



Norwegian University of  
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# Treatment of Stormwater Using Large Particle Size Fraction of Incineration Bottom Ash

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# Description of Master's Thesis Spring 2017

**Candidate name:** Audhild Bakken Rognstad

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**Title:** Treatment of Stormwater Using Large Particle Size Fraction of Incineration Bottom Ash

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## Background

Bottom ash (BA) is a major byproduct of municipal solid waste incineration (MSWI) from waste-to-energy facilities across EU and Norway. Despite its good geotechnical and mineralogical characteristics, BA has not been fully recycled either as a construction material or as an adsorbent due to risks associated with leaching of contaminants. Previous studies on BA show that both leaching and adsorption increase with decreasing particle size, mainly due to increased dissolution and surface area. However, a recent work done at the Department of Civil and Environmental Engineering laboratory at Norwegian University of Science and Technology (NTNU) shows that this relationship between decreasing particle size and increasing adsorption may not be linear, as larger particle size fraction ( $> 8$  mm) showed very high adsorption compared to smaller particles ( $< 2$  mm) but without high leaching of metals. This presents an opportunity for reusing larger particles in adsorption of contaminants from stormwater. However, more data is required on adsorption capacity, equilibrium isotherms and leaching under various contaminants and hydraulic loads in order to make an expert judgement. Therefore, the present thesis will evaluate adsorption potential of larger particle size of BA in pilot scale column tests.

## Objectives

The main objectives of the project were:

- 1) What is the metal adsorption capacity of the assessed  $\geq 12.5$  mm fraction of MSWI BA for stormwater treatment?
- 2) What is the leaching potential of the assessed  $\geq 12.5$  mm fraction of MSWI BA?

**Collaboration partners:** INEXCES and Klima 2050

**Location:** The master thesis will be conducted at the Department of Civil and Environmental Engineering. The candidate should have regular meetings with advisor(s). The laboratory work will be conducted at the laboratories at the Department of Civil and Environmental Engineering.

**Advisors:** Aamir Ilyas and Tone Merete Muthanna

# Preface

This MSc thesis is the final contribution to the five-year program in Civil and Environmental Engineering at the Norwegian University of Science and Technology (NTNU). The thesis was written during the spring 2017, in the 10<sup>th</sup> and final semester at the Water and Wastewater Division in Department of Civil and Environmental Engineering. The work was carried out January to June 2017 and includes three experiments conducted in the drinking water laboratory and the analytical laboratory at campus Valgrinda at NTNU.

The thesis has an untraditional format with a scientific paper as the main product. This was done in agreement with the supervisor Tone Merete Muthanna. The paper is a manuscript planned to be submitted to the journal VANN. Additional information about the work, and results not included in the paper, is found in Appendix A - E. The work will also be presented at ICUD 2017, International Conference of Urbane Drainage, in September 2017. The Norwegian summary was also submitted to the magazine Byggeindustrien (Construction Industry), as a part of the column Nytt fra NTNU (News from NTNU). It has an expected publication date September 28<sup>th</sup> 2017 in magazine number 15.

The research was funded partly by the INXCES project – INovations for eXtreme Climatic EventS, a JPI Waterworks project, and the SFI project Klima 2050, Centre for Research-based Innovation (klima2050.no).

I would like to thank my supervisor post doctor Aamir Ilyas for all the support and help throughout the whole project and associate professor Tone Merete Muthanna for guidance as my supervisor from NTNU. I would also like to thank Jardar Lohne for guidance on writing a paper and Trine Hårberg Næss and Thuat Trinh in the analytical lab for always giving me a helping hand. A last thank you to my fellow students Even, Lars and Guro who helped me in the laboratory work, and to Markus Glad who has been most helpful with the program Latex and who has proof-read my thesis.

Trondheim, June 9<sup>th</sup>, 2017



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Audhild Bakken Rognstad



# Sammendrag

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

Hvis vi kan rense overvann for tungmetaller ved hjelp av bunnaske fra avfallsforbrenningsanlegg, slår vi to fluer i én smekk: Vannrensing og gjenbruk av avfall. En nylig avlagt masteroppgave ved NTNU viser at dette kan være mulig.



*Figure 1: Overvann forurenses med tungmetaller fra blant annet biltrafikk. Foto: Audhild Bakken Rognstad.*

Tungmetaller er ikke nedbrytbare i naturen og vil dermed kunne bioakkumuleres. De kan derfor gi alvorlige kort- og langtidsvirkninger for både flora og fauna. Tungmetaller kommer fra for eksempel kjøretøy, industri og slitasje på bygninger. I regnvær vil disse havne i overvannet. Filter tilsatt adsorbenter for å forbedre fjerninga av den løste fraksjonen av tungmetaller fra overvann oppnår stadig større interesse hos forskere. Adsorbenten kan være et materiale som vanligvis defineres som et avfallsprodukt. Dette kan for eksempel være sagmugg eller bark fra skogbruk, avfall fra gruveindustrien eller aske fra kullforbrenningsanlegg.

### **Bunnaske med store partikler**

I denne masteroppgava har det gjennom pilot-skala laboratorieforsøk blitt undersøkt kap-

asiteten bunnaske fra avfallsforbrenningsanlegg har til å fjerne tungmetaller fra overvann. Tidligere forskning har vist at mindre partikkelstørrelser av slik bunnaske har god adsorpsjonsevne, men at de har et stort lekkasjepotensiale av uønskede stoffer, som for eksempel tungmetaller. Derfor har vi sett på større partikler av bunnaske – større enn 12,5 mm – som kan ha god adsorpsjonsevne og lavere lekkasjepotensiale.

### **Adsorpsjonstester**

Et batch-adsorpsjonsforsøk ble først gjennomført for å undersøke den teoretiske adsorpsjonskapasiteten til bunnaska. Her ble fem forskjellige konsentrasjoner testa. Resultatene fra forsøkene viste at bunnaska hadde evne til å adsorbere 92-100 % av tungmetallene Cu, Ni, Pb og Zn. Etter at disse lovende resultatene ble funnet, ble det gjennomført et kolonneforsøk. Kolonner med høyde 40 cm og 10 cm diameter ble brukt. Et imitert overvann bestående av springvann tilsatt 2 mg/L Cu, Pb, Zn og Ni ble benyttet og følgende hydrauliske belastninger ble undersøkt; 100 mL/min, 200 mL/min, 300 mL/min og 400 mL/min. Belastningen simulerte større regnværshendelser for å se om den hydrauliske belastningen ville påvirke rensegraden. Resultatet viste at rensegraden går ned ved økt hydraulisk belastning, men at den øker til nivået før belastningen når den hydrauliske belastningen går ned igjen. Generelt ble det observert en lovende adsorpsjonsevne for materialet. Det viste seg at levetida til bunnaska som adsorpsjonsmateriale, kan være 15 år dersom en rensegrad for bly på 60 % er ønskelig.

### **Lekkasjeundersøkelse**

Siden bunnaksa kommer fra forbrent søppel, kan den inneholde stoffer som er farlige for miljøet. En pH-avhengig lekkasjetest ble utført med den europeiske standarden CEN/TS 14429 som retningslinje. Dette for å evaluere lekkasjepotensialet til bunnaska. Tre ulike pH-verdier ble undersøkt; pH 4, askas naturlige pH (~10) og pH 12. Lekkasjetesten viste at bunnaska i hovedsak hadde lavere lekkasjeverdier enn utvalgte grenser for overvann gitt i norske og svenske retningslinjer.

### **Lovende resultater**

Resultatene viser at stor fraksjon av bunnaske fra avfallsforbrenningsanlegg kan benyttes som materiale i sorpsjonsfiltre for å fjerne tungmetaller i overvann. Praksis i dag er at bunnaska legges på deponi. Vi kan altså ha funnet et bruksområde for aska. En utfordring er at bunnaske fra forbrenningsanlegg er et heterogent materiale. Innholdet og dens egenskaper vil kunne variere fra anlegg til anlegg og tidspunkt til tidspunkt. Derfor oppfordres det til mer forskning, for å avdekke ytterligere lekkasje- og adsorpsjonsdata for det lovende filtermaterialet.



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

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Norwegian University of Science and Technology, 2017

## Abstract

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  $\geq 12.5$  mm. Five plexiglas 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 good adsorption abilities of bottom ash  $\geq 12.5$  mm towards heavy metals. A leaching test following CEN/TS 14429 was carried out to evaluate the leaching potential. It showed in general leaching below Norwegian and Swedish limits. Hence, the leaching potential was considered low. To enable industrial use, further research should be carried out in field conditions.

**Keywords:** adsorption, alternative adsorbents, bottom ash, heavy metals, stormwater.

## 1 Introduction

Climate change and urbanisation cause challenges in management of urban drainage. Changing rainfall patterns and more intense rainfall events accentuate these (Hanssen-Bauer et al. 2015, Willems et al. 2012). Increasing urbanisation results in a larger share of impervious surfaces. This leads to higher runoff volume, increased sediment loads and extensive pollutant transport. The concentration of pollutants in the stormwater will also increase, as a result of urbanisation. Dissolved toxic metals in road runoff is of an environmental concern, because metals are not degradable in the environment and may cause short- and long-term adverse effects in the aquatic system (Genç-Fuhrman et al.

2007). To avoid contamination of receiving surface and ground waters, there is a need of stormwater treatment.

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). A certain water volume is usually detained, letting the particles flocculate and sediment. 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 of the contaminants, a retention pond can be constructed in combination with a sand filter and a sorption filter. Then the sorption filter will work as a polishing step for the pre-treated stormwater runoff (Wium-Andersen et al. 2012). In this context, this paper evaluates the treatment function of adsorbent amended filters. Generally the stormwater treatment filters work with sand and other commercial adsorbent materials. However, due to high costs of those adsorbents, these filters can also consist of so-called alternative adsorbent materials. These include waste materials from mining operations, agro-forestry and solid waste incineration used as adsorbents (Ilyas & Muthanna 2016).

Bottom ash (BA) from incineration plants is considered an alternative adsorbent material. BA from coal-burning thermal power plants has been studied to remove dyes (Gupta et al. 2004) and heavy metals (Asokbunyarat et al. 2015, Gorme et al. 2010). It showed promising results. BA is the major by-product from municipal solid waste incineration (MSWI) from waste-to-energy facilities (Dijkstra et al. 2006b). MSWI BA has been investigated as a potential adsorbent towards metals, proving an effective adsorbent (Shim et al. 2003, Ilyas & Muthanna 2016). However, 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. 2006b). 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 that the leaching of various pollutants from MSWI BA can be below environmental limit values (Phoungthong et al. 2016, Ribbing 2007). 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.

Therefore, this paper reports on a laboratory-based study further exploring the use of

a  $\geq 12.5$  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  $\geq 12.5$  mm fraction MSWI BA, the following research questions were addressed:

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

## 2 Materials and Methods

### 2.1 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, but 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 ( $\text{Cl}_2\text{Zn}$ ,  $\text{Cl}_2\text{Cu}$ ,  $\text{Cl}_2\text{Ni}$  and  $\text{Cl}_2\text{Pb}$ ) to spike tapwater to create an artificial stormwater solution.

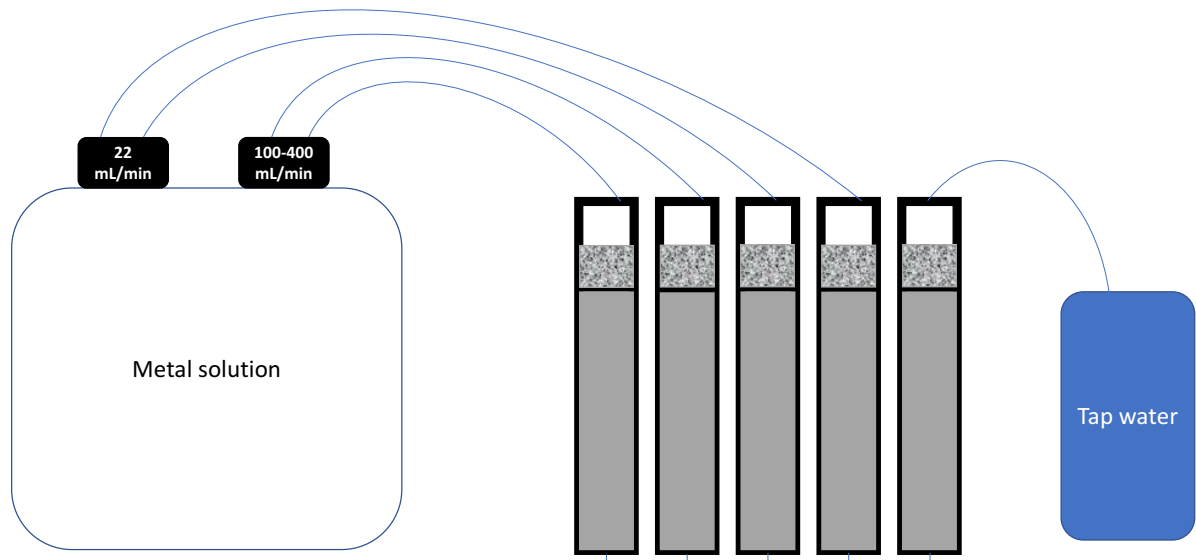
### 2.2 Batch Adsorption Test

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 how the concentration changed the adsorption capacity. A 200 mL 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. Three replicates of each concentration were tested to be able to evaluate the standard deviations and mean value. The pH of the solutions were adjusted to approx. 7 by addition of 0.1 M  $\text{HNO}_3$  before added to the bottles. This was done to simulate the same pH as found in natural stormwater (Monrabal-Martinez et al. 2016). The bottles were placed on a shaking table for 24 hours at 130 rounds per minute (rpm).

### 2.3 Column Adsorption Test

The column experiment was carried out to recreate an in-situ stormwater filter. Five plexiglass columns (height of 40 cm, diameter 10 cm) were filled with approx. 2.5 kg BA to a 30 cm height, with an additional 5 cm layer of rocks on top. The rocks were added

to evenly distribute the water. The rocks and the BA were separated by a geotextile. 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 for this initial feasibility study. The metal-chloride powders were dissolved in a beaker before added to a 1 m<sup>3</sup> tank. Hot water was used to facilitate the dissolution of Pb. Nevertheless, problems in dissolving all the metal powders were still encountered. Therefore, lower concentrations than 2 mg/L were obtained. Inflow samples were collected to monitor this. Peristaltic pumps supplied the columns with artificial stormwater from the 1 m<sup>3</sup> tank.



**Figure 2:** *Experimental setup for column tests.*

Parallel column tests were conducted, see Figure 2; two columns with constant inflow (22 mL/min), two columns fed with variable inflow and one control column was fed with tap water. Four hydraulic loads were applied to evaluate the impact of the adsorption at different flow rates: 100, 200, 300 and 400 mL/min. These were chosen based on the rational formula. The objective was to treat road runoff. Therefore, the runoff coefficient was assumed to be 0.9 as for asphalt (Lindholm 2004). The precipitation intensity was chosen based on local intensity-duration-frequency (IDF) data. 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 IDF data. Durations of 45 and 180 minutes were chosen, since those can be relevant when designing and managing stormwater infrastruc-

ture (Hailegeorgis et al. 2013). The area was assumed to be 1 m<sup>2</sup>. Each hydraulic load was run for a period which resulted in an equal volume of metal solution (288 L) received for each column. The pumps were calibrated and set to deliver a certain hydraulic load. However, they delivered variable flow, so the flow through the columns were measured at each sampling. The measured flows were used for the calculations.

## 2.4 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 was performed. It followed a European standard leaching behaviour test with initial acid/base addition (CEN/TS-14429 2005). 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, see Appendix D. Three different pHs was 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. This was done according to the procedure by CEN/TS 14429. 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 pH-dependence tests, see Appendix D.1.

## 2.5 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. Since the XRF method used was customized to normal geological material in Norway, the data had to be considered semi-quantitative. An X-ray diffraction photometer (XRD, Bruker D8 Advance.Diffrac.Suite.Eva) was performed on the BA to analyse its mineralogy. For the XRD, only the concentration which was crystalline was reported. The BA normally consists of a greater part of amorphous material (Phoungthong et al. 2016). Therefore, the XRD results also had to be considered semi-quantitative in this study.

## 2.6 Analytical Methods

The samples from batch, column and leaching tests were filtered with 0.45  $\mu$ m 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). pH and electrical conductivity were measured for all samples. The removal efficiency (%)

from both the batch and column test was calculated based on the reduction of metal concentrations in the effluent samples:

$$Removal\ efficiency = \frac{C_i - C_e}{C_i} \quad (1)$$

where  $C_i$  is the influent concentration (mg/L) and  $C_e$  is the effluent concentration (mg/L). The adsorption capacity from the batch and column experiment was evaluated studying the Freundlich and Langmuir isotherm as in Gorme et al. (2010). To evaluate the long-term adsorption capacity of the MSWI BA investigated, a breakthrough curve was created from 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:

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

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 (Thomas 1944) fitted to the effluent metal concentration, through  $R^2$ , were also investigated for adsorption capacity of the BA. The Thomas model is described as follows:

$$\frac{C_e}{C_i} = \frac{1}{1 + e^{\frac{k_{th}}{Q}(qX - C_0 V_{eff})}} \quad (3)$$

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 (Paus et al. 2014). A design suggestion was calculated based on a procedure found in Wium-Andersen et al. (2012).

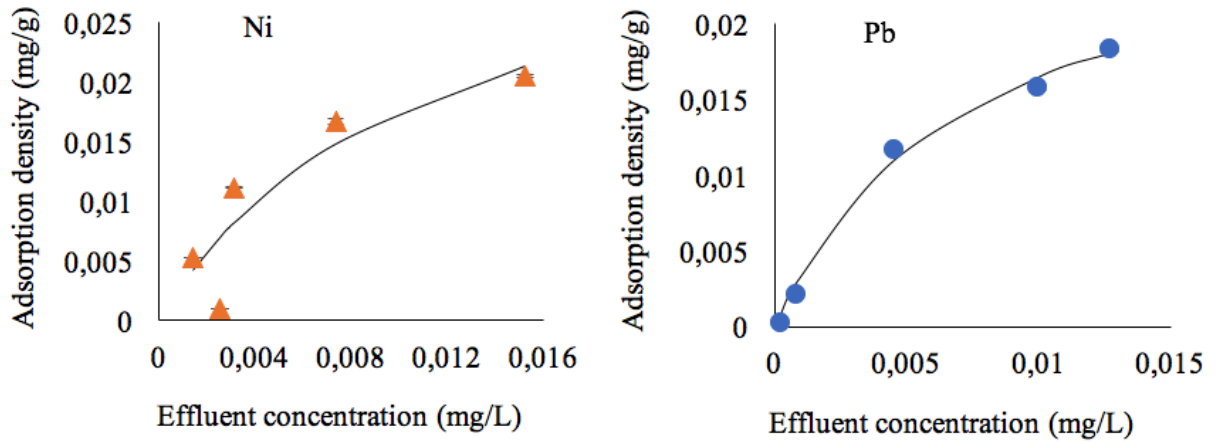
## 3 Results and Discussion

### 3.1 Batch Adsorption Test

Removal rates of the metals were studied through batch adsorption tests, and 92-100 % removal was observed for all the metals at all concentrations, see Appendix A. Isotherm fitting showed that Langmuir had the best fit for Ni and Pb when considering the  $R^2$ , see Figure 3. This result is the same isotherm as Tran et al. (2010) reported for both Ni and Pb 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) observed in their work. They reported that some combinations of sorbate and sorbent had a strong correlation between the water-phase concentration and the mass sorbed by the solid face. Others had a weaker or no correlation. However, they had an L/S ratio of 100 L/kg which might have affected the adsorption mechanisms



differently. Generally, the relative standard deviations (RSD) of the samples from the ICP-MS were low, except for 50 % of the Zn results. This could be a reason for the poor fit of isotherm for Zn.



		Ni	Pb	Zn
Freundlich	$K_f$ (mg/g(L/mg) <sup>1/n</sup> )	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

**Figure 3:** 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  $R^2$ .

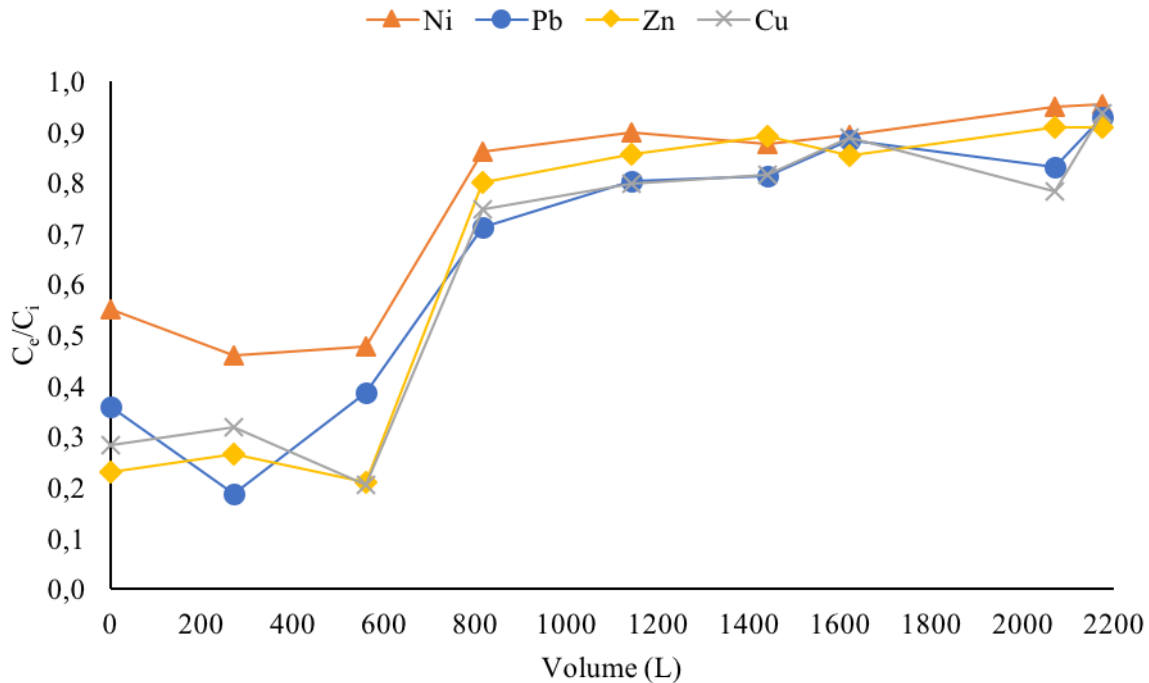
The sorption capacities for representative stormwater metal concentrations were calculated based on the Freundlich isotherm constants presented in Figure 3. These capacities were compared with other sorbents, see Table 1. The representative stormwater concentrations were found in Lindholm (2004).

	MSWI BA <sup>a</sup> (mg/kg)	Olivine II <sup>b</sup> (mg/kg)	Shell sand <sup>b</sup> (mg/kg)	Coal BA 2-4.75 mm <sup>c</sup> (mg/kg)	50 % CVF (MNC1) <sup>d</sup> (mg/kg)
Ni	5	78	23	-	-
Pb	15	345	1632	23	-
Zn	635	246	485	-	260

**Table 1:** Calculated sorption capacities for different sorption materials at equilibrium concentrations of 31, 4.4 and 197  $\mu\text{g/L}$  for Pb, Ni and Zn, respectively, using Freundlich isotherm constants. a: This study. b: Wium-Andersen et al. (2012). c: Gorme et al. (2010). d: Paus et al. (2014).

### 3.2 Breakthrough Analysis

Based on the promising results found in the batch adsorption test, a column adsorption experiment was conducted. As seen from the breakthrough curve in Figure 4, the removal rate was close to 80 % for all the metals in the first part of the experiment, except for Ni. This is in line with what Shim et al. (2003) found when studying adsorption of Cu and Ni onto MSWI BA, where the removal efficiency of Ni was lower than that for Cu. Such high removal rates can be explained from the XRD results. This showed that the material was mostly amorphous in nature, but also indicated the presence of quartz ( $\text{SiO}_2$ ), melilite ( $\text{Ca}_2\text{MgSi}_2\text{O}_7$ ), plagioclase ( $\text{NaAlSi}_3\text{O}_8 - \text{CaAl}_2\text{Si}_2\text{O}_8$ ) and magnetite ( $\text{Fe}_3\text{O}_4$ ). 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 proven to be a good adsorbent towards metal cations in wastewater (Erdem et al. 2004). Magnetite was investigated as an adsorbent towards heavy metals in a composite with chitosan (Tran et al. 2010). The composite proved a good adsorbent towards Pb and Ni. Hence, the presence of several mineral species could explain the good adsorption of heavy metals onto MSWI BA.



**Figure 4:** Breakthrough curve.  $C_e$  = effluent concentration.  $C_i$  = inflow concentration.

After passing a volume of 600 L through the columns the removal rate was remarkably reduced. This was taken as the breakpoint. At this point, the columns had an L/S ratio of 233 L/kg. Breakthrough was defined as the point when 95 % or more of the heavy

metals was not removed. This was obtained for Ni after 1318 hours (at the end of the experiment), which corresponds to a volume of 2170 L, and an L/S ratio of 843 L/kg. None of the other metals reached 95 %. Nevertheless, Cu reached 94 %, Pb 93 % and Zn 91 %, so they were close to breakthrough at the end of the experiment as well. Hatt et al. (2011) investigated breakthrough in filtration systems based on three different soil-based filter medias. They had lower in-concentrations than 2 mg/L and saw breakthrough for Zn, but not for Cu.

The increase in removal efficiency for Pb and Cu at the end of the curve (2060 L) in Figure 4, was most likely caused by a laboratory error. Two hours before the sample was taken at 2060 L, the metal solution tank was found empty. Therefore, the columns had been without flow for an unknown number of hours (< 12 h).

### 3.3 Column Adsorption Test

In the column test the Langmuir isotherm was identified as the best fit for Pb and Cu, with an  $R^2$  of 0.69 and 0.43, respectively. For Zn and Ni, no isotherm was identified. The affinity of the adsorbent for the adsorbate,  $b$  in the Langmuir isotherm, was found highest for Pb, which is the same result as found in the batch test, see Figure 3. This indicated that the highest adsorption affinity was found for Pb.

The XRF results, the Thomas model and the mass balance calculations gave the following adsorption capacities for the four metals investigated, see Table 2. The Thomas model fitted the data well with an  $R^2$  in the range 0.82-0.92, see Table A.7. The Thomas model sorption capacity for Zn was in the range of what Paus et al. (2014) reported for sand filter with 30-50 % compost.

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

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

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 sorption capacity and the mass balance calculation for Ni. The XRF differs the most from the two others.

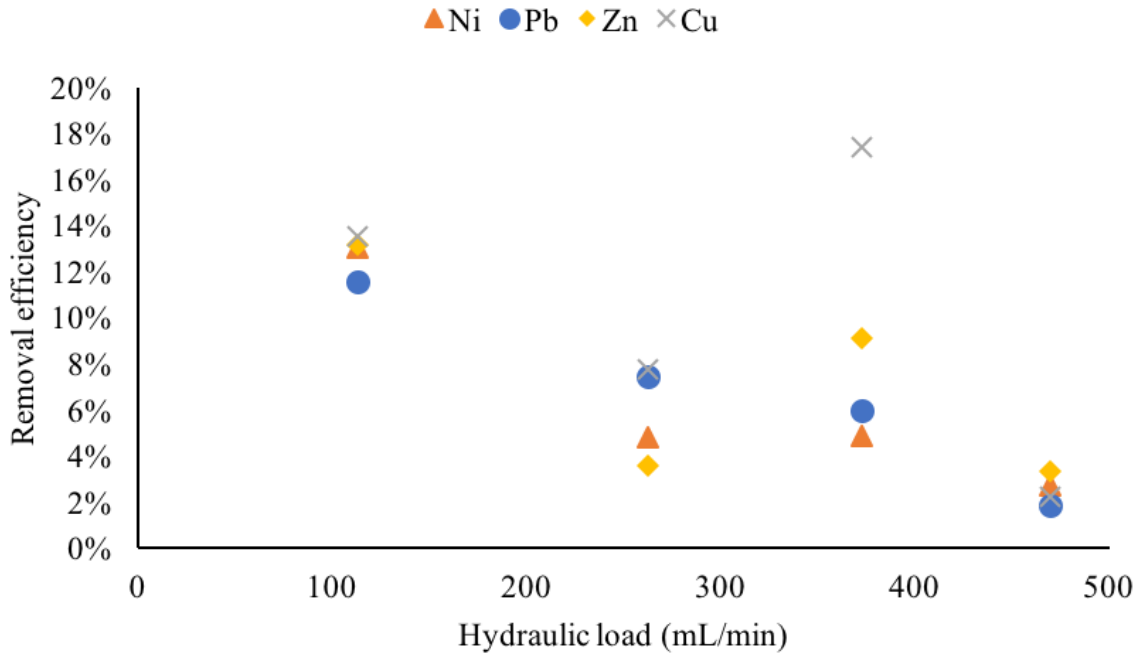
This was probably because the XRF data was semi-quantitative and should be handled with care. The XRF data gave a reduction in Cu content after the adsorption test and is therefore not considered here.

The XRF results from before and after the column adsorption test showed that the content of Pb and Ni was 2-4 times higher after the adsorption experiment than before. This is an indication of the actual adsorption capacity of the BA towards Pb and Ni. The Zn content was found to be almost equal to the sample before the adsorption experiment, and the Cu content was reported to be half of what was found before the adsorption experiment. This can be explained by the fact that MSWI BA is a very heterogeneous material, and the sample taken before the experiment might have higher concentration of Cu. This was supported by the values reported in Su et al. (2013), where the bulk chemical composition of MSWI BA used in their study was presented. This composition corresponded well to the values found in the present study, except that the Cu content was found to be almost 8 times higher in the present study compared to Su et al. (2013).

The average pH of the inflow solution was 8.28, see Appendix B. This is higher than what is expected in stormwater, which has a mean pH of 7.85 in the city of Trondheim (Monrabal-Martinez et al. 2016). This might have affected the results, since higher pH affects dissolution and speciation of metals. A higher pH in the influent solution might have reduced the amount of dissolved metals.

### **3.4 Variable Hydraulic Loads**

From the results, it was seen that a higher hydraulic load generally reduces the removal efficiency of the BA, see Figure 5. The exception was at 370 mL/min for Cu and Zn. However, all the metals had equally poor removal rates at the highest hydraulic load, 470 mL/min. This was an expected result and is in line with what Främ (2002) reported when investigating metal sorption to natural filter substrates. The same trend was observed by Gorme et al. (2015), during investigation of heavy metal removal by coal BA. The reduced removal efficiency at higher flow rates can be explained by the lower contact time obtained between the BA and the metals. This is due to the high hydraulic conductivity of the MSWI BA, which was found to be 2292 cm/h, see Appendix C. The value of Cu removal efficiency at 370 mL/min can be considered an outlier, possibly due to sampling error.

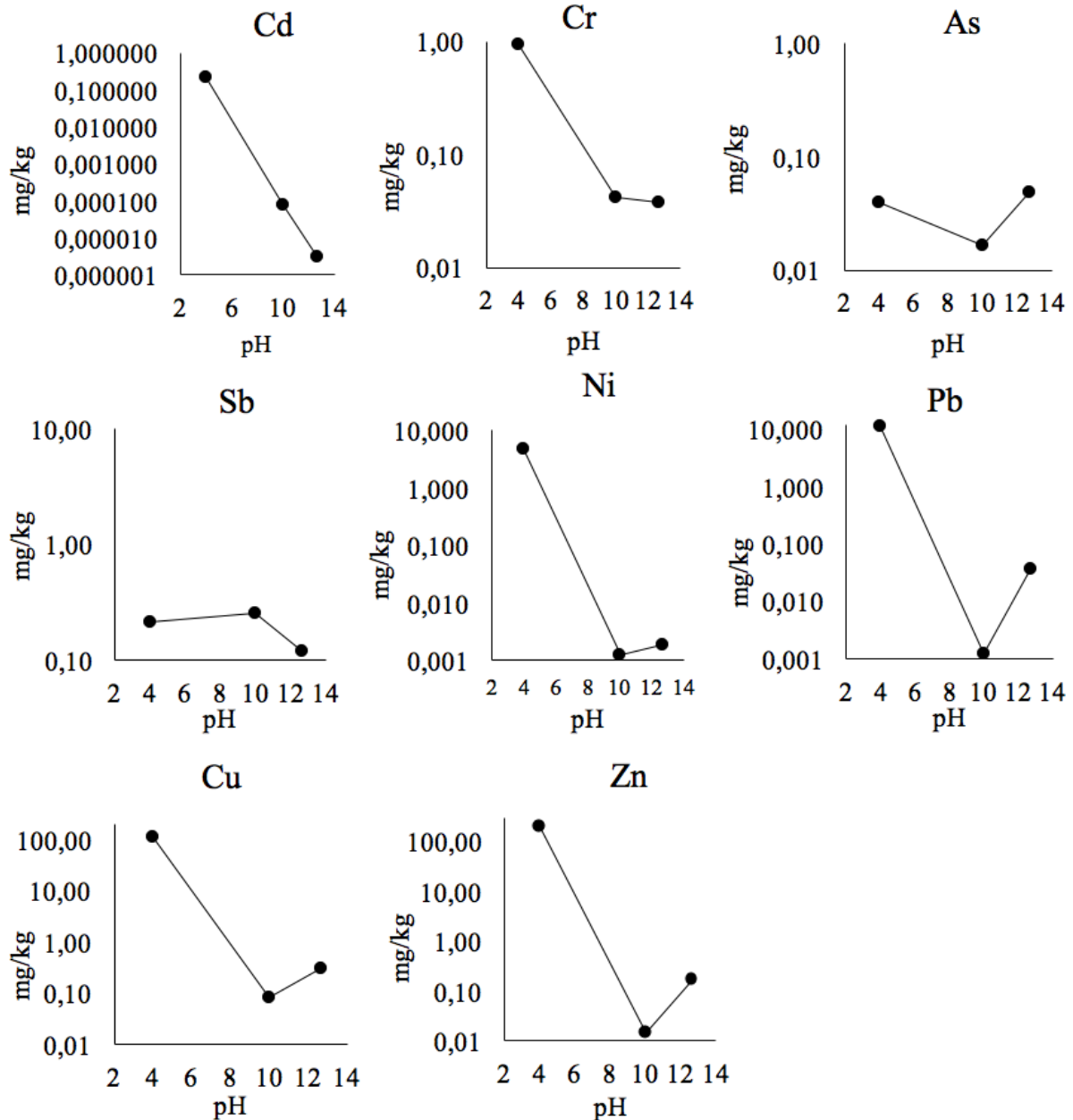


**Figure 5:** The removal efficiency (%) of Pb, Ni, Cu and Zn in columns with variable hydraulic loads.

The tests were conducted to investigate the maximum hydraulic load that the filter can handle without losing desired performance. At 470 mL/min the filter seemed to reach breakthrough ( $< 5\%$  removal) for all four metals. Therefore, it was concluded that the filter is ineffective at loads similar to or higher than 470 mL/min. However, it was observed that between each high hydraulic load test, when the columns were fed with a base flow of 22 mL/min, the removal efficiencies rose to a level similar to what was observed before the test. At this time of the experiment these efficiencies were in the range of 17-36 % for all metals except Ni, which generally had a lower removal rate. This indicates that the removal efficiency during high loads was reduced, but the material was able to recover its removal efficiency when returned to base flow. This is important in an application perspective. 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 of regenerate its removal efficiency. No pounding was observed during any of the four tests, which was expected based on the high hydraulic conductivity found for the BA. This indicates that below the 470 mL/min the BA filter will not experience any pounding. If the storm intensity of the load is calculated as in Monrabal-Martinez et al. (2017), 470 mL/min corresponds to a 10 minutes duration rainfall event with a 50 years return period in Trondheim, Norway. This indicates that the filter would be robust in face of the intensifying rain storms due to climate change.

### 3.5 Environmental Assessment

Results from the CEN/TS 14429 leaching test are shown in Figure 6. The presented metals were chosen because they are regulated by 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 ( $< 0.015 \mu\text{g/L}$ ).



**Figure 6:** Results for a selection of metals from the leaching test CEN/TS 14429.

From Figure 6 it can be seen that the leaching is dependent on pH of the leachant. 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 Figure 6. The leaching potential was at its highest for pH 4, while the lowest leaching potential was

found for the native pH for the MSWI BA (pH  $\sim$ 10). We see a slight increase in leaching potential at pH 12 for the majority of the metals. The exception is Sb, which has its highest leaching potential at natural pH. 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 %.

When compared to results found for MSWI BA particle size  $< 4$  mm (Ilyas & Muthanna 2017), the leaching was considerably lower for the  $\geq 12.5$  mm particle fraction. For example, leaching of Cr, Cu, Pb and Zn was found to be 79 %, 93 %, 59 % and 31 % lower for MSWI BA  $\geq 12.5$  mm than what was reported for MSWI BA  $< 4$  mm, respectively.

To evaluate the environmental suitability of MSWI BA as a filter material, its leaching values at its natural pH were compared to certain limits found in Norwegian and Swedish environmental guidelines. The leaching values found in the present study were all below the limits examined. The only exception was Cr compared to the values in Åstebøl et al. (2012). These values are limits for the concentration in stormwater in Norway, not the leaching values at L/S 10 L/kg. Therefore, it is expected that these values will be lower than what was found through CEN/TS 14429, because the leachate will be diluted into the stormwater. Thus, the slightly higher values for Cr found through CEN/TS 14429 in the present study is not considered an environmental concern. However, the recipient has to be considered in each case. All the metal concentrations were below Swedish guidelines for stormwater discharge (Regionplane och Trafikkontoret (Swedish Regional Planning and Traffic Office) 2009), which is considered the most relevant regulation for leaching from BA in a stormwater treatment system. Because the leaching values were below leaching limits from construction materials (Naturvårdsverket (Swedish Environmental Protection Agency) 2010), it can be argued that the BA can be used as such. This could for example be as ballast stone in road construction. Hence, it might have been found a new way of reusing the MSWI BA, and the amount sent to landfill can be reduced.

### 3.6 Practical Application

Both 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. Monrabal-Martinez et al. (2017) estimated the lifetime by dividing the expected cumulative pollutant retention at a desired metal removal by the pollutant load expected to be treated by the filter. Paus et al. (2014) used the Thomas model, Equation (3). This method was chosen to calculate the filter lifetime in the present study as well, because this model is supported by extensive experimental. See Appendix E.2 for lifetime calculation procedure.

A metal reduction of 60 % was investigated, unlike 90 % in Paus et al. (2014), since this was the highest removal rate obtained with the fitted Thomas model, see Figure A.3. Since the removal efficiency will be unnaturally high with a metal concentration of 2 mg/L in the columns, the Thomas model was adjusted to representative stormwater metal concentrations found in Lindholm (2004). The adsorption capacity at representative stormwater concentrations was calculated from the isotherms fitted the results from the batch experiment, see Appendix E.1. The lifetime for both column and field situation was estimated, see Table 3. For Ni, a removal efficiency of 60 % was never achieved, see Figure 4. 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 represented.

	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	379.5	5
	Field	0.64	0.03	0.197	6457.5	87

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

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. This is more years than what Paus et al. (2014) reported for Zn onto sand and compost media. However, the Zn batch results fitted to Freundlich had an  $R^2$  of only 0.49 where the Pb data had 0.98. Hence, the Pb results are more reliable than the Zn results. However, these service years estimated must be handled with caution, because the experiments were conducted under controlled laboratory conditions, which differ from real field conditions. Therefore, the values presented can be seen as a general indication of potential service years.

A design suggestion was made for Pb removal based on the approach in Wium-Andersen et al. (2012). A stormwater loading of  $16.11 \text{ m}^3/\text{m}^2\text{y}$  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  $31 \mu\text{g}/\text{L}$  and the sorption capacity of the BA from the batch test at this concentration was found to be 15 mg/kg. If we consider a lifetime of 15 years, a requirement of  $485 \text{ kg BA}/\text{m}^2$  was found. In the perspective of a four-lane



highway, if two lanes are drained to one filter, this corresponds to 48.5 kg BA per length meter of road.

From Figure 6, it can be seen that the leaching potential for the majority of the metals will increase with decreasing pH. The natural pH of MSWI BA was found to be  $\sim 10$ , which indicates that it has an alkaline character. 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, see Appendix D.1. Therefore, it is argued that the BA itself will work as a buffer and avoid increase in leaching when exposed to stormwater.

Because a reduction in removal efficiency with increasing hydraulic load was observed, it seems important to regulate the water inflow to the filter to increase the hydraulic contact. This can be done through implementing a treatment system having a retention pond with a weir before the sorption filter controlling the flow into it. Vollertsen et al. (2009) successfully tested a system with a retention pond, followed by a sand filter and a sorption filter as a polishing step at the end. Such a system could be tested with MSWI BA as the media in the sorption filter. Then the particle bound pollution will settle in the retention pond and the dissolved metal fraction will be adsorbed onto the MSWI BA. Gorme et al. (2015) did such a test for coal BA with promising results.

## 4 Conclusions

The metal adsorption capacity of the  $\geq 12.5$  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. Compared to other materials and with representative metal concentrations found in stormwater, the sorption capacities of MSWI BA was found to be lower than olivine and shell sand, except for Zn adsorption. 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. However, the efficiency quickly climbed back up to pre-high hydraulic load, when the system was stress tested.

The leaching from  $\geq 12.5$  mm fraction of MSWI BA was found to be lower than limits found in Norwegian and Swedish guidelines. Compared to XRF data the leaching was  $< 1$  % for all metals investigated. The leaching was found to be pH-dependent with the highest leaching potential at low pH. Because of the BA's high buffer capacity, it is not

expected to find higher leaching values in stormwater than the ones found at natural pH for the BA. Therefore, it is concluded that the leaching potential of the  $\geq 12.5$  mm fraction of the BA is not a limiting factor for the usage of MSWI BA as an adsorption media in stormwater treatment. Though, the recipient has to be considered.

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

## 5 Acknowledgements

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*The following reference list includes both references found in the paper and in the appendices.*

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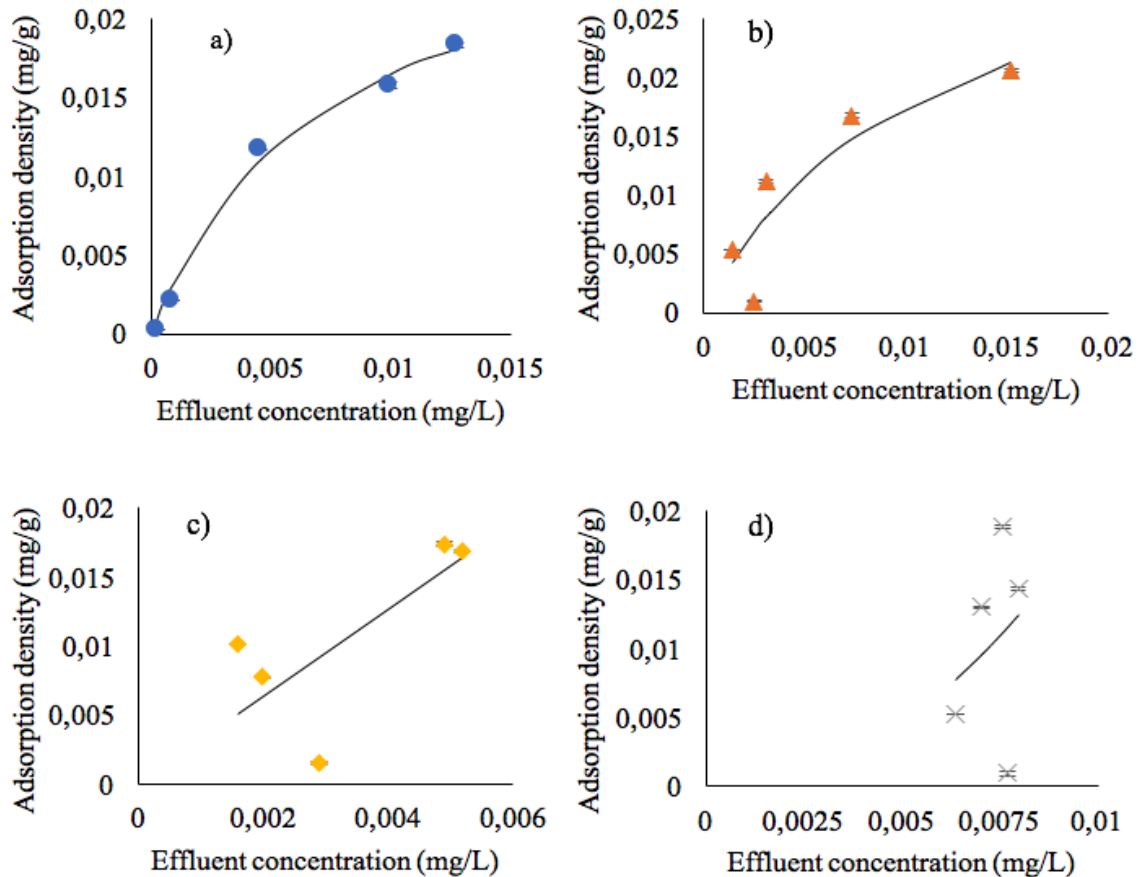
# Appendices

## A Batch Adsorption Results

Table A.1, Figure A.1 and Table A.2 present the batch adsorption results for all of the metals investigated.

Metal concentration (mg/L)	Ni (%)	Pb (%)	Zn (%)	Cu (%)
2	99	99	100	100
1.5	100	99	100	99
1	100	100	100	99
0.5	100	100	100	99
0.1	97	99	98	92

**Table A.1:** Removal rate (%) for Ni, Pb, Zn and Cu found in the batch adsorption test.



**Figure A.1:** The batch results (symbols) and fitted isotherms (lines). Standard deviations from the three replicates at each concentration are included, partly hidden behind the symbols. a) Pb fitted to Langmuir. b) Ni fitted to Langmuir. c) Zn fitted to Freundlich. d) Cu fitted to Freundlich.

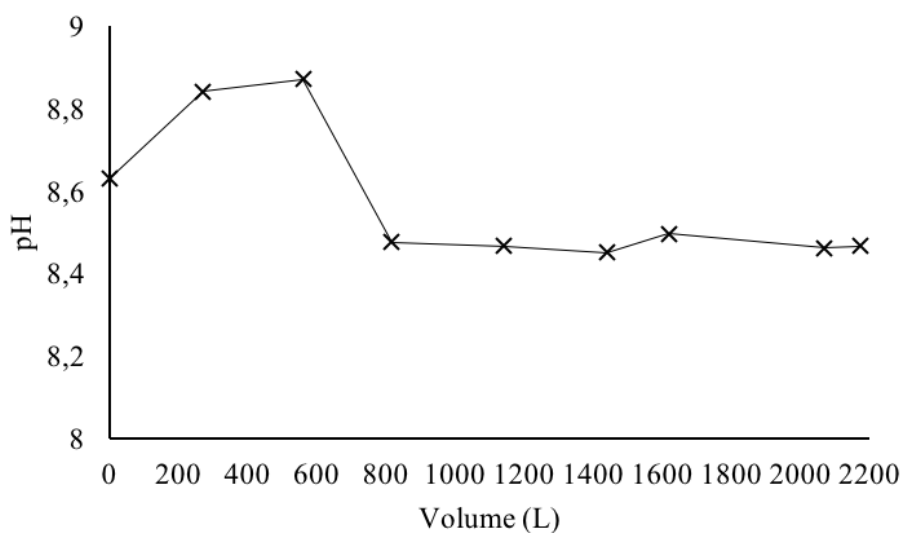
		Ni	Pb	Zn	Cu
Freundlich	$K_f$ (mg/g(L/mg) <sup>1/n</sup> )	0.151	0.135	3.258	15.419
	$1/n$ (-)	0.623	0.624	1.006	2.094
	$R^2$	0.778	0.975	0.488	0.073
Langmuir	$Q_0$ (mg/g)	0.036	0.028	3.196	1.251
	$b$ (L/mg)	93.706	143.137	0.997	1.159
	$R^2$	0.905	0.993	0.488	0.054

**Table A.2:** Constants from best fit of Freundlich and Langmuir isotherms and  $R^2$ .



## B pH Measurements During Column Test

The pH was measured throughout the whole column test. It is seen from Figure A.2 that the pH behaved as expected compared to the removal efficiency in the columns (Shim et al. 2003). It increased with increasing removal efficiency, and decreased throughout the experiment as the columns approached breakthrough. This can be explained by the point of zero charge (PZC) of the minerals in BA (i.e. iron oxides) which is estimated to be around 7-8.00 (Kosmulski 2009). This means BA particles have negative charge that promotes adsorption of metal cations. Moreover, high pH in combination with high concentrations can also lead to precipitation of metals. However, when the pH of the metal solution is below the PZC of the BA, the BA particles are positively charged, which hinders adsorption of metal ions.



*Figure A.2: The pH measurements during the column test.*

## C Hydraulic Conductivity Test

A saturated hydraulic conductivity test was performed using a version of the constant head method described in ASTM-F1815-11 (2011). The flow through the column was measured three times, and a mean  $Q$  value was calculated. The saturated hydraulic conductivity,  $K_{sat}$  was calculated according to Equation 4:

$$K_{sat} = \frac{QL}{hAt} \quad (4)$$

where  $K_{sat}$  is the saturated hydraulic conductivity (cm/h),  $Q$  is the quantity of effluent collected in a period of time ( $\text{cm}^3$ ),  $L$  is the length of soil column (cm),  $h$  is the hydraulic head,  $A$  is the sectional area of the soil core ( $\text{cm}^2$ ) and  $t$  is the time required to collect  $Q$  (h).

Mean Q ( $\text{cm}^3$ )	L (cm)	h (cm)	A ( $\text{cm}^2$ )	t (s)
500	30	30	$\pi \times 5^2$	10

**Table A.3:** Values used to calculate the saturated hydraulic conductivity of the MSWI BA investigated.

The saturated hydraulic conductivity of the MSWI BA investigated was calculated to be 2292 cm/h.

## D pH-Dependence Leaching Test

### D.1 Acid and Base Addition

Prior to the pH-dependence leaching test, the acid and base neutralization capacity of the BA was analysed. This was done according to guidelines in CEN/TS 14429. Continuous addition of a known volume of 1 M HNO<sub>3</sub> and 1 M NaOH into bottles containing 180 mL distilled water and 20 g BA was done. It started with an L/S of 9 L/kg and aimed at a final L/S ratio of 10 L/kg after acid or base addition. Acid and base was added until the desired pH of 4 and 12 was obtained. The bottles were equilibrated and stirred for 48 h on a shaking table at 130 rpm. The pH was measured several times during the 48 h to follow the pH-development and acid or base was added if needed. By the end of the test, the volume of acid and base needed to obtain pH 4 and pH 12 in the bottles was known. This was later used to conduct the pH-dependence test, see Table A.4.

	pH 4	Natural pH	pH 12
Volume acid, 1 M HNO <sub>3</sub> (mL)	10	0	0
Volume base, 1 M NaOH (mL)	0	0	2
Volume distilled water (mL)	190	200	198

**Table A.4:** Volumes of acid, base and distilled water used in the pH-dependence test.

The buffer capacity for the MSWI BA was calculated as the number of moles of H<sup>+</sup> needed to change the pH of a 1 L solution with one unit. This was calculated from the fact that 10 mL 1 M HNO<sub>3</sub> changed the pH in the 190 mL distilled water and BA solution used in the leaching test from 10 to 4. Hence, the buffer capacity was calculated to be approx. 0.009 mol H<sup>+</sup>/L.

### D.2 Sample Preparation

The CEN/TS 14429 require that the material is dried prior to the test. Since the MSWI BA is a product of incineration, it could be considered dry, but a drying test was performed to see if the weight reduction was considerable. 15 g BA was dried at 105 °C for 24 hours.

Weight before (g)	Weight after (g)	Weight reduction (%)
14.990	14.817	1.15

**Table A.5:** Values from drying test of MSWI BA.

The weight reduction was only 1.15 %, and therefore considered negligible. Hence, the MSWI BA was considered dry, and no drying of the BA was performed prior to the the

leaching test. An L/S of  $10 \pm 0.2$  mL/g would still be achieved, as the standard required.

### D.3 pH-Development During the Test

pH was measured during the leaching test to follow the pH-development, according to CEN/TS 14429. The electrical conductivity was measured after 48 hours.

	pH after 2 hours	pH after 4 hours	pH after 44 hours	pH after 48 hours	EC after 48 hours (uS/cm)
pH 4-1	3.00	2.11	4.09	4.14	8050
pH 4-2	2.17	1.87	3.79	3.82	7890
Natural pH-1	10.07	10.08	9.83	9.76	948.2
Natural pH-2	10.06	10.06	10.13	10.06	788.7
pH 12-1	11.71	12.88	12.62	12.59	2246
pH 12-2	11.53	12.88	12.77	12.75	1973
Blank pH 4	1.37	0.84	0.83	0.86	30.24
Blank natural pH	9.47	6.80	6.61	6.85	2.07
Blank pH 12	11.50	12.99	12.99	12.99	2171

**Table A.6:** *pH at different hours during the pH-dependence leaching test. EC = electrical conductivity.*

## E Thomas Model

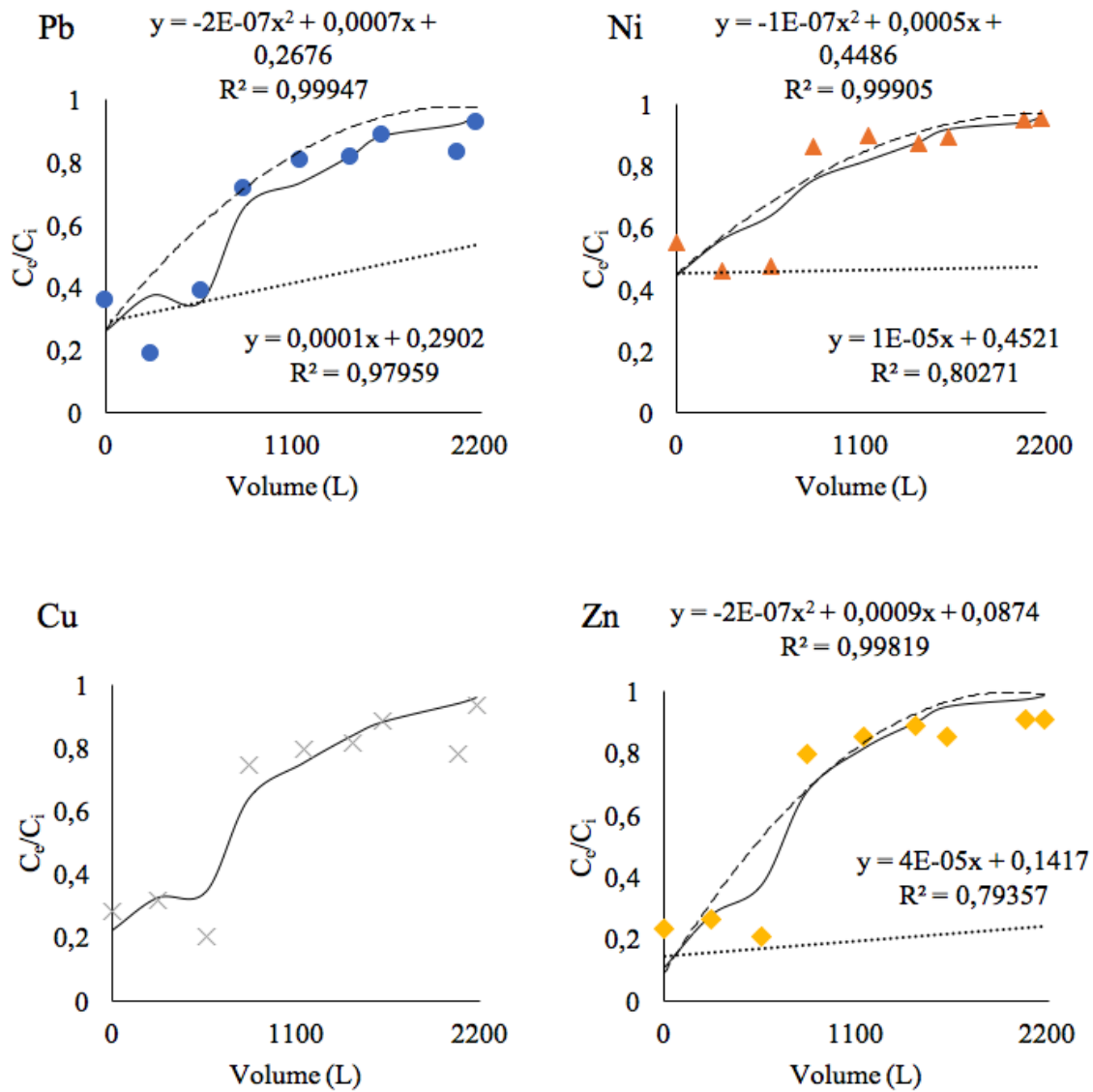
### E.1 Adjusted Thomas Model

The Thomas model was fitted to the column data. It showed good fit, according to the  $R^2$ , see Table A.7. Since the column was fed with 2 mg/L of metals, which is a concentration higher than what is expected in stormwater, the Thomas sorption capacity and rate constant were adjusted for representative stormwater concentrations. These were found in Lindholm (2004). The adjustment was done as in Paus et al. (2014), with the use of isotherm constants fitted from batch experiment. No isotherm fit was found from the batch test for Cu. Therefore, its Thomas sorption capacity and rate constant were not adjusted for representative stormwater concentrations.

	Pb	Ni	Zn	Cu
$q$ (mg/g)	0.29	0.08	0.43	0.33
$k_{tk}$	0.03	0.02	0.04	0.03
$R^2$	0.89	0.81	0.90	0.90
$C_{sw}$ (mg/L)	0.031	0.0044	0.197	0.072
$q_{sw}$ (mg/g)	0.02	0.01	0.64	-
$k_{th,sw}$	0.39	0.18	0.03	-

**Table A.7:** Thomas sorption capacity and rate constant from the column test,  $q$  and  $k_{th}$ , and adjusted for representative storm-water concentrations,  $q_{sw}$  and  $k_{th,sw}$ . Stormwater concentrations,  $C_{sw}$ , from Lindholm (2004).  $R^2$  from fitting Thomas model.

The new Thomas sorption capacity and rate constant were used to model the behaviour of the filter media over time at representative stormwater metal concentrations. The Thomas sorption capacity and rate constant from the column tests were applied to a constant inflow concentration of 2 mg/L. This generated the following curves for the four metals, see Figure A.3.



**Figure A.3:** Symbols = Measured concentrations from column test. Line = Thomas model fitted to column concentrations. Broken line = Thomas model with constant inflow concentration of 2 mg/L (upper equation). Dotted line = Thomas model adjusted for representative stormwater concentrations (lower equation).

## E.2 Lifetime Calculations

When determining the lifetime of the filter, Equation (5) was used (Paus et al. 2014):

$$y = \frac{d_f V_b A_c}{P C A_f V_m} \quad (5)$$

where  $y$  is the number of service years,  $d_f$  is the depth of the filter (m),  $V_b$  is the accumulated volume through the filter before a certain metal breakthrough (L),  $A_c$  is the catchment area (m<sup>2</sup>),  $P$  is the annual precipitation (m/y),  $C$  is the runoff coefficient,  $A_f$  is the filter area (m<sup>2</sup>) and  $V_m$  is the volume of the filter media (L).  $V_b$  was calculated from the equations for the fitted curves in Figure A.3 and  $P$  was set to 895 mm/y as for the town Trondheim, Norway (Monrabal-Martinez et al. 2017).  $C$  was set to be 0.9. The ratio  $A_c/A_f$  was assumed to be 20 as used for rain gardens in Paus et al. (2014).

When determining the volume of the filter media,  $V_m$ , the BA particles were assumed to be spherical. The volume of one average particle was calculated, the mean weight of ten BA particles were found and the total weight of BA in the columns was known. From this and the total volume of the column, the volume of the filter media,  $V_m$ , was estimated.