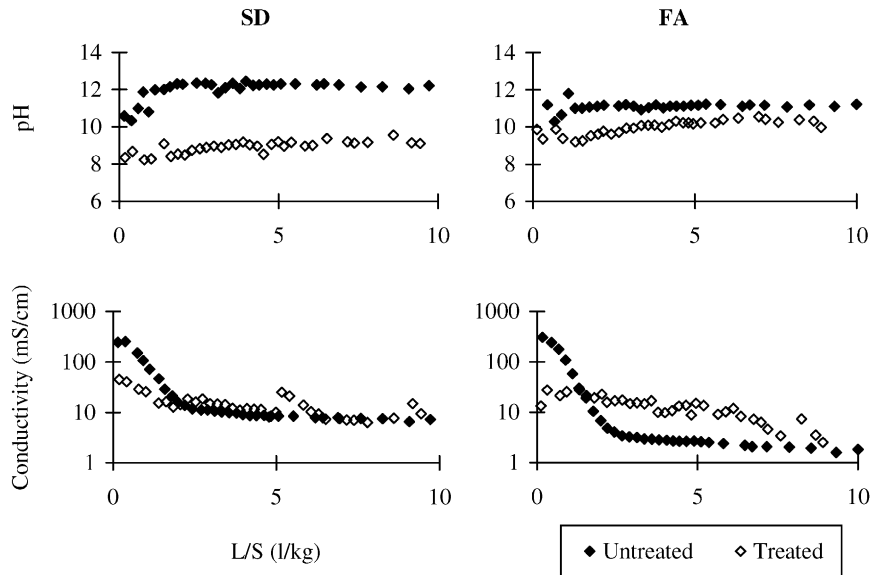


Fig. 4. pH and conductivity in the leachate from model landfills with treated and untreated APC-residues as a function of the amount of rainfall infiltrating through the landfill expressed as the liquid to solid ratio (L/S). SD is a semidry APC-residue and FA is a fly ash.

The content of Pb (14–100 µg/l) and Cd (2–7 µg/l) in the wastewater was low, but wastewater from treatment of FA contained up to 1 mg/l of Cr. Reducing the Cr content might be needed prior to discharge of the wastewater and ongoing research focuses on this issue.

Filtration at 0.4 µm did not significantly change the composition of the wastewater compared to no filtration (data not shown). Thus, any metals associated with colloids in the wastewater greater than 0.4 µm were effectively removed by the filtration through the land-filled material.

The on-site treatment yields wastewater with a high content of salts. This can, due to low contents of trace metals, potentially be discharged to marine waters, possibly after reducing pH by a simple aeration. The latter has been tried out and is without any difficulties [6]. In non-coastal areas evaporation of the wastewater maybe an alternative approach, recovering the salts for industrial use or use as a de-icing agent on roads.

3.2. Leaching

After on-site pretreatment and landfilling, the residues in the landfill was subject to leaching by infiltrating rainfall, here simulated by deionized acidic water.

Fig. 4 shows that pH in the leachate was 1–4 units lower from the treated residues than from untreated residues. This is partly due to the FeSO₄-treatment and partly to the

Table 3
Leachate concentrations from model landfills with treated and untreated semidry APC-residue (SD) at L/S ratios corresponding to the initial leachate and to L/S ratios between 5 and 61/kg

	Treated SD		Untreated SD	
L/S (l/kg)	0.18	5.8	0.15	5.5
pH	8.35	8.97	10.59	12.30
Specific conductivity (mS/cm)	45	14	240	8.4
Cl (mg/l)	13000 ^{a, b}	6100	310000	280
Ca (mg/l)	5800	2300	65000	1100
K (mg/l)	2700	890	40000	62
Na (mg/l)	2600	890	37000	720
S (mg/l)	280	260	110	290
Si (mg/l)	2.0	1.3	0.71	0.30
Sr (mg/l)	22	8.8	250	3.4
Al (µg/l)	230	370	4800	11
Ba (µg/l)	1400	210	37000	100
Cd (µg/l)	0.41	0.54	1100	0.07
Co (µg/l)	0.81	0.064	2.2	0.36
Cr (µg/l)	7.8	11	390	540
Cu (µg/l)	3.4	1.6	2100	4.0
Fe (µg/l)	<0.04	70	6100	18
Hg (µg/l)	0.05	1.7	2.6	1.3
Mo (µg/l)	510	290	390	29
Ni (µg/l)	2.9	0.28	<15	1.0
Pb (µg/l)	85	38	4300000	6000
Zn (µg/l)	24	4.8	32000	460

^a At L/S 0.41 l/kg.

^b At L/S 5.4 l/kg.

Table 4

Leachate concentrations from model landfills with treated and untreated fly ash (FA) at L/S ratios corresponding to the initial leachate and to L/S ratios between 5 and 6 l/kg

	Treated FA		Untreated FA	
	0.12	5.6	0.16	5.8
L/S (l/kg)	0.12	5.6	0.16	5.8
pH	9.86	10.21	11.19 ^a	11.20
Specific conductivity (mS/cm)	13.2	9.1	310	2.4
Cl (mg/l)	3000	2900 ^b	390000	25
Ca (mg/l)	340	490	4500	450
K (mg/l)	2700	1100	85000	98
Na (mg/l)	1400	880	70000	800
S (mg/l)	250	430	550	460
Si (mg/l)	2.0	3.5	1.0	6.0
Sr (mg/l)	2.4	2.0	52	2.0
Al (μg/l)	480	670	230	420
Ba (μg/l)	96	64	1100	70
Cd (μg/l)	0.091	0.050	3500	0.10
Co (μg/l)	0.053	0.028	<2	0.03
Cr (μg/l)	108	440	220	460
Cu (μg/l)	2.6	1.7	35	1.0
Fe (μg/l)	36	17	54	20
Hg (μg/l)	0.53	0.46	3.0	<0.01
Mo (μg/l)	420	390	3500	240
Ni (μg/l)	0.79	0.30	<17	1.0
Pb (μg/l)	7.1	1.6	1600000	8.0
Zn (μg/l)	5.0	7.2	<68	16

^a At L/S 0.45 l/kg.

^b At L/S 5.2 l/kg.

carbonization taking place when the treated APC-residue is spread on the landfill. Fig. 4 also shows that the specific conductivity of the initial leachate from the treated residue was reduced by at least a factor of 10. This is due to the removal of salts such as Cl, Na, K, and Ca during the pretreatment, as also suggested by the composition of the wastewater (Table 2).

Tables 3 and 4 show the composition of the initial leachate and leachate corresponding to a L/S of 5–6 l/kg. In the initial leachate, the concentrations of salts (Cl, Na, K, and Ca) were one to two orders lower in the leachate from the treated residues than in the leachate from the untreated residues. The concentrations of trace metals in the initial leachate from the treated residues were low, whereas the initial leachate from the untreated residues contained high concentrations of especially Pb and Cd (for SD also Cu and Zn). After leaching to L/S 5–6 l/kg the concentrations of salts in the leachate appeared higher for the treated than for the untreated residues. For SD this may not be the general trend as the specific conductivity, as shown in Fig. 4, was almost equal for untreated and treated SD except at L/S-ratios between 5 and 6 l/kg. The leachate after L/S 5–6 l/kg had for both untreated and treated residues a fairly low content of trace metals except for untreated SD, where the leachate contained 6000 μg/l Pb.

Fig. 5 shows the accumulated leaching of Pb, Cd, and Cr as a function of L/S for the model landfills with treated SD and FA and for the model landfills with untreated SD and

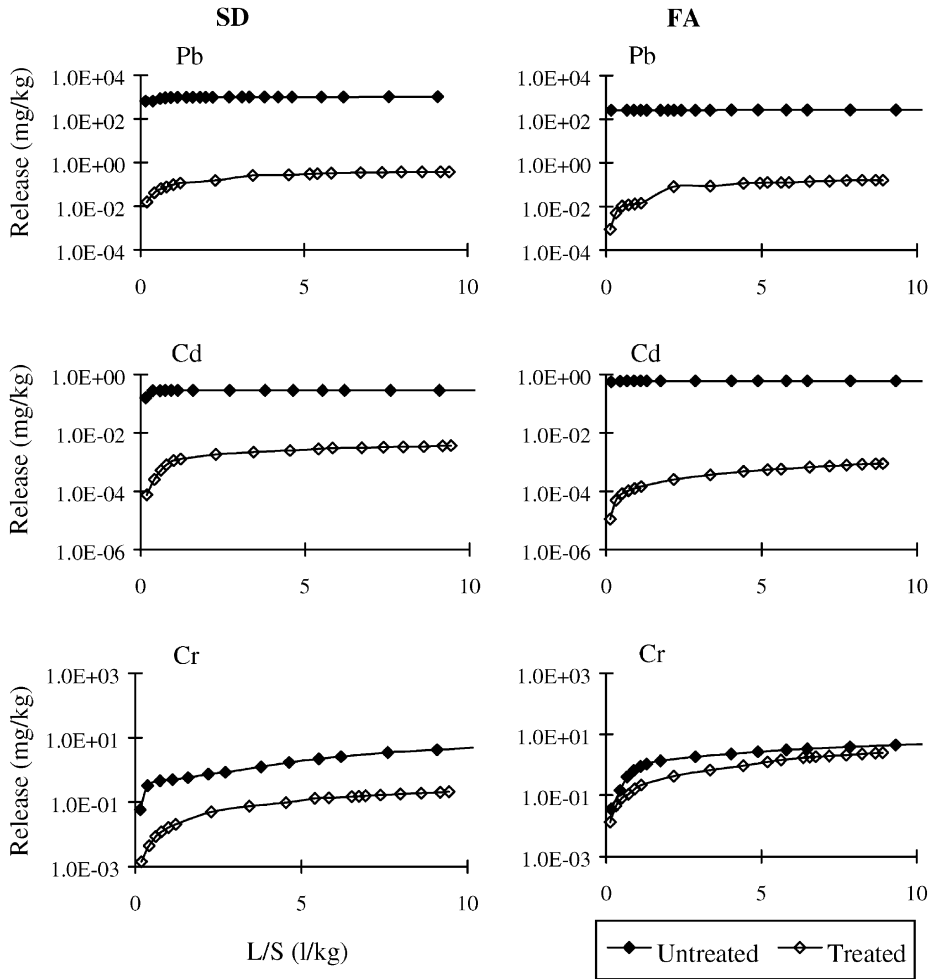


Fig. 5. Leaching of Pb, Cd, and Cr from model landfills with treated and untreated APC-residues as a function of the amount of rainfall infiltrating through the landfill expressed as the liquid to solid ratio (L/S). SD is a semidry APC-residue and FA is a fly ash.

FA. The pretreatment of the residues reduced the initial leaching by up to four orders of magnitude. The reduction in leaching is partly due to the lowering of pH by the treatment [2] but certainly also to the binding of metals by the introduced ferrihydrite. The latter has been previously demonstrated by pH-static leaching test [6]. In the case of Cd, also the removal of chloride, resulting in less complexation of Cd, may have contributed to the reduction in leaching from the pre-treated residues.

Fig. 6 shows metal concentrations of filtered and unfiltered leachate samples from the two model landfills with treated residues suggesting that Pb and Cd occasionally were associated with colloids larger than $0.4 \mu\text{m}$, whereas Cr was found as dissolved Cr or on

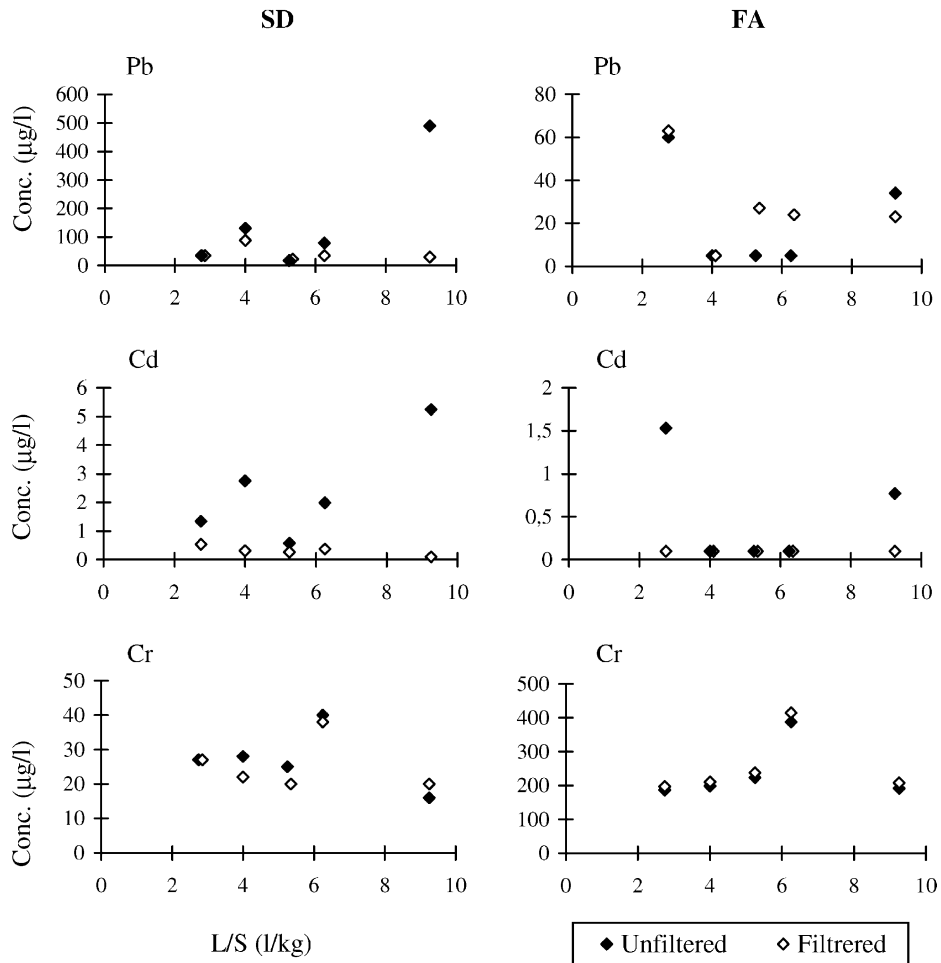


Fig. 6. Concentrations of Pb, Cd and Cr in filtered and unfiltered selected samples of leachate from treated APC-residues. SD is a semidry APC-residue and FA is a fly ash. Filtration was done with 0.4 µm filters.

colloids smaller than 0.4 µm. This is in compliance with the expected behaviour of these elements [2]. However, the concentrations of Pb, Cd, and Cr associated with colloids larger than 0.4 µm was, in all cases, low. This suggests that the drainage system used generally was able to prevent migration of Pb, Cd, and Cr associated with larger colloids.

3.3. Physical properties

The total thickness of the 25 layers of treated residue was 17 cm for SD and 15 cm for FA. The dry matter content of these layers was 30–32% for SD and 33–36% for FA. Treated SD was soft with low carrying capacity (not quantified), but leaching hardened it, so it became

Table 5

The total release of Pb, Cd and Cr during active and passive operation until L/S 21/kg for untreated/treated residues and the relative reductions in release by treating the residues

	SD			FA		
	Untreated (mg/kg)	Treated (mg/kg)	Reduction (%)	Untreated (mg/kg)	Treated (mg/kg)	Reduction (%)
Pb	990	0.28	99.97	260	0.18	99.93
Cd	0.29	0.0092	96.83	0.61	0.011	98.20
Cr	0.52	0.15	71.15	1.9	2.9	–52.63

SD is a semidry APC-residue and FA is a fly ash.

firm. In contrast, treated FA was firm even before leaching started. During the artificial rain events water ponded on the landfilled material (not quantified). The hydraulic conductivity as well as the geotechnical properties of the material should be further studied prior to designing such a pretreatment and landfilling system.

3.4. General discussion

The pretreatment of the APC-residues produces a wastewater high in salt content, but low in trace metal content, and a solid material that has significantly improved properties with respect to leaching. Pretreating the residues substantially reduces the release of Pb and Cd during passive operation of the landfill. Especially the high release of metals during the first stages of leaching is avoided by pretreating the residues. The first stages of leaching, for example corresponding to L/S 21/kg, may at a full-scale landfill equal approximately 130 years of leaching (percolation: 200 mm/y, estimated dry density: 1.3 kg/l, height of landfill: 10 m). Pretreatment of the APC-residues, therefore, is important both with respect to protecting the surrounding surface and groundwater bodies and with respect to reducing the costs over time of leachate collection and treatment.

The total release of Pb, Cd and Cr during pretreatment (active operation) and leaching of the landfilled material (passive operation) until L/S 2 is shown in Table 5. Release data for the untreated residues are also shown. It is evident that the pretreatment overall significantly reduced the release of Pb (1500–4000 times) and Cd (30–50 times), while Cr was only slightly reduced (0.7 time) for the semidry residue and slightly increased (0.5 time) for the fly ash.

Table 6

The distribution of the total release of Pb, Cd and Cr during active and passive operation until L/S 21/kg for treated residues

	SD		FA	
	Active	Passive	Active	Passive
Pb	50%	50%	67%	33%
Cd	82%	18%	98%	2%
Cr	68%	32%	86%	14%

SD is a semidry APC-residue and FA is a fly ash.

Table 6 shows the distribution of the total release during pretreatment and subsequent leaching until L/S 2 l/kg. For Cd and Cr most of the release was with the wastewater, while Pb was released both with the wastewater and the leachate, but in very low amounts.

4. Conclusion

On-site pretreatment and landfilling of APC-residues seems feasible with the Ferrox-process. The treatment reduced the leaching of Pb and Cd substantially, especially in the initial leachate, where reductions up to three orders of magnitude were observed. The total release of Pb and Cd, including both the release with the wastewater during the pretreatment and the release during leaching, was likewise reduced several orders of magnitude. For most of the elements, the major part of the release was with the wastewater. The wastewater from the process contained low concentrations of Pb and Cd, and moderately low concentrations of Cr. As intended a large amount of salts was removed from the residue during the treatment, leading to a decrease in leaching of salts.

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