



Potential Use of Incineration Bottom Ash in Construction: Evaluation of the Environmental Impact

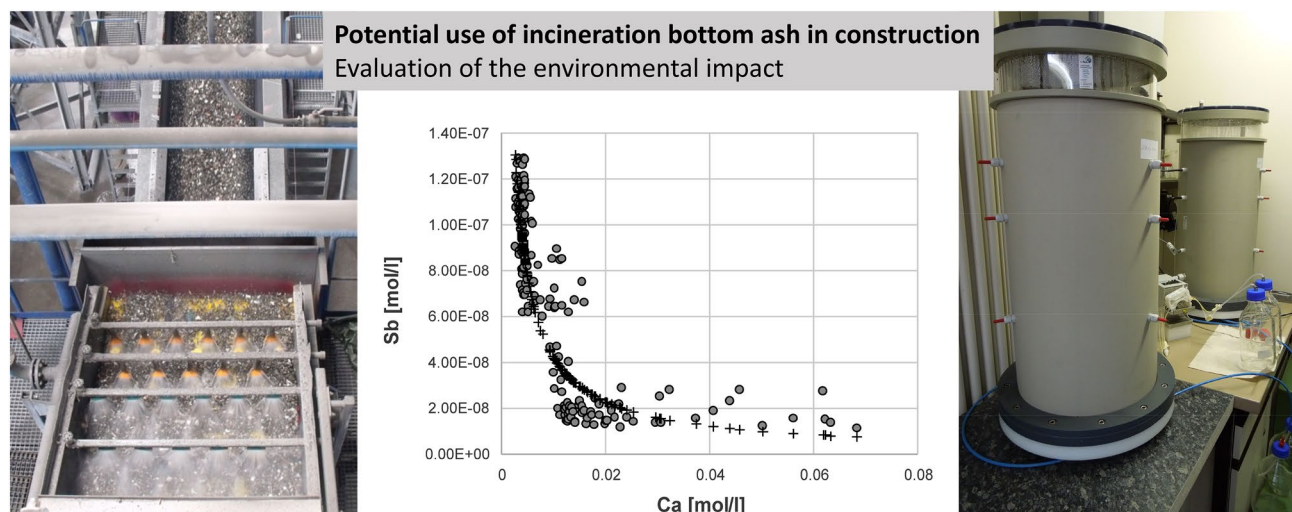
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Abstract

Knowledge of the long-term leaching behavior of potentially harmful substances is crucial for the assessment of the environmental compatibility of reusing municipal solid-waste incineration bottom ash (MSWI BA) in construction, i.e., as a road base layer. BA fractions obtained from wet-processing aiming at the improvement of environmental quality were used to investigate the mobility of relevant substances. Eluates from laboratory-scaled leaching procedures (column percolation and lysimeters) were analyzed to learn about the long-term release of substances. Unsaturated conditions and artificial rainwater (AR) were used in the lysimeter tests to simulate field conditions. In addition, batch test eluates were generated at usual liquid-to-solid ratios (L/S) for compliance testing purposes. A variety of cations and anions was measured in the eluates. The wet treatment reduces the leaching of chloride and particularly sulfate by more than 60%. The release of typical contaminants for the treated MSWI BA such as the heavy metals Cu and Cr was well below 1% in the conducted leaching tests. An increase in the Sb concentration observed in the lysimeter experiments starting at L/S 0.75 L/kg and in the column experiment at L/S 4 L/kg is assumed to be related to decreasing concentrations of Ca and thus to the dissolution of sparingly soluble calcium antimonate. The same leaching mechanism applies with V, but the concentration levels observed are less critical regarding relevant limit values. However, on the long term the behavior of Sb could be problematic for the application of MSWI BA in construction.

Graphic Abstract



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Extended author information available on the last page of the article

Keywords MSWI bottom ash · Leaching · Batch tests · Lysimeter · Antimony

Statement of Novelty

Wet-mechanical treatment is an option to generate secondary raw material from waste incineration residues with high quality regarding environmental compatibility as a surrogate in construction. Especially from long-term lysimeter tests with treated bottom ash, the leaching behavior of such materials was investigated for almost 5 years using field-like conditions. Particularly the unusual leaching behavior of antimony was shown and related to the immobilization of calcium, i.e. increasing antimony concentrations with proceeding ageing. This phenomenon must generally be observed when bottom ash is applied in construction since threshold values might be exceeded in a later stage of lifetime. The findings will induce research on suitable countermeasures to avoid environmental burden.

Introduction

Approximately 68 million tonnes of municipal solid waste (about 27% of total amount) were incinerated in 2017 in the EU-28 countries [1] which resulted in about 15 million tonnes of MSWI BA and represents a valuable source for secondary raw materials. After separation of metals the remaining mineral fraction is mostly reused as unbound aggregate for construction of road base layers. Even though, the environmental behavior of MSWI BA in reuse scenarios in construction has been intensively studied [2–7], there is still a substantial need for research e.g. regarding advanced pre-treatment options to improve the mechanical performance, to reduce the environmental impact and finally to save natural resources as well as landfill space.

In Germany, the regulations on the reuse of mineral waste are in the responsibility of the 16 Federal states so far. Quite stringent threshold values for substances of environmental concern (including Sb) are targeted to be regulated on national level [8]. Even though, harmonized leaching procedures are required by the European Commission [9] and are being developed (e.g. [10]) on European level not all European member states have implemented appropriate regulations. Further, the specification of threshold values remains in the responsibility of the member states so far. Therefore, the risk assessment approaches are different even in countries with well-developed legislation [11].

Considering the waste input [12], operational conditions of incineration [13] and pre-treatment technologies [14–16], the subsequent influence on the leaching behavior has to be investigated to assess a potential environmental risk in

reuse applications [17]. Furthermore, a comparison of data obtained by simple laboratory tests to data obtained under field conditions is of high interest for this purpose [18–20].

A wet-processing technology for municipal solid waste incineration bottom ash (MSWI BA) has been optimized regarding the amount and quality of the fractions produced in the framework of a project funded by ZIM (Central Innovation Program for SMEs of German Federal Ministry for Economic Affairs and Energy). The technology focuses on the mineral fractions, which are the main mass fraction, in order to enable their reuse as construction products. In particular, the aim was the removal of chloride and the transfer of sulfate to the finest fraction. Within the project, we performed accompanying investigations regarding the mass balance of the process and the leaching behavior of the various processed fractions [21].

In this manuscript, eluate analyses from batch leaching, column percolation, and lysimeter tests in laboratory scale were thereby discussed to learn more about the long-term release of hazardous substances from bottom ash in civil engineering applications, e.g. road construction [2, 22].

Particularly, attention was paid on the potentially critical leaching behavior of Sb which is mainly introduced to bottom ash from additives in plastics. The specific behavior of antimony in MSWI BA has been discussed by previous studies [20, 23–26]. It is intended to regulate Sb in MSWI BA in Germany within a new ordinance on the reuse of mineral waste [8, 27].

Experiment

Material

The material was sampled on November 5, 2013 at a bottom ash treatment plant in Germany. Ferrous (Fe) and non-ferrous (NFe) metals had already been removed by standard methods (magnets, eddy current separation). A wet-mechanical process step was implemented in the plant to remove the finest fraction below 0.25 mm [21]. Two mineral fractions (0.25–4 mm and 4–60 mm) were generated by different sieving steps (see Fig. 1).

The mineral fractions from 0.25–4 to 4–60 mm were sampled, air-dried, and homogenized separately. The 4–60 mm fraction was additionally sieved to 45 mm. Representative subsamples of both fractions were then combined in the lab at a ratio of 40:60 to the final 0.25–45 mm test material employed for the leaching experiments, since this condition was expected as the status on an intermittent storage site before reuse of such material.

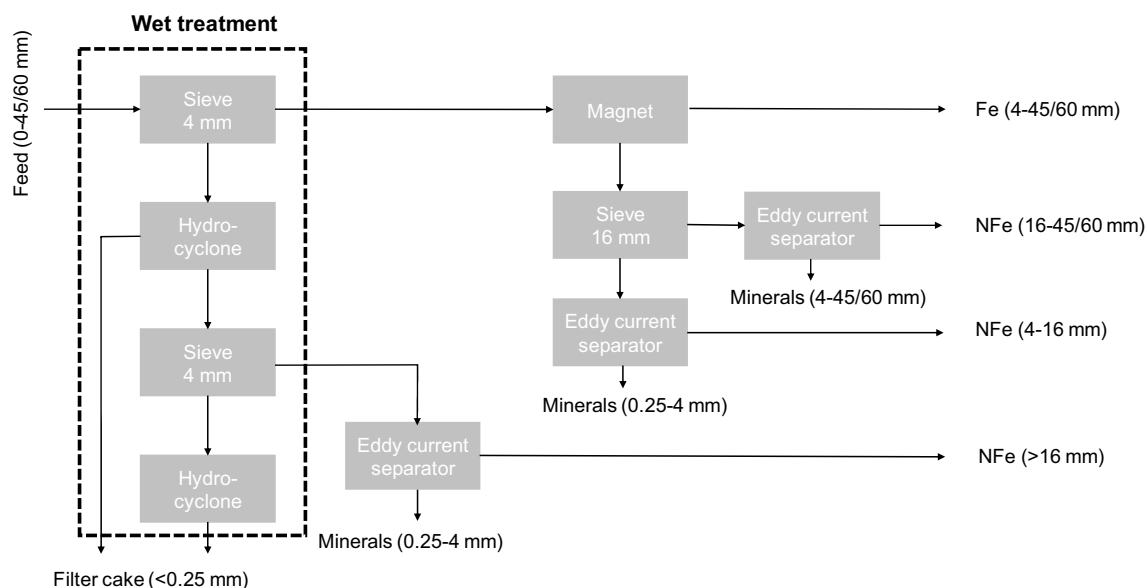


Fig. 1 Sketch of the wet treatment process for MSWI bottom ash (Fe and NFe are ferrous and non-ferrous metals)

Samples from the filter cake (fraction below 0.25 mm) separated in a chamber filter press were also sampled.

The particle density of the mixed material was determined by means of helium pycnometry (AccuPyc, micromeritics) [28]. The moisture content was measured by drying at 105 °C following DIN EN 14346. Loss on ignition was determined by heating the pre-dried sample (12 h at 105 °C) for 4 h at 550 °C [29].

Size-reduced subsamples were acid-digested in a microwave oven (μPREP-A, MLS GmbH, Leutkirch, Germany) according to DIN ISO 11466 [30] to analyze the total contents of metals. Such samples were also subjected to a carbonate determination following the principles of DIN EN ISO 10693 [31] and using a special apparatus (D. Klosa, Hambühren) with pressure sensors to measure the CO₂ development. Chloride and sulfate were analyzed after aqueous extraction with an excess of sodium carbonate.

Leaching Tests

With the test material described in 2.1, four different leaching tests were performed: (i) a lysimeter test up to a liquid-to-solid ratio (L/S) of 2.96 L/kg, (ii) a column test up to L/S = 9.6 L/kg, and (iii) batch tests using a L/S (iii) 2 and (iv) 10 L/kg.

Two laboratory scale lysimeters (polypropylene cylinders, 30 cm in diameter, 57.5 cm in height, ecoTech GmbH, Bonn) were assembled in March 2014 and are still being operated. Approximately 60 kg of test material (59.3 kg dry mass) was introduced to each of the lysimeters in layers. The resulting bulk density of the sample in the lysimeters was 1.46 g/cm³;

the corresponding porosity was 45%; and one pore volume amounts to 18.4 L.

A quartz sand layer of 2 cm (1–2 mm particle size) was put on top of the test material to facilitate the distribution of irrigation done by a system of needles fed from a storage bottle by a peristaltic pump. Artificial rainwater (AR) is used as leachant (pH 6, see Table S1) [32]. The lysimeters were operated under unsaturated condition and irrigated once a week with 816 mL leachant, corresponding to an average annual precipitation rate of 600 mm/a, leading to a liquid-to-solid ratio (L/S) of about 0.7 L/kg per year of operation. The day of the irrigation events each week was chosen randomly within working days. A 0.45 μm polyamide membrane was placed at the bottom of the lysimeters. Leachates were sampled every two weeks. Inserted sensors measured temperature, pH, and redox potential in-line. The duration of the experiment to the time of writing this paper was 37,296 h (more than 4 years); complete analyses of the leachates were performed with 110 different L/S ratios ranging from 0.02 to 2.96.

A tracer was used between L/S 1.9 and 2.7 to check the generation of possible preferential pathways during the lysimeter experiments. For this purpose, KBr substituted KCl in the AR leachant (see Table S1 in the supporting material of this publication). The concentration of Br in the leachant was 1000 mg/L. About 48 L of the leachant with bromide permeated through the lysimeters during this period, whereby about 2.6 pore volumes were intended to be exchanged. This period is called “tracer experiment” in the following.

Laboratory column percolation tests were performed according to DIN 19528 [33]. The inner diameter of the

Borosilicate glass columns was 10 cm, the filling height 40 cm, and the resulting sample dry mass approximately 4.9 kg for all columns. The porosity of the material packed in the columns was 42%. Following DIN 19528, a flow rate of 4.35 mL/min was applied to obtain 5 h contact time between leachant and sample. Whereas such column percolation tests are usually performed up to an L/S of 4 L/kg, the tests in this study were extended to an L/S of approximately 10 L/kg to make it possible to compare the release of substances with the other leaching tests and especially to study the long-term behavior of Sb. In contrast to the lysimeter experiments, saturated conditions prevailed in the laboratory column tests. The duration of the column experiment was 186 h. The use of Milli-Q water as leachant is standard for this type of test. In addition, artificial rainwater (AR) was used to make it possible to compare with the lysimeter results.

Batch tests were performed according to DIN 19529 [34] (L/S 2 L/kg) and DIN EN 12457 [35] (L/S 10 L/kg). A representative sample mass of approximately 2.5 kg for each test was split into several subsamples, which were shaken in several glass bottles for 24 h. After 15 min of settling, the supernatants were recombined. Pressure filtration was subsequently conducted through a 0.45- μ m cellulose nitrate membrane filter for liquid–solid separation. All leaching tests were performed in at least duplicates.

Analysis of Heavy Metals and Anions in Eluates

Leachates were aliquoted after sampling. A small subsample was analyzed immediately for conductivity, pH, turbidity, and total organic carbon (TOC) as basic parameters. For chemical analysis, one aliquot was preserved using concentrated nitric acid [36] for measurement of cations, and one aliquot remained untreated for anion analysis. The eluates were stored at 4 °C until measurement.

TOC concentration was determined with a TOC-VCPH analyzer (Shimadzu, Berlin, Germany) by the difference method with external calibration. pH values were measured with a Schott CG 841 pH-meter equipped with a WTW SenTix 41 pH electrode [37], the electric conductivity with a WTW LF 437 microprocessor conductivity meter [38], and turbidity by a HACH 2100 N turbidity meter [39].

Cations (Al, As, Ba, Ca, Cd, Co, Cr, Cu, Fe, Hg, K, Mg, Mn, Mo, Na, Ni, Pb, Sb, Sn, Sr, V, Zn, Se) were quantified on an iCAP 7000 ICP-OES equipped with an ASX-260 autosampler (Thermo Scientific, Dreieich) according to [40]. An external calibration with mixed standards was carried out for the ICP-OES. Anions (F^- , Cl^- , NO_3^- , Br^- , NO_2^- , PO_4^{3-} , SO_4^{2-}) were determined on an IC320 ion chromatograph with an AS 40 autosampler, an ASRS 300 suppressor, an AS-9-HC separation column with column oven, and a conductivity detector according to DIN EN ISO 10304 [41]. The separation was isocratic with a mixture of carbonate/

bicarbonate in aqueous solution (8 mmol/L/1 mmol/L) as eluent. ICP-MS measurements were performed from tenfold dilutions with a Thermo Scientific iCAP Qc [42].

Results and Discussion

Mass Balance of the Process

The objectives of the wet treatment process for MSWI bottom ash were to produce higher quality mineral material for use as secondary building material and the recovery of the purest possible metals. Therefore, the treatment was executed without prior ageing of the bottom ash. The process was realized in two plants in Germany, one near a waste incineration plant and a second where the ash material was delivered by combined ship and truck transport. The capacity was in the range of 50–60 tons per hour. Details of the process are described elsewhere [21]. A sketch of the process is displayed in Fig. 1.

The mass balance was estimated over an 8-month operation period in which more than 65,000 tons were treated. The results are displayed in Table 1.

The intended use as secondary building material was targeted in a ratio of 60% fine and 40% coarse mineral material by the operator of the recycling plant in order to obtain the desired sieving line and improve the soil-mechanical characteristics. All analyses and leaching experiments in the present study were therefore performed with a 60:40 mixture of the two fractions. However, the mass balance for sulfate was estimated with the analyses of the separate fractions. With the data in Table 1, it can be calculated that 60.4% of the sulfate input is concentrated in the filter cake ($123,500 \text{ mg SO}_4/\text{kg} \times 12.2\%$)/(24,966 mg SO_4/kg), see Table 1. Chloride can be removed from the initial material only by dissolution and not by separation of a certain grain-size fraction and is therefore dependent on the frequency of washing water renewal.

Table 1 Mass balance of the wet treatment process

Fraction	Share (%)	Sulfate (mg/kg)
Filter cake (<0.25 mm)	12.2	123,500
Fine aggregates (0.25–4 mm)	30.7	17,800
Coarse aggregates (4–60 mm)	53.8	6800
Fine non-ferrous metals (0.25–4 mm)	0.03	
Medium non-ferrous metals (4–16 mm)	0.9	
Coarse non-ferrous metals (> 16 mm)	0.7	
Fine ferrous metals (nails, bolts, caps, etc.)	1.7	
Total	100	24,966

Bulk Parameters

For numerous elements, the pH value is the dominant factor controlling the solubility [43]. Wet extracted bottom ash exhibits pH values above 12 before start of the ageing processes (e.g. formation of CaCO_3 from the reaction of Ca(OH)_2 with CO_2 from the air transported by rainwater). From the solubility of Ca(OH)_2 , a maximum pH value of 12.2 can be calculated. At such high pH values, high concentrations of certain heavy metals can be measured in standard leaching tests. Lead, for example, forms soluble hydroxy complexes (Pb(OH)_4^{2-}), resulting in concentrations above 0.2 mg/L in batch tests with $\text{L/S} = 10 \text{ L/kg}$ [44]. Below pH 12, the concentrations are considerably lower. In the present work, the pH value was below 12 in all experiments after reaching equilibrium conditions ($\text{L/S} > 0.21 \text{ L/kg}$), see Fig. 2.

In the batch test at $\text{L/S} = 2 \text{ L/kg}$ and $\text{L/S} = 10 \text{ L/kg}$, only the final pH values can be recorded (10.6 for $\text{L/S} = 2$ and 10.7 for $\text{L/S} = 10$).

Redox potential was measured continuously in the lysimeter experiment and was in the range of 20–40 mV. The electrical conductivity dropped from maximum values above 20,000 $\mu\text{S/cm}$ in the beginning by more than 90% to values well below 2000 $\mu\text{S/cm}$.

Total Content of MSWI BA Constituents

Table 2 shows the total content of substances in the bottom ash samples used for the leaching experiments. The total content of the finest fraction that was removed by the wet-mechanical process step in the BA treatment plant is displayed in Table 2 for comparison. Noticeable is the considerably higher content of Ca and sulfate in the filter cake. Enrichment by other elements such as Pb and Sb might

Table 2 Total content of substances in the test material in mg/kg

Substance	Treated bottom ash 0.25–45 mm	Filter cake < 0.25 mm
Al	36,310.0	35,415.0
As	4.3	7.3
Ba	1,725.6	646.0
Ca	90,678.4	162,725.0
Cd	4.6	8.9
Co	64.1	34.8
Cr	436.9	270.1
Cu	3725.0	2654.0
Fe	57,188.2	28,185.0
K	2414.0	1826.0
Mg	7694.4	8670.0
Mn	1098.2	723.0
Mo	18.4	20.6
Na	6770.8	3054.0
Ni	249.3	147.1
Pb	1062.4	1583.0
Sb	47.9	103.7
Sn	40.1	50.2
Sr	293.0	452.0
V	56.2	58.4
Zn	3413.4	4770.0
Cl	2840.0	6700.0
SO_4	13,400.0	123,500.0
CO_3	33,140.0	

not be significant, due to the common inhomogeneity of MSWI BA. However, at least the wet-mechanical process successfully reduced the sulfate content in the mineral fraction above 0.25 mm foreseen for use (Table 1), since a high portion of sulfate is transferred to the filter cake (Table 2).

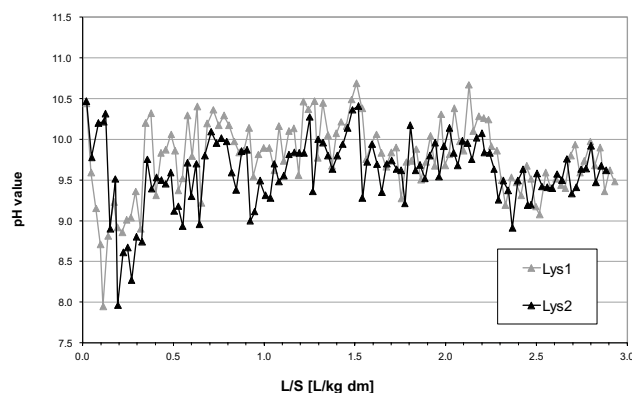
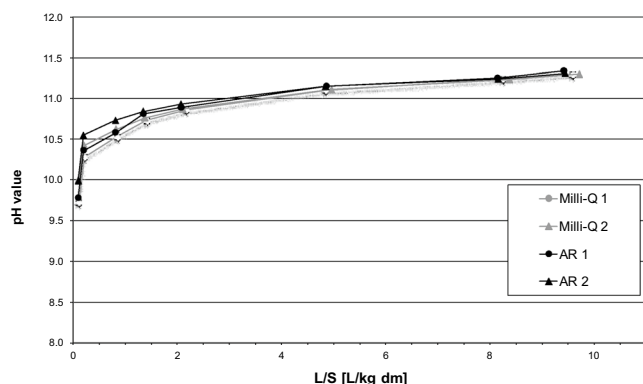


Fig. 2 pH value as a function of the L/S ratio in the column experiments using MilliQ and AR as eluent (left, two replicates each) and the lysimeter experiment (right, two replicates)

Release of Contaminants

The irrigation scheme resulted in unsaturated conditions in the lysimeters. Since the day of weekly irrigation changes randomly, the collected eluate volume in the 2-week sampling periods fluctuated, too. The collected eluate volume was 1568 ± 226 mL on average for both lysimeters in all the experiments, i.e., it took approximately 12 weeks (12 irrigation and 6 sampling events) and an L/S span of about 0.16 to exchange one pore volume (18.4 L).

It turned out during the period of the tracer experiment that the well measurable breakthrough ($c/c_0 > 0.1$) of the bromide tracer took a span of 5 sampling events and 0.13 difference in the L/S.

The bromide concentration then increased to a maximum of about 870 mg/L after an L/S difference of 0.73 or after exchanging about 2.4 pore volumes. The full concentration of bromide in the leachant was not measured in the eluates, probably due to sorption to the sample. After switching back to the original composition of the AR (see Table S1), the bromide concentration decreased again, but was still enhanced (approximately 270 mg/L) after an L/S difference of 0.2 (i.e., definitely more than one pore volume) at the time of writing this manuscript. Overall, the tracer experiment

allows us to assume that no significant preferential pathways existed throughout the lysimeter experiments.

The release of contaminants in the four conducted leaching tests was generally low and above 1% only for the alkaline or alkaline earth elements Na, K, Ca, and Sr, as well as for the anions chloride and sulfate, see Table 3. Although the same initial material was used in all leaching experiments, the release of contaminants differs for some substances considerably, e.g. Al, Cr, Sb, Sn, and Zn. But the variation is not significant (max. value/min. value < 2) for Ca, Cu, K, Na, Sr, and sulfate. All the results from the four leaching tests are displayed in Fig. 3. Plotting all data in a log/log-diagram resulted in a fitted straight line with a slope of 1.09 and an intercept of 2.89 ($y = 0.0013x^{1.09}$, with y = release and x = total content) with a fair correlation of 48%, see the dotted line in Fig. 3. This behavior proves that all that can be predicted from the content is the magnitude of the release of a certain element. Similar findings were obtained in the work of Yin et al. [45]. Their correlation was in the range of 50% with somewhat lower slopes and intercepts. However, the authors performed leaching experiments at a L/S ratio of 10 L/kg only, at natural pH and several other static pH values.

In batch tests, it is not possible to obtain any information on the time-dependent release of a certain element, i.e., the

Table 3 Release of contaminants with incineration bottom ash in four different leaching tests

Bottom ash (0.25–60 mm)		L/S 2 L/kg batch test	L/S 2.96 L/kg lysimeter	L/S 9.6 L/kg column test	L/S 10 L/kg batch test	Min./max.
Content	mg/kg	% release	% release	% release	% release	
Al	36,310.0	0.003	0.004	0.075	0.182	50.7
As	4.3	0.627	0.006	0.233		102.3
Ba	1725.6	0.013	0.009	0.033	0.181	20.5
Ca	90,678.4	1.702	1.467	2.364	2.161	1.5
Cd	4.6		0.002	0.009		3.9
Co	64.1		0.076			
Cr	436.9	0.002	0.010	0.055	0.032	5.8
Cu	3725.0	0.008	0.006	0.009	0.008	1.5
Fe	57,188.2	4.984E–04	9.439E–05	1.399E–05		35.6
K	2414.0	5.074	11.144	7.144	13.422	2.6
Mg	7694.4	0.018	0.003	0.010	0.019	6.0
Mn	1098.2		0.000	0.000		1.8
Mo	18.4	3.824	1.014	4.977		4.9
Na	6770.8	5.338	6.947	4.943	9.216	1.4
Ni	249.3		0.006	0.033		5.3
Pb	1062.4		4.560E–05			
Sb	47.9	0.670	0.033	0.322	0.364	20.2
Sn	40.1	0.419	0.007	0.202		63.0
Sr	293.0	2.474	2.530	3.344	4.355	1.8
V	56.2	0.029	0.211	0.148		7.2
Zn	3413.4	1.172E–04	1.255E–03	7.617E–04	0.006	52.5
Cl	4236.2	53.628	37.315	29.887	46.739	2.8
SO ₄	8415.9	29.498	31.474	46.492	36.359	1.6

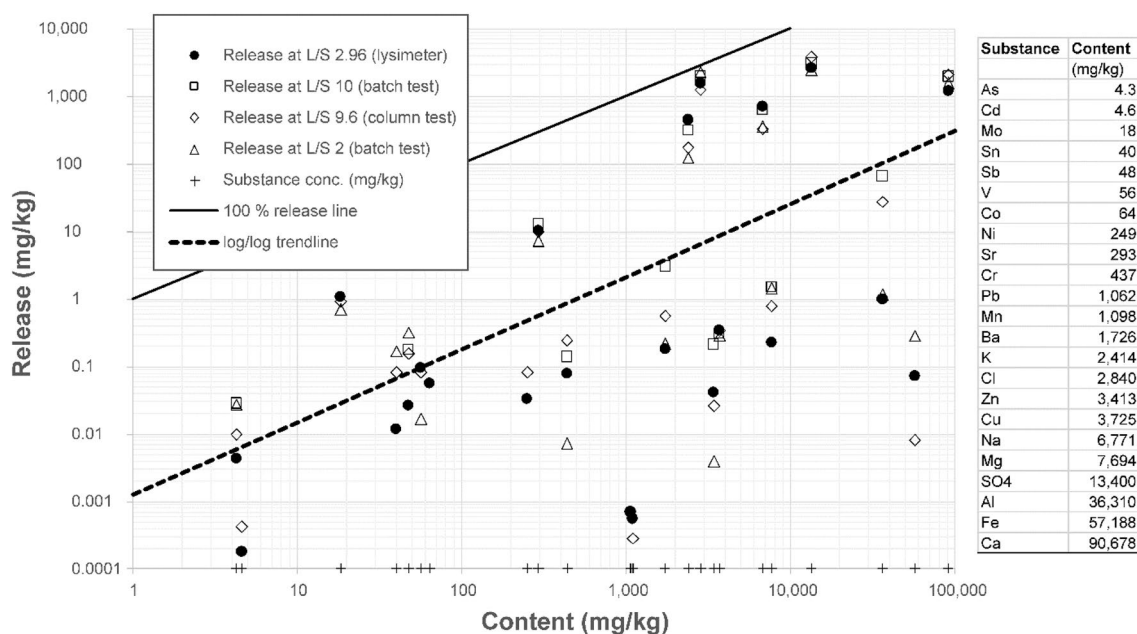


Fig. 3 Release of various cations and anions in four different leaching tests as a function of total content. There is only a fair correlation between total content and release of the respective substances

release at other L/S ratios than that of the designated test. In column test and lysimeter investigations, leachate rates can be obtained at different L/S ratios. Usually it is observed that the highest concentrations are measured in the beginning of the experiment at low L/S ratios. Then the concentrations drop to lower values. This behavior is displayed in Fig. S1 (Supporting Material) for chloride and copper. The cumulative release can be calculated from the individual results by simply adding the values of the fractions. In the case of chloride, the cumulative release curve reaches an almost constant value soon, whereas for other elements the cumulative release increases on a longer time scale because the release does not approach a value close to zero [46, 47]. The concentration curves of chloride, sulfate, Ca, Sr, Ba, Cr, Mo, Cu, Sb, and V for the lysimeter and column experiments, together with the curves of cumulative release, are displayed in the supporting material of this publication (Figs. S2–S5). The concentrations of the illustrated substances in the eluates were all well above the respective LOQ (limits of quantification). It can be seen in the Figs. S2 and S4 for these substances that the overall agreement between the two parallel lysimeter and column tests is quite good.

For some substances which are not illustrated the concentrations were not measurable (Hg, Mn) or very low and partly in the range of the respective LOQ which might have influenced the variance of concentrations. This applies particularly for Cd.

In the lysimeter experiment, a considerably different behavior of antimony and vanadium was observed. The

release of antimony (Sb) was in the range of 2 µg/L for the first 10,000 h of the experiment (L/S = 0.8 L/kg). Then a steep increase to 10 and more µg/L was observed. In the same period, the Ca concentration dropped from values around 2000 mg/L to less than 500 mg/L, see Fig. 4. The reason for this is the proceeding carbonization of the material resulting in the formation of sparingly soluble CaCO₃ from better soluble Ca compounds such as CaCl₂ or Ca(OH)₂.

A correlation of Ca and Sb concentration was first reported for a landfill leachate by Johnson et al. [24]. According to their results, the solubility of heavy metal oxyanions such as Sb(OH)₆[−] could be controlled by Ca metallates. Plotting the measured concentrations of antimony versus the Ca concentration reveals the direct relation, see Fig. 5. Johnson et al. proposed Ca(Sb(OH)₆)₂ as a solubility controlling phase [24] with a Sb/Ca ratio of 2. However, other Sb/Ca ratios, as well, are discussed in the literature [23, 48], i.e., Sb/Ca = 2/1.13. Using the Solver module, the experimental data for Sb and Ca from the lysimeter experiments were entered in a Microsoft Excel table (EA, evolutionary algorithm) according to the following equation describing the solubility of Sb as a function of Ca:

$$K_L = [Ca^{2+}]^m [Sb(OH)_6^-]^2$$

with K_L the solubility product of calcium antimonate.

The best fit resulted in a value of 5.0×10^{-19} for K_L and 1.78 for the exponent m , see Fig. 5, left. With these values,

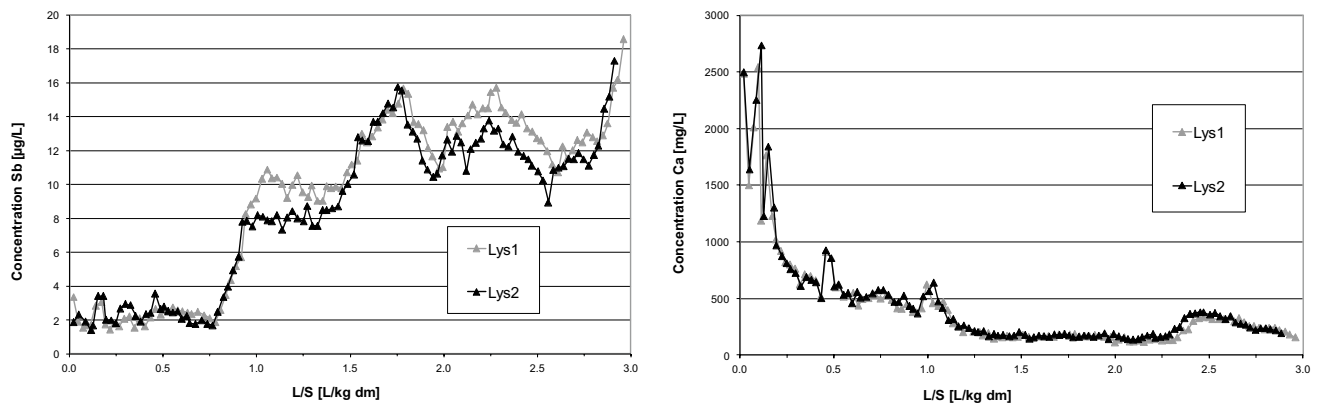


Fig. 4 Concentration of Sb and Ca as a function of the L/S ratio

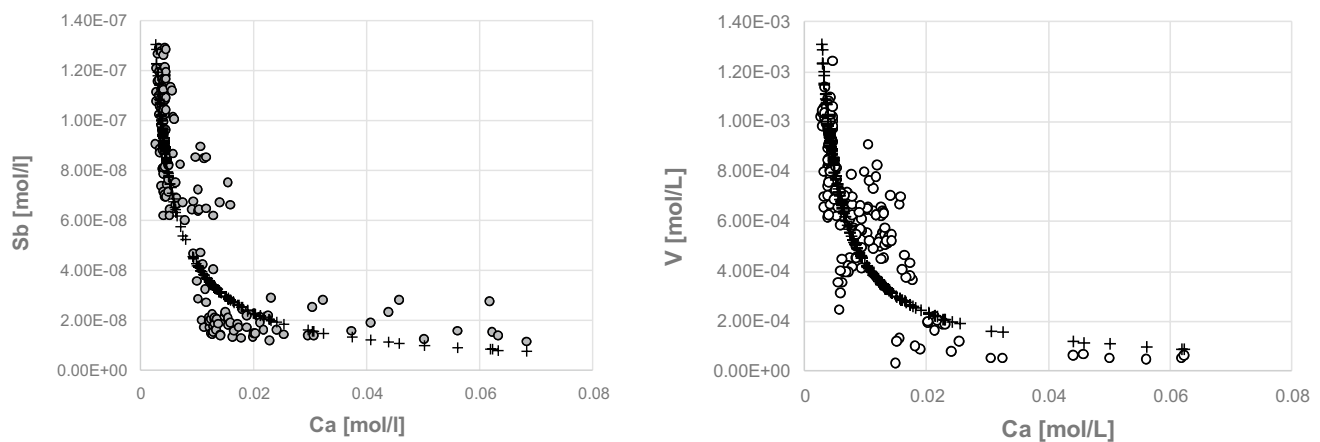


Fig. 5 Concentration of antimony and vanadium in the lysimeter experiment as a function of Ca concentration (circles). The crosses are the result of a fit, see text

the chemical formula to balance positive and negative charges would be $\text{Ca}(\text{Sb}(\text{OH})_6)_2 \times \text{Ca}_{0.78}(\text{OH})_{1.56}$.

Vanadium showed a comparable behavior in the lysimeter experiment, so that CaV_2O_6 could be assumed as a solubility controlling phase [49]. The best fit of the experimental data was achieved with $K_L = 5.5 \times 10^{-11}$ and exponent $m = 1.76$, see Fig. 5, right.

$$K_L = [\text{Ca}^{2+}]^m [\text{V}_2\text{O}_6]$$

The release of antimony and vanadium from bottom ash is clearly related to the concentration of calcium, as can be seen in Fig. 5. This has already been shown for antimony in the work of Johnson et al. [24]. Measured Sb concentrations in landfill leachates (Swiss bottom ash mono-fill) were in the range of 1.1×10^{-7} to 4.7×10^{-7} mol/L. The respective Ca concentrations were in a narrow range between 3.2×10^{-3} and 16.2×10^{-3} mol/L. In the present work, the range of the Ca concentration was wider

(2.8×10^{-3} to 68.3×10^{-3} mol/L) and the Sb concentrations lower (0.1×10^{-7} — 1.3×10^{-7} mol/L). The lower Sb release might be due to the slight reduction of Sb in the tested mineral fraction as a result of enrichment of Sb in the filter cake, see Table 2.

The solubility of antimony could surely also be influenced by other solid phases controlling the solubility of Sb that are involved in the present experiment [26] or by other parameters such as alkalinity, carbonate concentration, or pH. The variation of pH in the lysimeter experiment investigation ranged between 8.5 and 10.5. In this limited pH range no clear correlation of pH and Sb concentration was observable. Influence of pH on the solubility of Sb in bottom ash was discussed in the literature [23]. The data from a field study by Sormunen et al. suggest that high Sb concentrations occur when the amount of seepage water is low and at pH values below 9 [20] (range investigated 8–11.5). However, the authors did not analyze the leachates for Ca.

The German draft ordinance for reuse of mineral waste [8] defines three recovery categories (HMVA-1, HMVA-2, HMVA-3) with decreasing demands on the release of hazardous substances. The limit values for Sb concentration at an L/S of 2 L/kg using column tests or batch tests (DIN 19528 or DIN 19529) are 10, 60 and 150 µg/L respectively. The concentration of Sb in µg/L at L/S 2 L/kg can be retrieved from the value at L/S 2 L/kg on the cumulated release curve in mg/kg and dividing by 2 L/kg. The release at L/S 2 L/kg for Sb in the lysimeter experiments is about 0.015 mg/kg (see Fig. S5 in Supporting Information). Thus, the corresponding concentration can be calculated to 7.5 µg/L, close to the limit value of HMVA-1. The limit value would be exceeded if the concentration at the final L/S of the experiment (2.96 L/kg) was taken for evaluation (release 0.026 mg/kg, resulting concentration 13 µg/L). In case of V the limit value of HMVA-1 is also close to the limit value (limit values: 55, 150 and 200 µg/L; release at L/S 2 L/kg = 0.063 mg/kg, resulting concentration is 31.5 µg/L).

Limit value of HMVA-1 for Sb is exceeded in the standardized column test at an L/S of 2 L/kg (DIN 19528) with a concentration of 13 µg/L for Sb (cumulative release 0.026 mg/kg, see Figure S3 in Supporting Information). With 13 µg/L for V, the concentration is below the limit values.

The results from lysimeter experiment are closer to real field conditions than the column test due to larger sample size and overhead irrigation rather than up-flow conditions. However, for the assessment of the environmental compatibility the column test according to DIN 19528 is legally binding in Germany. Therefore, holding the German limit value of Sb of HMVA-1 might be problematic for MSWI BA, especially due to the inhomogeneous nature of material. In the present investigation the wet treatment of the ash could have an influence on Sb mobilization because the Ca content is lower in the treated ash (high Ca content in the filter cake, see Table 2). As explained above concentration of Ca controls the mobilization of Sb.

Conclusions

The leaching experiments showed that the aim to reduce Cl^- and SO_4^{2-} concentrations in the leachates by the wet treatment technology could be reached [14], in contrast to untreated MSWI BA. More than 60% of the sulfate is accumulated in the filter cake. This is an important effect when considering the suitability of the intended reuse of such material in construction. This finding might be even more important for the treatment of construction and demolition (C&D) waste. Sulfate content in C&D waste is increasing because of the increasing use of building products

that contain sulfate. Wet processing could improve the quality of the secondary building material [50], but there are reservations due to greater effort and, in the end, higher cost.

Antimony is present in residues from municipal solid waste incineration [25]. The investigation of the long-term leaching behavior of Sb was possible only in column and lysimeter experiments and not in batch tests where temporal resolution of the results cannot be obtained. An increase of the Sb concentration was observed in the lysimeter experiment after 10,000 h at an L/S ratio of 0.75 L/kg and in the column experiment at a L/S ratio of 4. The basic characterization of waste materials according to DIN 19528 [33] with L/S ratios of 0.3, 1, 2, and 4 L/kg is therefore appropriate for revealing complex solubility behavior, rather than the simpler compliance test up to an L/S of 2 L/kg only. The leaching behavior of Sb might be critical in view of reuse scenarios with comparably stringent limit values for Sb such as planned in Germany (10 µg/L) [8], due to increasing release of Sb with decreasing concentrations of Ca. The cumulative release of Sb at L/S 2.96 L/kg was 0.026 mg/kg dry matter (dm) and is still increasing. This could be an effect of the removal of the finest fraction by wet treatment which contains approximately 20% of the initial Ca content. The addition of Ca compounds might not solve the problem, because in the longer term any calcium species will be transformed to sparingly soluble limestone anyway. Perhaps additives containing iron, which lead to adsorption of Sb, to iron hydroxide, or to the formation of Fe-antimonates, could be an alternative [26]. However, further research is needed to identify possible countermeasures for avoiding harmful Sb release in a later stage of the lifetime of constructions based on waste materials.

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Compliance with Ethical Standards

Conflict of interest The authors declare that they have no conflict of interest.

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Electronic supplementary material

Waste and Biomass Valorization, Springer

Potential use of incineration bottom ash in construction – Evaluation of the environmental impact

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Table S1: Composition of artificial rainwater (AR)

Substance	Concentration ($\mu\text{mol/L}$)
KCl	40.0
$\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$	50.0
NH_4Cl	39.9
NaNO_3	60.0
$\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$	10.0
HNO_3 (63%)	2.0
pH	5.7

Fig. S1: Concentrations of chloride and calcium in leachate fractions taken from two replicates of laboratory-scaled lysimeter experiments (Lys1, Lys2) at increasing L/S (liquid to solid) ratios

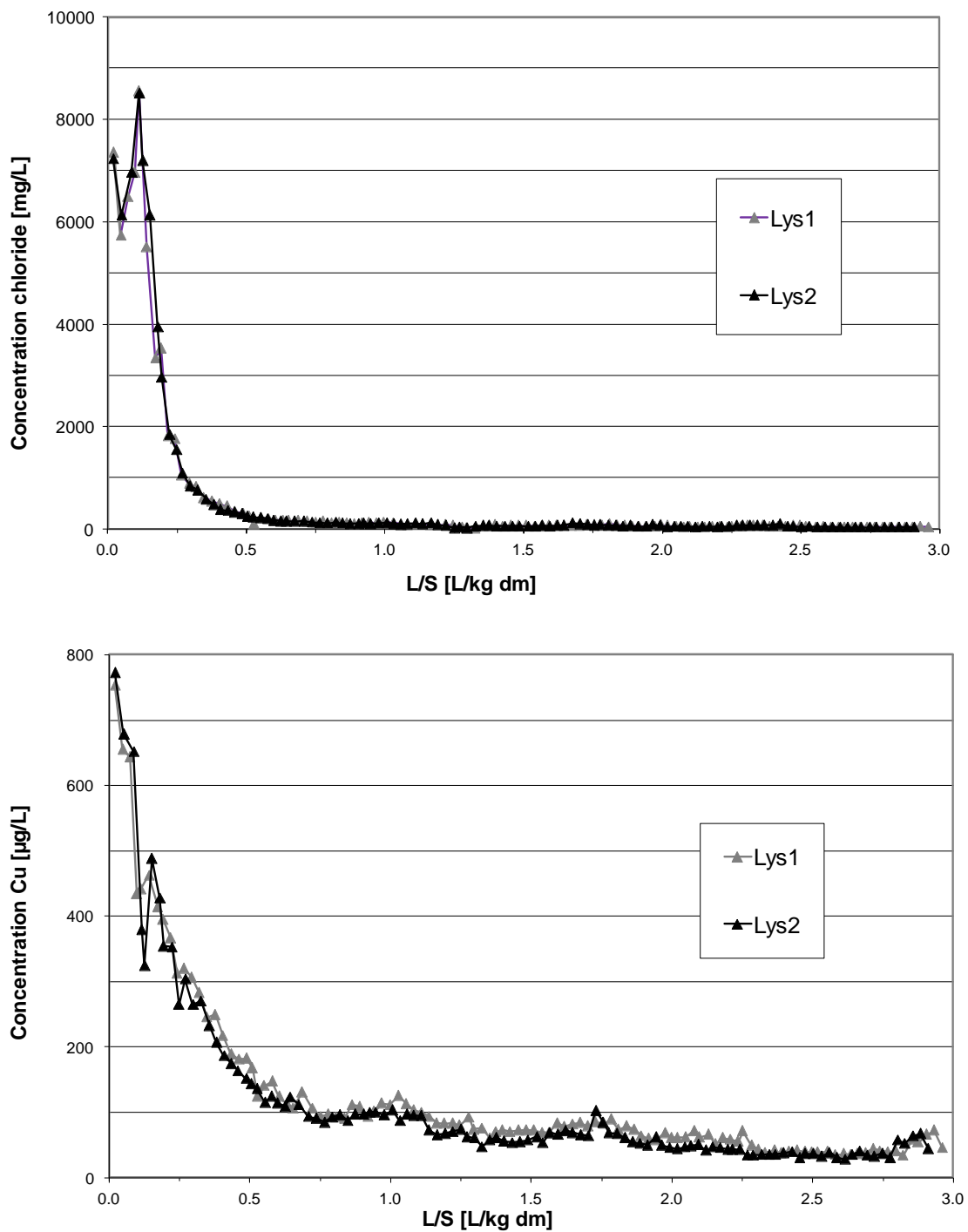


Fig. S2: Concentration of substances in eluate fractions taken from column percolation experiments (two replicates S1, S2) at increasing L/S ratios using demineralized water MilliQ) and artificial rain (AR) as eluent

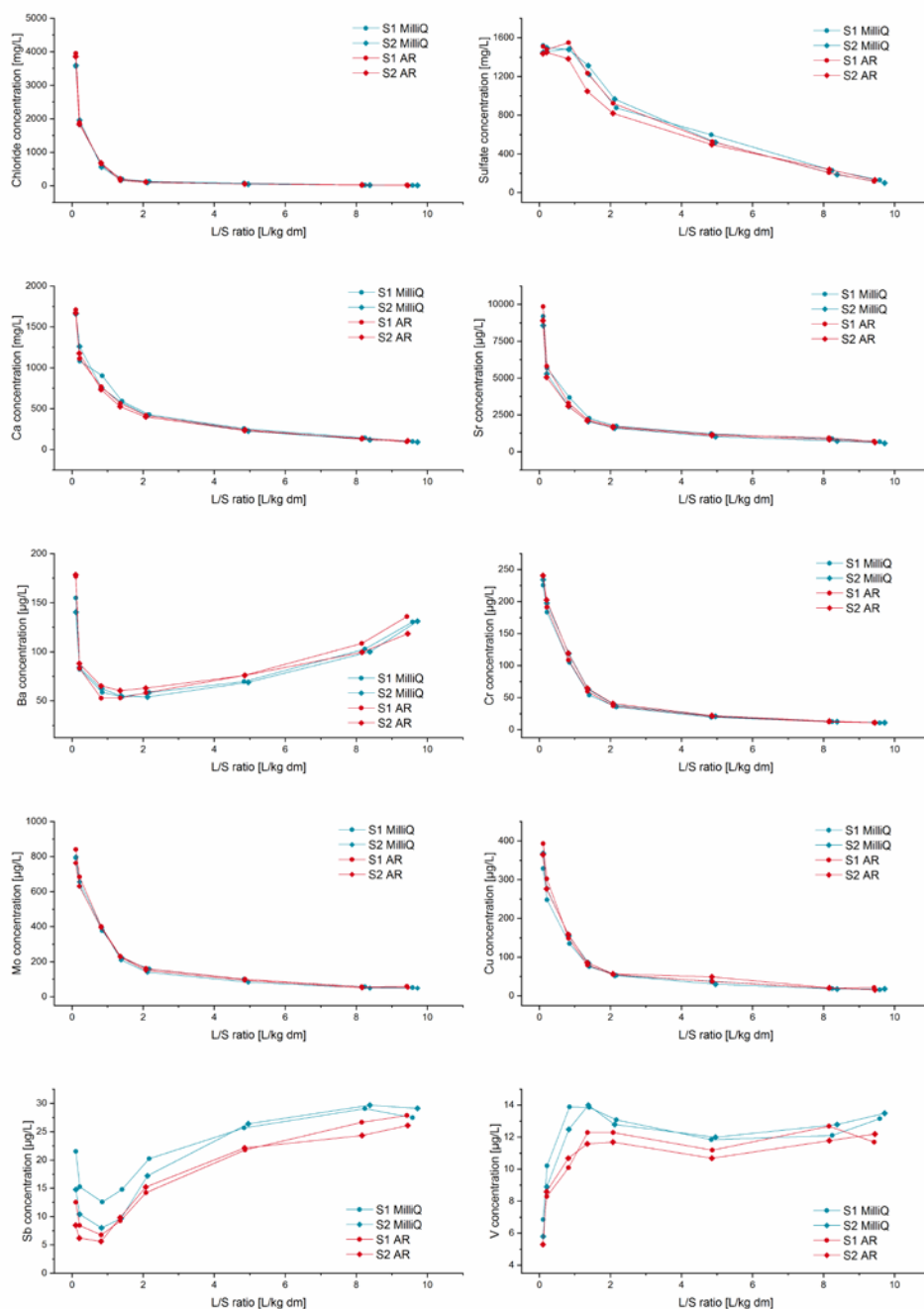


Figure S3: Cumulative release of substances as a function of L/S ratio in column percolation experiments (two replicates S1, S2) using demineralized water (MilliQ) and artificial rain (AR) as eluent

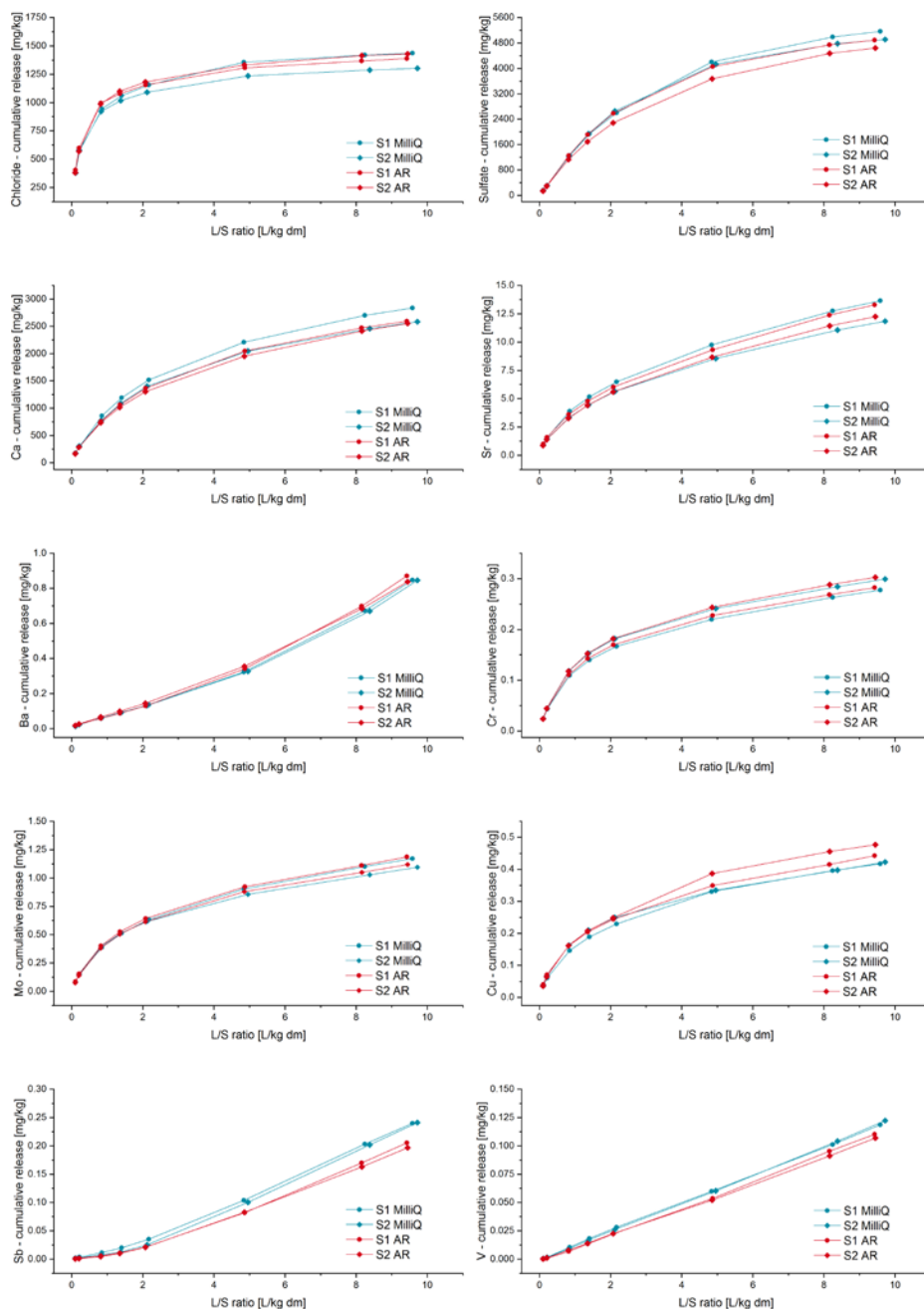


Fig. S4: Concentration of substances in eluate fractions taken at increasing L/S ratios in lysimeter experiments using artificial rain (AR) as eluent (two replicates Lys1, Lys2)

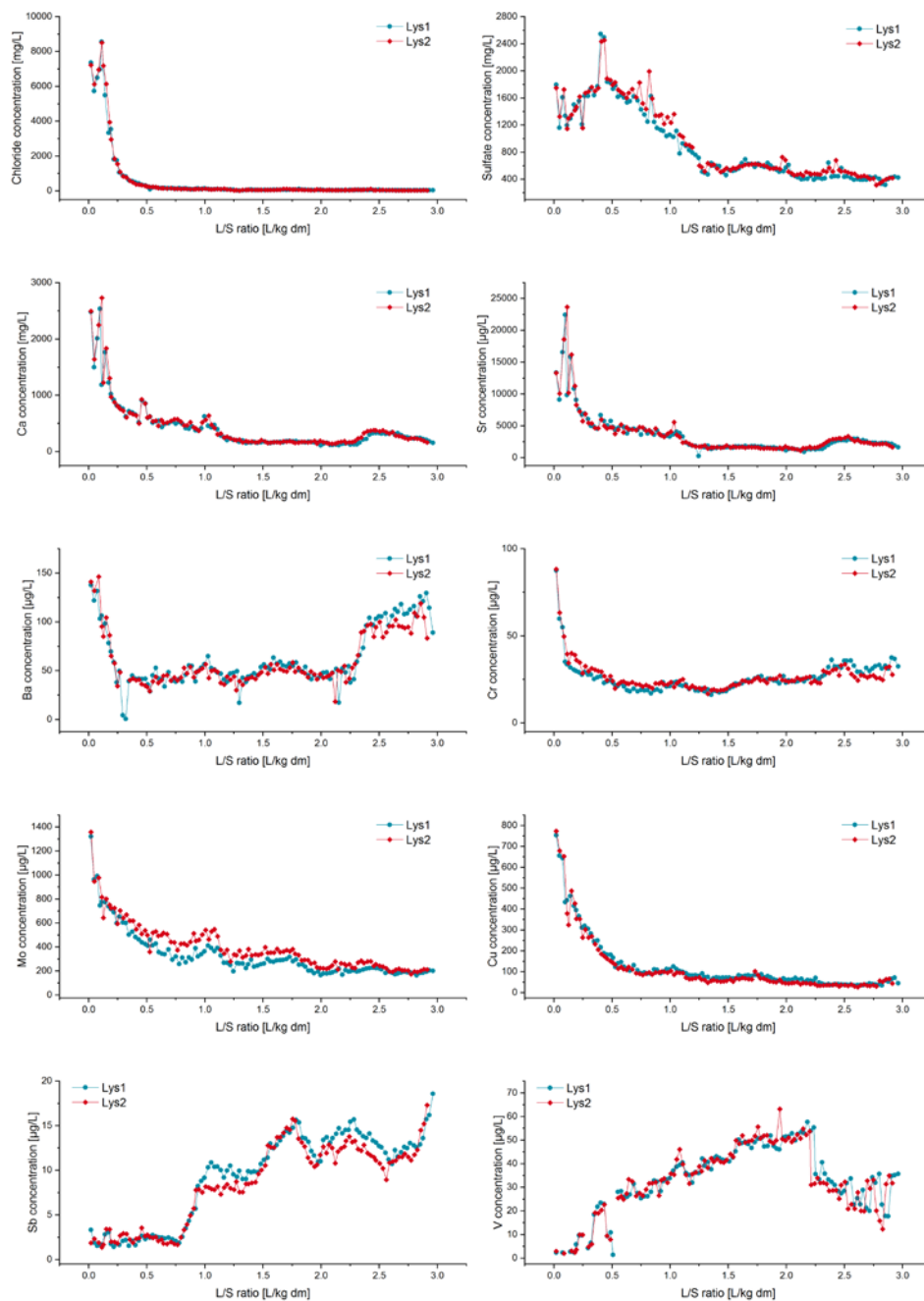


Fig. S5: Cumulative release of substances as a function of L/S ratio in a lysimeter experiments using artificial rain (AR) as eluent (two replicates Lys1, Lys2)

