Treatment of Stormwater Using the Large Particle Size Fraction of Incineration Bottom Ash

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Summary

Adsorbent amended filters to improve treatment of the dissolved fraction of heavy metals from stormwater are currently receiving increased research interest. This paper presents a pilot scale laboratory-based study carried out to investigate the metal removal capacity of municipal solid waste incineration bottom ash particles ≥ 12.5 mm. Five columns with height 40 cm and 10 cm diameter were used to study the adsorption performance of the bottom ash. An artificial stormwater consisting of tapwater added 2 mg/L of Cu, Pb, Zn and Ni was used. Variable hydraulic loads were tested. The result showed promising adsorption abilities of bottom ash ≥ 12.5 mm towards heavy metals. The leaching test showed low leaching potential. To enable industrial use, further research should be carried out in field conditions.

Sammendrag

Rensing av overvann ved bruk av stor størrelsesfraksjon av bunnaske fra forbrenningsanlegg.

Filter tilsatt adsorbenter for å forbedre fjerninga av den oppløste fraksjonen av tungmetaller fra overvann oppnår stadig større interesse hos forskere. Denne artikkelen presenterer et pilotskala laboratoriebasert arbeid for å undersøke kapasiteten som bunnaskepartikler fra avfallsforbrenning ≥ 12,5 mm har til å fjerne tungmetaller. Kolonner med høyde 40 cm og 10 cm diameter ble brukt. Et kunstig overvann, bestående av springvann tilsatt 2 mg/L av Cu, Pb, Zn og Ni, ble benyttet. Forskjellige hydrauliske belastninger ble undersøkt. Resultatet viste lovende adsorpsjonsevne av tungmetaller for bunnaskepartikler ≥ 12,5 mm. Lekkasjetest viste i hovedsak lavere lekkasjeverdier enn det som finnes i nasjonale forskrifter for overvann. Derfor konkluderes det med at lekkasjepotensialet er lavt. For å muliggjøre industriell utnyttelse, burde videre forskning på fullskala utføres.

Introduction

Stormwater treatment systems, such as retention ponds, are proven robust and efficient to remove contaminants in association with particles and suspended solids (Wium-Andersen et al. 2012). However, it is the dissolved pollutants which are the most bioavailable, and they remain in the discharge from the pond. To remove the dissolved fraction, a retention pond can be constructed in combination with a sand filter and a sorption filter, which works as a polishing step (Wium-Andersen et al. 2012). This paper evaluates the treatment function of adsorbent amended filters. Due to high costs of commercial sorbents it is interesting to investigate so-called alternative adsorbent materials. These include waste materials from mining operations, agro-forestry and solid waste incineration (Ilyas & Muthanna 2016).

Bottom ash (BA) is the major by-product from municipal solid waste incineration (MSWI) from waste-to-energy facilities (Dijkstra et al. 2006). MSWI BA has been investigated and proven to be a potential adsorbent towards metals (Shim et al. 2003; Ilyas & Muthanna 2016). Despite its good geotechnical and mineralogical characteristics, MSWI BA has not been fully recycled neither as a construction material nor as an adsorbent due to risks associated with leaching of contaminants (Dijkstra et al. 2006). Ribé et al. (2014) evaluated the content of BA from a Swedish MSWI plant and reported content of metals such as Cr, Cu, Pb and Zn surpassing threshold levels. However, some studies have showed the leaching to be below regulatory limits for hazardous waste (e. g. Phoungthong et al. 2016). Nonetheless, previous studies on MSWI BA show that both leaching and adsorption of heavy metals increase with decreasing particle size, mainly due to increased dissolution and surface area respectively (Shim et al. 2003). However, work done by Ilyas & Muthanna (2017) indicate that this relationship may not be linear. Larger particle size fraction (> 8 mm) showed high adsorption compared to smaller particles (< 2 mm), but without similarly high leaching of metals.

This paper reports on a laboratory-based study further exploring the use of the ≥ 12.5 mm fraction of MSWI BA for removal of dissolved metals. The larger particles do not only have a lower risk of leaching, but they are also more suited for infiltration-based treatment due to high hydraulic conductivity. To address the environmental suitability of using ≥ 12.5 mm fraction MSWI BA, the following research questions were addressed:

1. What is the metal adsorption capacity of the assessed ≥ 12.5 mm fraction of MSWI BA for stormwater treatment?
2. What is the leaching potential of the assessed ≥ 12.5 mm fraction of MSWI BA?

Materials and Methods

Materials

This study used 2 years old BA from a MSWI plant in Trondheim, Norway. The BA was sieved through a 12.5 mm sieve. The particles retained on the sieve were used. Larger rocks (> 18 mm) were removed by hand, to obtain a more uniform particle size. The heavy metals investigated were Zn, Cu, Ni and Pb, as they are the most commonly found and studied metals in stormwater (Genç-Fuhrman et al. 2007). These were purchased from Sigma Aldrich Norway AS as chloride compounds (Cl_2Zn, Cl_2Cu, Cl_2Ni and Cl_2Pb) to spike tapwater to create an artificial stormwater solution.

Batch Adsorption

A batch adsorption experiment was conducted to investigate the theoretical adsorption capacity of the BA. Five metal concentrations were tested, 0.1, 0.5, 1, 1.5 and 2 mg/L, to see if the concentration changed the adsorption capacity. 200 mL of metal solution, which consisted of metal-chloride powders and milli-Q water, was added to bottles containing 20 g BA. A liquid-to-solid (L/S) ratio of 10 L/kg was obtained. The pH of the solutions was adjusted to 7, typical for stormwater, by addition of 0.1 M HNO_3 before added to the bottles (Monrabal-Martinez et al. 2016). The bottles were placed on a shaking table for 24 hours at 130 rounds per minute (rpm).

Column Adsorption

The column experiment was carried out to simulate an in-situ stormwater filter. Five plexiglass columns (height of 40 cm, diameter 10 cm) were filled with 2.5 kg BA to 30 cm height and additional 5 cm of rocks on top. In the bottom of the columns a grid was placed to prevent loss of the filter media. The preparation of the artificial stormwater was done by weighing the amount of the chloride compound corresponding to a concentration of 2 mg/L for each metal. This relatively high concentration was chosen to achieve quick breakthrough and detectable metal concentrations. The metal-chloride powders were dissolved in a beaker before added to a 1 m^3 tank.

Hot water was used to facilitate dissolution of Pb. Nevertheless, problems dissolving all the metal powders were encountered, resulting in lower concentrations than 2 mg/L. Inflow samples were collected to monitor this. Peristaltic pumps supplied the columns with artificial stormwater form the 1 m^3 tank. Parallel column tests were conducted; two columns with constant inflow of artificial stormwater, one control column only fed with tap water and two columns fed with variable inflow of artificial stormwater. Four hydraulic loads, chosen based on the rational formula, were tested: 100, 200, 300 and 400 mL/min. The hydraulic loads chosen corresponded to precipitation intensities for 45 and 180 minutes' duration and 2, 5, 50 and 100 years of return periods in the local IDF data.

Leaching Test

An adsorbent material is considered good when effectively removing heavy metals without releasing potential hazardous substances (Gorme et al. 2015). To evaluate the leaching potential of the investigated BA, a pH-dependence leaching test following European leaching behaviour test (CEN/TS 14429 2005) was performed. CEN/TS 14429 requires material with a grain size where 95 % is smaller than 1 mm. Therefore, the standard was followed as a procedure guideline. Three different pHs were investigated; pH 4, pH 12 and the natural pH of the BA with no acid or base addition. The test was performed in duplicates with a blank test for each pH. The bottles were equilibrated and agitated for 48 h on a shaking table at 130 rpm. Prior to the pH-dependence leaching test, the acid and base neutralization capacity of the BA was analysed. By the end of the test, the volume of acid or base to obtain pH 4 and pH 12 in the bottles was known, and it was later used to conduct the pH-dependence test.

Elemental Composition and Mineralogy

The elemental composition of the BA was measured by X-ray fluorescence spectrometry (XRF, Bruker S8 Tiger 4 kW) both before and after the column experiment. An X-ray diffraction photometer (XRD, Bruker D8 Advance.Diffrac.Suite. Eva) was performed to analyse its mineralogy. For the XRD, only the concentration which was crystalline is reported.

Analytical Methods

The eluates from batch, column and leaching tests were filtered with 0.45 µm filter, acidified with nitric acid for conservation, stored in a fridge and analysed for dissolved metal content by using Inductively Coupled Plasma Mass Spectrometry (ICP-MS). The removal efficiency (%) from both the batch and column test was calculated based on the reduction of metal concentrations in the effluent samples:

$$Removal\ efficiency = \frac{C_i - C_e}{C_i} \qquad \text{Equation 1}$$

where C_i and C_e is the influent and effluent concentration (mg/L). The adsorption capacity from the batch test was evaluated studying Freundlich and Langmuir isotherms, as done in Gorme et al. (2010). To evaluate the long-term adsorption capacity, a breakthrough curve was created form the columns with constant hydraulic load. To conclude on the adsorption capacity a mass balance calculation similar to that found in Paus et al. (2014) was used:

$$q = \frac{\int_0^{V_{eff}} (C_i - C_e)dV}{X} \qquad \text{Equation 2}$$

where q is the adsorption capacity (mg/g), V_{eff} is the cumulative effluent water volume at full metal exhaustion (L) and X is the mass of sorbent (g). Additionally, XRF results and the Thomas model fitted to the effluent metal concentration, through R^2, were investigated for adsorption capacity of BA. The Thomas model is described as follows:

$$\frac{C_e}{C_0} = \frac{1}{1 + e^{[\frac{k_{th}}{Q}(qX - C_0 V_{eff})]}} \qquad \text{Equation 3}$$

where k_{th} is the Thomas rate constant (mL/mg/min), Q is the flow rate through the column (mL/min) and q is the Thomas sorption capacity (mg/g). The lifetime of the filter media was calculated through to the Thomas model, as in

Paus et al. (2014). A design suggestion was presented based on calculations done in Wium-Andersen et al. (2012).

Results and Discussion

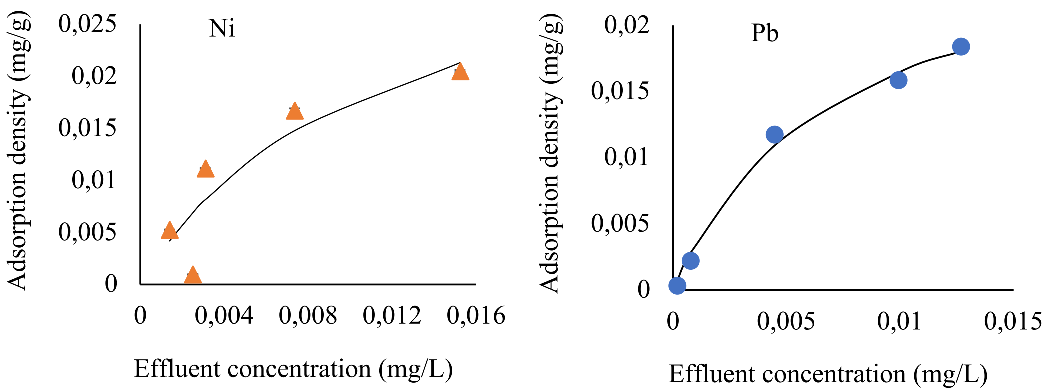
Batch Adsorption Test

Removal rates of 92-100 % were observed for all metals at all concentrations. Isotherm fitting showed that the Langmuir isotherm had the best fit, Fig. 1, which is the same isotherm as Tran et al. (2010) reported for both Pb and Ni adsorption onto chitosan/magnetite composite beads. No isotherm was found to fit the data for Cu and poor fitting was observed for Zn. This is in line with what Wium-Andersen et al. (2012) reported.

The sorption capacities were calculated based on Freundlich isotherm constants presented in Fig. 1. These capacities were compared with other sorbents in Table 1. The representative concentrations are from Lindholm (2004).

Breakthrough Analysis

For the column adsorption experiment the removal rate was close to 80 % for all metals, except Ni, in the first part of the experiment, as seen from the breakthrough curve in Fig. 2. This is in line with what Shim et al. (2003) found when studying adsorption of Cu and Ni onto MSWI BA. The high removal rate can be explained form results from the XRD results. This showed that the material was mostly amorphous in nature, but also indicated the presence of quartz (SiO_2), melilite ($Ca_2MgSi_2O_7$), plagioclase ($NaAlSi_3O_8$ – $CaAl_2Si_2O_8$) and magnetite (Fe_3O_4). Other authors have also reported the presence of quartz, magnetite and plagioclase in MSWI BA (Meima & Comans 1997; Su et al. 2013). Plagioclase and quartz were also reported in natural zeolite which has been proven a good adsorbent towards metal cations in wastewater (Erdem et al. 2004). Hence, the presence of several mineral species could explain the good adsorption of heavy metals.



		Ni	Pb	Zn
Freundlich	K_f (mg/g(L/mg)$^{1/n}$)	0.151	0.135	3.258
	1/n (-)	0.630	0.624	1.006
	R^2	0.778	0.975	0.488
Langmuir	Q_0 (mg/g)	0.036	0.028	3.196
	b (L/mg)	93.706	143.137	0.997
	R^2	0.905	0.993	0.488

Fig. 1 The batch results (symbols) and the isotherm fitted (lines). Ni and Pb fitted to Langmuir. Standard deviations from the three replicates at each concentration are included, partly hidden behind the symbols. Freundlich and Langmuir constants and R^2.

Breakthrough was defined as the point when 95 % or more of the metals was not removed. This was obtained for Ni after 1318 hours (at the end of the experiment), which corresponded to a volume of 2170 L. None of the other metals reached 95 % removal. Nevertheless, Cu reached 94 %, Pb 93 % and Zn 91 %, which is close to breakthrough at the end of the experiment.

Column Adsorption Test

The data fitted the Thomas model well (Table 2), with R^2 in the range 0.82-0.92. The Thomas model

Table 1. Calculated sorption capacities for different sorption materials at equilibrium concentrations of 31, 4.4 and 197 µg/L for Pb, Ni and Zn, respectively, using Freundlich isotherm constants.

	MSWI BA[a] (mg/kg)	Olivine II[b] (mg/kg)	Shell sand[b] (mg/kg)	Coal BA 2-4.75 mm[c] (mg/kg)
Ni	5	78	23	-
Pb	15	345	1632	23
Zn	635	246	485	-

[a] This study.
[b] Wium-Andersen et al. (2012).
[c] Gorme et al. (2010).

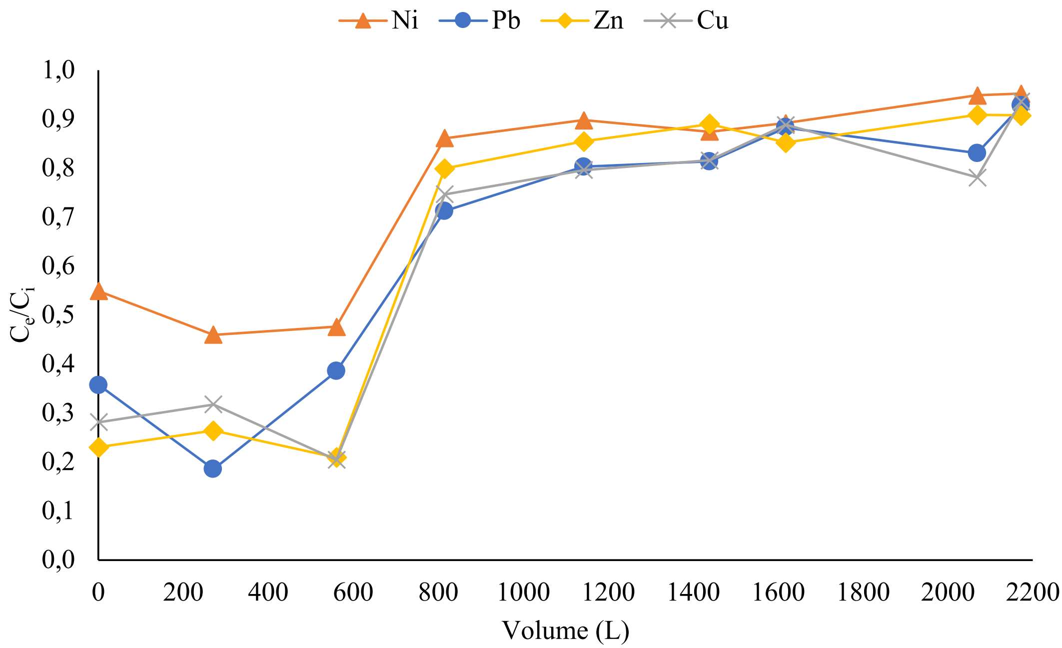


Fig. 2. Breakthrough curve. C_e = effluent concentration. C_i = inflow concentration.

Table 2. Calculated sorption capacities form XRF data, fitted Thomas model and mass balance calculation.

	Ni	Pb	Zn	Cu
XRF (mg/g)	0.219	0.642	0.148	-
Thomas model (mg/g)	0.078	0.285	0.426	0.331
Mass balance (mg/g)	0.302	0.288	0.404	0.304

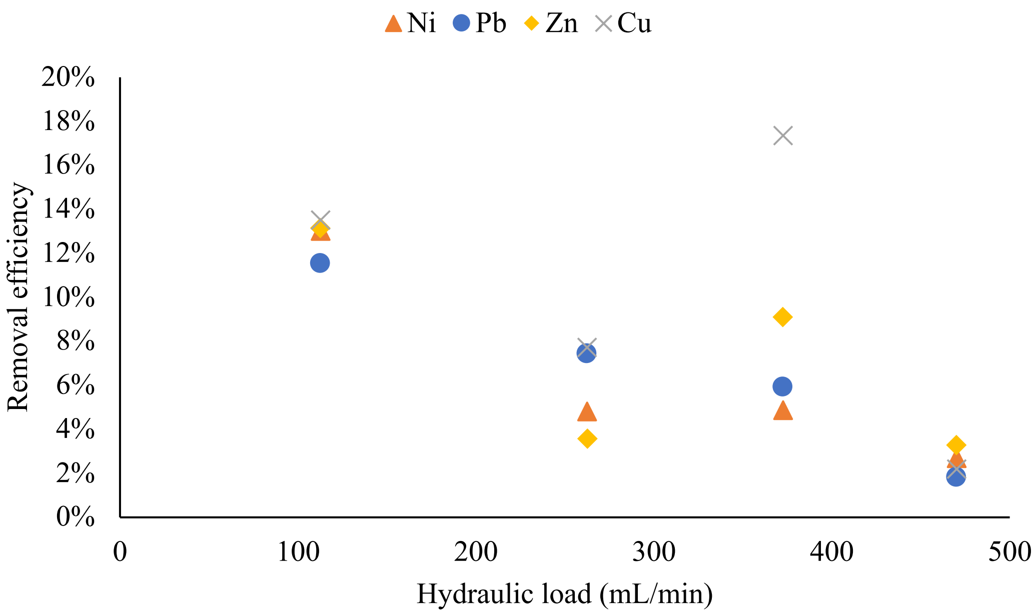


Fig. 3. The removal efficiency (%) of Pb, Ni, Cu and Zn in columns with variable hydraulic loads.

sorption capacity for Zn was in the range of what Paus et al. (2014) reported for sand filter with 30-50 % compost.

As seen from Table 2, the Thomas model and the mass balance calculation were quite similar, especially for Pb, Zn and Cu. Ni had the lowest R^2 when fitting the Thomas model (0.82). This might explain the difference between the Thomas model and the mass balance for Ni. The XRF differs the most from the two others, however this is semi-quantitative data and should be handled with care. The average pH of the inflow solution was 8.28, which was higher than expected. This might have affected the results, since adsorption is pH dependent.

Variable Hydraulic Loads

Higher hydraulic load generally reduced the removal efficiency of the BA, Fig. 3. There is a clear trend towards low removal at high hydraulic loads, which can be explained by reduced contact time. The same trend was observed by Gorme et al. (2015), when investigating heavy metal removal by coal BA. The value of Cu removal efficiency at 370 mL/min can be considered an outlier, possibly due to sampling error.

Between each increase in hydraulic load, the columns were fed with a base flow of 22 mL/min. Removal efficiency rose to a level similar to what was observed before the test between high hydraulic load tests. This indicates a good ability to recover performance after more extreme events. This is important for application. An increase in removal efficiency after periods with no flow through the columns was also observed. This indicates that the filter media has an ability to regenerate its removal efficiency.

Environmental Assessment

Results from the CEN/TS 14429 leaching test is shown in Fig. 4. The presented metals were chosen because they are regulated in national laws. The amount of Hg in the leaching results was found to be under the limit of quantification (LOQ) for the ICP-MS used (< 0.015 µg/L).

From Fig. 4 it can be seen that the leaching was dependent on pH of the lechant. Gianfilippo et al. (2016) reported on leaching from BA from Refuse Derived Fuel Incineration (RFD-I). Their results showed the same tendencies as what is reported in Fig. 4. The leaching potential was at its highest for pH 4, while the

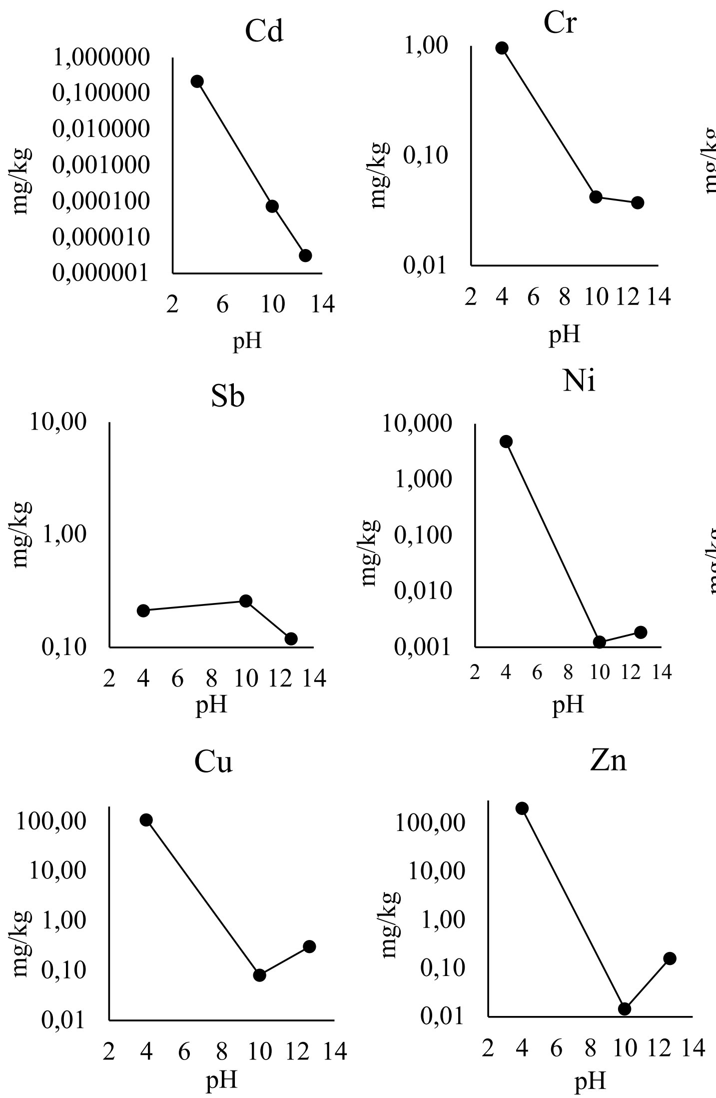


Fig. 4. Results for a selection of metals from the leaching test CEN/TS 14429.

lowest leaching potential was found for the native pH for the MSWI BA (pH ≈ 10). Compared to the XRF results, the amount of the different constituents released at native pH was in all cases less than 1 % of the total content. For Cr, Cu, Ni, Pb and Zn it was less than 0.01 %. Stormwater typically has a pH of 7-8 (Monrabal-Martinez et al. 2016). This indicates that the leaching from the BA will be higher in stormwater than at its natural pH. However, the MSWI BA has shown an acid neutralizing capacity, resulting in high buffer capacity.

When compared to results found for MSWI BA particle size < 4 mm (Ilyas & Muthanna 2017), the leaching was considerably lower for the > 12.5 mm particle fraction. Ranging from 93-31% lower for Cr, Cu, Pb and Zn, with the largest reduction for Cu and smallest for Zn. Comparing the leaching values to Norwegian and Swedish environmental guidelines, as well as values from Åstebøl et al. (2012), they were found to be below the limits, except for Cr. These values are limits for stormwater concentrations in Norway, rather than leaching values

Table 3. Best fit Thomas model sorption capacity and rate constant, predicted volume of stormwater before 40 % breakthrough and predicted service years in experimental columns and field.

	Situation	Sorption capacity (mg/g)	Sorption rate constant (mL/mg/min)	Influent concentration (mg/L)	Volume to 40 % breakthrough (L)	Service years
Pb	Column	0.29	0.03	2	200.7	3
	Field	0.02	0.39	0.031	1098	15
Zn	Column	0.43	0.04	2	379.4	5
	Field	0.64	0.03	0.197	6457.5	87

at L/S 10 L/kg, hence not accounting for the dilution. All the metal concentrations were below Swedish guidelines for stormwater discharge (Regionplane- och trafikkontoret 2009).

Practical Application

Monrabal-Martinez et al. (2017) and Paus et al. (2014) calculated the lifetime of the filter media they investigated in column experiments in two different ways. Paus et al. (2014) used the Thomas model, Equation 3. This same method was chosen in the present study as well. However, a metal reduction of 60 % was investigated, unlike 90 % in Paus et al. (2014), because the removal efficiency will be artificially high with such high inflow concentrations as 2 mg/L in the columns. The Thomas model was adjusted to concentrations found in stormwater in the field (Lindholm 2004). The adsorption capacity at natural stormwater concentrations were calculated from the isotherms fitted from the batch experiment. The lifetime for both column and field situation was estimated, see Table 3. For Ni, a removal efficiency of 60 % was never achieved, see Fig. 2. Therefore, no service years were found for Ni. No isotherm was fitted for Cu in the batch experiment, hence, only results for Pb and Zn are presented.

According to the Thomas model, the service years of the filter will be 15 years if fed with 0.031 mg/L of Pb. It will be 87 years if fed with 0.197 mg/L of Zn. A design suggestion was made for Pb removal based on the approach in Wium-Andersen et al. (2012). A stormwater loading of 16.11 m^3/m^2y was considered, which corresponds to an annual rainfall of 895 mm/y, a runoff coefficient of 0.9 and a ratio between cell surface area and impervious drainage area of 0.05. The concentration of Pb in the stormwater was expected to be 31 µg/L and the sorption capacity of the BA from the batch test at this concentration was found to be 15 mg/kg. Considering a lifetime of 15 years 485 kg BA/m^2 is found to be needed, which results in 48.5 kg BA per length meter for a four-lane highway.

Conclusions

The metal adsorption capacity of the ≥ 12.5 mm fraction of MSWI BA for stormwater treatment was evaluated. According to the batch experiment, the maximum adsorption capacity for Ni and Pb was found to be 36 and 28 mg/kg, respectively, according to the Langmuir isotherm. The sorption capacities of MSWI BA was found to be lower than olivine and shell sand, except for Zn, for representative metal concentrations for stormwater. From the column test the Thomas model gave adsorption capacities for Zn of 0.437 mg/g which is in the same range as reported for sand filters with 30-50 % compost. The removal efficiency was found to decrease with increasing hydraulic load.

The leaching from ≥ 12.5 mm fraction of MSWI BA was found to be lower than limits found in Norwegian and Swedish guidelines. The leaching was found to be pH-dependent with the highest leaching potential at low pH. Due to BA's high buffer capacity, it is not expected to find higher leaching values than the ones found at natural pH. Therefore, it is concluded that the leaching potential of the ≥ 12.5 mm fraction of the BA is not a limiting factor for usage.

However, MSWI BA is a heterogeneous material, and its characteristics will vary at different locations and times. Therefore, more research is needed to reveal additional leaching and adsorption data for MSWI BA.

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