

**Predicting the source strength of recycling materials
within the scope of a seepage water prognosis by
means of standardized laboratory methods**

Dissertation
zur Erlangung des Doktorgrades
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Preface

The presented thesis was written in the frame of a project called ‘seepage water prognosis’. It was funded by the Federal Ministry for Education and Science (BMBF). 41 German institutions among them research institutes of universities, public authorities and engineering companies were financed for three years respectively. The aim was to work out the scientific basis that is needed to carry out a seepage water prognosis (Oberacker und Eberle, 2002). According to the Federal German Soil Protection Act (Federal Bulletin, 1998) a seepage water prognosis is required in order to avoid future soil impacts from the application of recycling products. The participants focused on the development of either methods to determine the source strength of the materials investigated, which is defined as the total mass flow caused by natural leaching or on models to predict the contaminants transport through the underlying soil. Annual meetings of all participants as well as separate meetings of the two subprojects were held.

The department of Geosciences in Bremen participated with two subprojects. The aim of the subproject that resulted in this thesis was the development of easily applicable, valid, and generally accepted laboratory methods for the determination of the source strength. In the scope of the second subproject my colleague Veith Becker developed a computer model for the transport prognosis with the source strength as the main input parameter.

In this doctoral thesis four manuscripts have been included.

Chapter 2.1.: Predicting the source strength of various recycling materials – Methods and preliminary results

Authors: Tanja Lager, Kay Hamer, Jürgen Schröter und Horst D. Schulz

In this article an overview and a detailed description of all applied laboratory batch methods and column tests are given. Some material parameters of the four materials investigated, sandy and fine fraction of harbour sediments, municipal waste incineration ash and demolition waste are shown. In addition, first results of the experiments conducted with the fine fraction of harbour sediments are presented. The manuscript includes a model for the calculation of the acid neutralisation capacity consumption in a horizon of the respective fraction of harbour sediments.

Chapter 3.1.: Mobility of heavy metals in harbour sediments - an environmental aspect for the reuse of contaminated dredged sediments

Authors: Tanja Lager, Kay Hamer und Horst D. Schulz

The results of four different batch experiments performed with sandy and fine fractions of harbour sediments are presented and release-controlling factors are compared. In addition pore water data from the unsaturated fine fraction are shown. The column experiments run under saturated and unsaturated conditions with the sandy fraction of the material are compared. Regarding all applied methods heavy metals, arsenic and molybdenum are considered with respect to a possible reuse of the two fractions of harbour sediments.

Chapter 3.2.: Comparison of Leaching Tests to Determine the Release of Inorganic Contaminants from Demolition Waste

Authors: Markus Delay, Tanja Lager, Horst D. Schulz und Fritz H. Frimmel

This manuscript evolved in cooperation with Markus Delay from the Engeler-Bunte-Institute in Karlsruhe. The focus lies on the comparison of column tests performed by Markus Delay and myself. The columns were both run under water-saturated conditions but had different dimensions, flow regimes and absolute time spans. In a next step, the results of the laboratory test methods were compared with field-scale experiments (lysimeters) performed with the same material and two batch tests.

Chapter 3.3: Determination and Quantification of the release of Inorganic Contaminants from Municipal Waste Incineration Ash

Authors: Tanja Lager, Markus Delay, Volker Karius, Kay Hamer, Fritz H. Frimmel und Horst D. Schulz

Markus Delay and myself wrote this manuscript in equal shares. Data from the leaching tests performed with municipal waste incineration ash are presented. The script thereby focuses on the comparison of the different methods applied, which are batch tests, column tests and field-scale experiments. Markus Delay conducted one column test, the batch tests as well as the second column test were performed by myself. Modelling with the program PHREEQC (version 2.8) (Parkhurst and Appelo, 1999) was performed in corporation with Volker Karius from the Geosciences Centre at the University in Göttingen.

The references of all manuscripts are displayed comprising in chapter 4. Figures and tables are numbered within the respective script independently from the entire thesis. All data, including data that are not listed in tables or displayed in figures in the respective chapter, are listed in the appendix.

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1. Introduction and objectives

Next to the remediation of contaminated sites the main purpose of the Federal German Soil Protection Act (Federal Bulletin, 1998) is the protection and restoration of the soil functions on a permanent and sustainable basis. This includes the protection against harmful changes that can derive from the application of materials onto or incorporated into the soil (Federal Bulletin, 1999). Thus the responsible handling of mineral wastes and other recycling materials that accrue in large quantities like municipal waste incineration ash, demolition waste material and harbour sediments is a major task.

In Germany, about 4 million tonnes of municipal waste incineration ash are generated every year out of 40 million tonnes of municipal waste (German Federal Environmental Agency, 1999). At the same time between 75 and 85 Mio tonnes of demolition waste material, not included excavated soil, accrue each year (German Federal Environmental Agency, 2002). Furthermore, waterways have to be dredged regularly in order to be able to guarantee a secure shipping traffic. 40 million m³ of sediments are dredged annually in the area of the North and Baltic Sea, about 5 million m³ inland (Hafenbautechnische Gesellschaft, 2002). In Bremen 400.000 m³ accumulate each year.

In order to be able to evaluate the feasibility of an application of these recycling materials in view of the consequences for the underlying soil the Federal German Soil Protection Act (Federal Bulletin, 1998) demands a seepage water prognosis. Threshold, precaution and action values are defined in the act for the assessment. The main focus therewith lies on the contaminants passage from the contaminated material through the underlying soil into the groundwater. The prognosis may be divided into two steps, the determination of the source strength of the material and the modelling of the contaminants passage through the soil. The source strength is defined as the total mass flow caused by natural leaching (Oberacker and Eberle, 2002) and should be determined at the place of sampling, which is the transition area between unsaturated and saturated zone. In order to find suitable methods and models for both steps the Federal Ministry for Education and Science funded a project called 'seepage water prognosis'. 41 institutions applied various methods on the same materials. For this purpose the Federal Department of Material Investigation (BAM) generated three reference materials according to ISO Guideline 30 (1992). The municipal waste incineration ash was a mixture of ashes from 4 different waste incineration plants in Germany, the demolition waste material was

made of 13 different materials and the soil was generated by mixing 16 different soils, all deriving from tar factory sites.

The aim of the project described in this work was to find methods to determine the source strength of the different recycling materials focussing on inorganic contaminants. Of the reference materials generated by the Federal Department of Material Investigation (BAM) only the municipal waste incineration ash and the demolition waste material were investigated in the frame of the investigations for this doctoral thesis. The soil was exclusively enriched with organic contaminants. Additionally the fine and the sandy fractions of harbour sediments from the harbour sediment disposal site in Bremen Seehausen were examined.

As the source strength is defined as the total mass flow caused by natural leaching the main requirements next to the general acceptance and the practicability is the validity of the method (Oberacker and Eberle, 2002). Validity comprises the reproducibility of the method, the applicability for different materials and the reflection of the natural conditions. Numerous scientists did a lot of investigations in the past 10 years in order to find methods that fulfil these requirements (e.g. van der Sloot et al., 1997; van der Sloot, 2000; Wahlström, 2000; Kosson et al., 2002). In this study a variety of batch tests as well as percolation tests were performed and compared. In the frame of the project 'seepage water prognosis' the Research Centre for Environment and Health (GSF) in Neuherberg, southern Germany and the North Rhine-Westphalia State Environment Agency (LUA NRW) in Essen conducted field scale column experiments (lysimeters) with municipal waste incineration ash and demolition waste material. The contaminant concentrations from these lysimeters are closest to the natural leaching of the investigated material and are thus taken as reference value for the source strength.

At first, the various methods applied will be presented in chapter 2. By means of some parameters first statements can be made about the comparability of the methods. Chapter 3 comprises the results of the application of these methods to the three materials investigated with regards to the determination of the source strength.

2. Methods

2.1. Predicting the Source Strength of Different Recycling Materials – Methods and Preliminary Results

Tanja Lager, Kay Hamer, Jürgen Schröter and Horst D. Schulz

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Predicting the Source Strength of Different Recycling Materials – Methods and Preliminary Results

Tanja Lager, Kay Hamer, Jürgen Schröter and Horst D. Schulz

Abstract

The German Federal Soil Protection Act (Federal Bulletin, 1998) demands a prediction of the transport of pollutants with the seepage water if waste materials are reused. Within the framework of a program funded by the Federal Ministry for Education and Science (BMBF) called ‘Seepage Water Prognosis’ methods to obtain relevant parameters for the prediction of the transport of pollutants through the unsaturated zone should be employed, compared and, if required, refined. Our investigations on diverse contaminated materials aim at the determination of their source strength and temporary variations. The three materials investigated in this project are (1) sediment dredged from the port of the city of Bremen, (2) municipal waste incineration ash, and (3) demolition waste. The study presented here focuses on inorganic contaminants. Crucial are the pH and E_H conditions as well as the liquid/solid ratio. Four batch tests, the DEV S4 test, the soil saturation extract, ammonium-nitrate extract, the pH-stat test, and column experiments were performed and compared. The acid neutralisation capacity is determined by means of pH-stat tests. Thus the time period until a soil horizon is acidified can be calculated. The oxidation and reduction capacity can be calculated on the basis of the potential electron transfer of redox reactions and thus enable a prediction when a sediment layer will be oxidised or reduced. In this paper we focus on the presentation of the laboratory methods. Preliminary results of the batch experiments are presented. In addition, results of the pore water analysis of the fine fraction of harbour sediments, providing the most realistic estimation of the source strength gained by dialysis and centrifugation, are shown.

Introduction

According to the German Federal Soil Protection Act (Federal Bulletin, 1998) a ‘Seepage Water Prognosis’ has to be performed before depositing mineral wastes or recycling products onto or into soils. The prognosis is also required for the evaluation of contaminated depositions. According to the Soil Protection Act, the seepage water prognosis has to assess the potential

groundwater contamination via seepage water in the foreseeable future. This prognosis has to be performed in two steps:

- Determination of the source strength of the contaminated material, which is defined as the total mass flow caused by natural leaching (Oberacker and Eberle, 2002); in case of contaminated sites the source strength corresponds to the concentration of the seepage water at the place of sampling (Fig. 1)
- Transport prognosis of the passage of a contaminant into the groundwater through the underlying soil and the unsaturated zone, with the source strength as the main input parameter.

The place of assessment is defined as the transition area between unsaturated and saturated zone (Fig. 1). The decision whether contaminated material can be safely deposited at a specific site without harming the environment may be made by comparing the results of the seepage water prognosis with the legal threshold values.

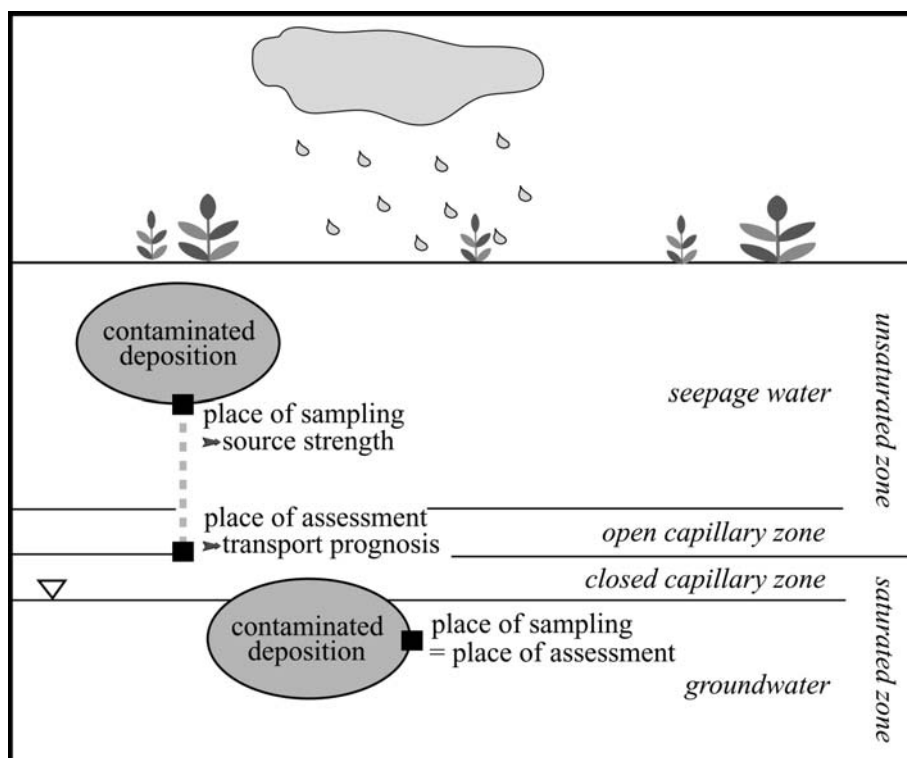


Figure 1: Seepage water prognosis: determination of the source strength at the place of sampling and performance of a transport prognosis to determine the contamination of the seepage water at the place of assessment

It is thus necessary to determine the quantity and quality of seepage water that is released from a deposit into the underlying soil (unsaturated zone or groundwater) in a given timeframe. Crucial for the variability of the source strength are the pH and E_H conditions in the soil or in the deposit

regarded (van der Sloot, 1998). Starting from the bulk concentration of the solid-phase, batch experiments like the standardised German DEV S4 test (DIN, 1984), soil saturation extract (Federal Bulletin, 1999) ammonium-nitrate extract (DIN, 1997b) or the pH-stat test (Obermann and Cremer, 1992; Karius and Hamer, 2001b) provide only a snapshot of the potential mobilisation of contaminants. Therefore, it is important to indicate the proceeding changes of the pH and E_H conditions, whereby sorption, dissolution and ion exchange reactions are influenced and thus the entire seepage water quality.

In the study presented here, we focus on different methods to obtain the source strength from the materials investigated, which consist of harbour sediments, municipal waste incineration ash (MWI ash), and demolition waste to discuss the applicability and validity of the experiments.

For further information regarding the BMBF research program ‘Seepage water Prognosis’ and the scientific work of 41 participants reference is made to Oberacker and Eberle (2002).

Material

The harbour sediments investigated here were sampled from a drainage field of the disposal site for dredged material in Bremen- Seehausen. Annually, a volume of about 400.000 m³ (measured *in situ*) harbour sediments is dredged to maintain the water depth in the city harbours of Bremen (harbour Authorities, personal communication). Because of the enrichment of various pollutants in these sediments, they are deposited at a landfill site, after a preceding drying period of approximately one year. In Bremen, the life span of the landfill for dredged material constitutes a period of 15-20 years – providing sufficient time to find alternative remediation strategies.

The harbour sediments can be subdivided into two fractions. A separation according to grain-size takes place when the drainage field is filled. The sandy fraction accumulates close to the outlet of the pipeline through which the dredged material is pumped into the drainage field with the use of excess water. The main amount of the sediment, the sandy, clayey silt fraction is deposited throughout the whole drainage field. After a drying period of almost one year, the sandy fraction usually has a water content of about 5 wt.-% OS and can be used as building material or drainage sand within the disposal site. The water content of the fine fraction of the harbour sediments averages approximately 35 wt.-% OS and could consequently be either deposited or reused. During the drainage period the material is besieged 3-4 times, which results in a homogenisation of the fine fraction of harbour sediment (Hamer et al., 1997). The physical properties of this material allows an utilisation as:

- building material for noise protection embankments, dikes, restoration layers

- mineral landfill surface sealing (von Lührte, 1997; von Lührte et al., 1999)
- raw material for bricks and lightweight aggregates (Karius and Hamer, 2001a, b; Hamer and Karius, 2002)

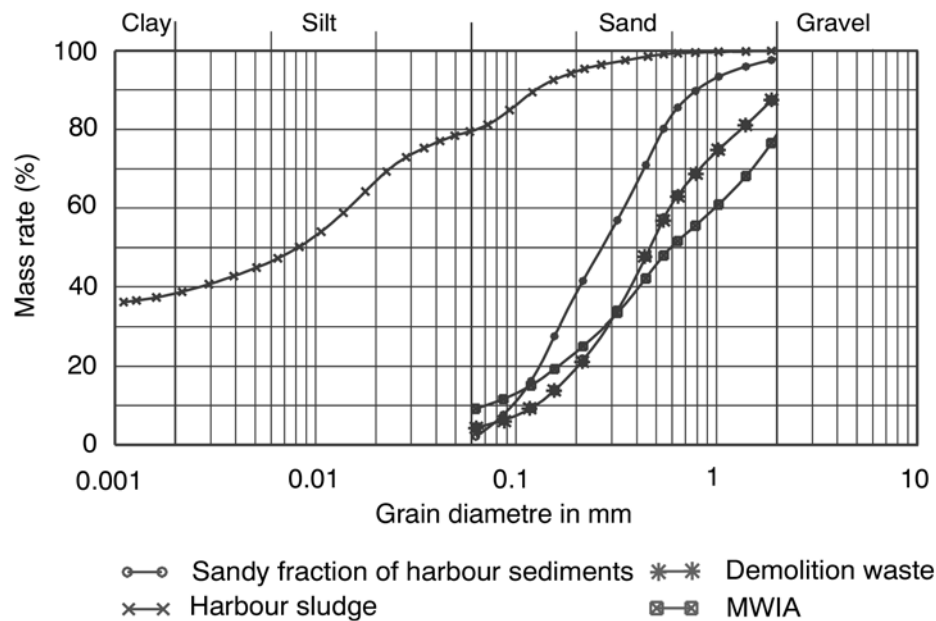


Figure 2: Grain-size distribution of various investigated materials

Municipal waste incineration ash (MWI ash) and demolition waste are considered as reference materials and are as such investigated by every institute involved in the BMBF research programme. Both materials are provided and prepared by the Federal Institute for Material Research in Berlin (BAM). The municipal waste incineration ash as well as the demolition waste are clayey, silty gravely sands and both have a water content of less than 5 wt.-% OS. Figure 2 shows the grain-size distribution of all materials investigated.

The material characterisation, which will be carried out with all materials mentioned includes following parameters:

- water content (DIN, 1989)
- specific weight (grain density) (DIN, 1997a)
- grain-size distribution (DIN, 1996)
- potential cation exchange capacity (DIN, 1997c); effective cation exchange capacity (DIN, 1997d)
- specific surface area (BET)
- total content

Determination of the total content is carried out by total digestion of the sample by the application of microwave heating and subsequent analysis. The procedure was developed according to DIN 38414-7 (1983).

The following parameters will be analysed in all samples after filtration (0.2 μ m):

- main elements (Al, Ca, Fe, K, Mg, Na, Si, SO₄) by ICP-AES
- trace elements (As, Ba, Cd, Co, Cr, Cu, Mn, Mo, Ni, Pb, Rb, Sr, V, Zn) by ICP-MS
- Total Carbon (TC), Inorganic Carbon (IC), Total Organic Carbon (TOC)
- anions (Cl, SO₄, NO₃) by HPLC
- pH, E_H, electric conductivity

Methods

Various batch experiments have been developed in order to evaluate the mobilisation of inorganic contaminants released from wastes and contaminated soils, e.g. the DEV S4 test (DIN, 1984), ammonium-nitrate extract (DIN, 1997b) and soil saturation extract (Federal Bulletin, 1999). They all give information about the short- term leachability of the contaminants.

Table 1: Test conditions and some release controlling factors of the applied methods

Method	Grain-size	pH value	Liquid/solid ratio [L/kg] / flow conditions	Elution time	Elution agent	parallel tests
DEV S4 test	< 1 cm	dependent on material	10 / overhead shaker	24 h	deionised water	3
Soil saturation extract	original	dependent on material	~1 / kneading / stirring for 15 min respectively	48 h	deionised water	3
ammonium- nitrate extract	< 2 mm	dependent on material and NH ₄ NO ₃	0.4 / overhead shaker	3 h	1 m NH ₄ NO ₃	3
pH stat test	< 2 mm, 63 μ m – 2 mm	4, 5, 6, 7, 11	10 / stirrer	24 h	deionised water	3
Saturated column	< 2 mm	4	variable / 0.1 mL/min	weeks to month	artificial rainwater	1
Unsaturated column	< 2 mm	4	variable / 50 mL/day in 1 h	weeks to month	artificial rainwater	1
Dialysis	original	dependent on material	adequate to natural / none	weeks	pore water	3
Centrifugation	original	dependent on material	adequate to natural / none	1 h	pore water	3

The pH-stat test was first developed by Obermann and Cremer (1991) and has been refined by Karius and Hamer (2001a, b). Meanwhile, a preliminary standard EW 98p has been developed by LAGA (2001). The column experiments applied, the two pore water extraction methods, the dialysis technique, and a special centrifugation technique for unsaturated material are internal developments of our institute. Tab. 1 shows the test conditions of the methods, which were applied in this study.

In addition to the laboratory experiments, *in situ* pH- and E_H -measurements of the sampled sediments were conducted at the dredged material deposit in Bremen- Seehausen.

The DEV S4 test

The German standard leaching test (DIN, 1984) is a commonly applied method. It was developed to assess the environmental compatibility of residues, wastes and slurries. It permits the determination of substances that are leached in the short term. Here, 100 g of a sample with a maximum grain-size of 10 mm are leached with 1000 mL deionised water at room temperature for 24 hours in an overhead shaker.

The DEV S4 test is supposed to indicate the potential hazard to waters if the regarded material is deposited in contact with water. However, the liquid-solid ratio of 10 foreseen by this method may be considered as an unrealistic event. In addition, shaking of the sample causes abrasions that produce new surfaces, which, in turn, leads to a higher mobilisation rate as compared to undisturbed deposition.

Ammonium nitrate extract

The standardised extraction of trace elements with ammonium- nitrate solution (DIN, 1997b) is performed with a liquid-solid ratio of 0.4. 20 g of the soil sample are leached with 50 mL 1M ammonium-nitrate at room temperature for 2 hours in an overhead shaker.

The ammonium-nitrate extract has a more natural liquid/solid ratio, but is only applicable to trace elements. Problems occur mainly on account of the preferential extraction of copper and cobalt by means of NH_4^+ - complexation (J. Gerth, TU Hamburg-Harburg, personal communication with). The supplementary high ionic strength leads to an increased retention of e.g. arsenic in soils with a predominantly variable charge, e.g. samples rich in iron oxide (Dankwarth and Gerth, 2002).

Soil saturation extract (SSE)

The soil saturation extract was developed for leaching inorganic contaminants (Federal Bulletin, 1998). Deionised water is added to 250 g of the sample, until the sample is fully imbued. The soil sample is thoroughly mixed and left to stand at room temperature for 24 hours. Then the sample is put into a centrifuge tube and deionised water is added until the flow limit is reached. This equals a liquid/solid ratio of approximately 1 L/kg. After further 24 hours of storage at 5°C the sample is centrifuged for 30 min at 20°C.

The soil saturation extract has a liquid/solid ratio that resembles natural conditions better than the DEV S4 test and ammonium-nitrate extract. However, the added amount of water is not strictly defined in the soil saturation extract ('adding of water until the flow limit is reached'). This adversely affects the reproducibility of this method. In addition, the soil saturation extract is not applicable to other materials than soils. Parallel tests have to be carried out when applying this method to materials with a sandy or even coarser grain-size, in order to obtain a sufficient amount of sample for all analyses required. Furthermore municipal waste incineration ash is known to undergo consolidation reactions when coming into contact with water, or oxygen and carbon dioxide of the ambient air (Hirschmann, 1999; Pfrang-Stotz and Reichelt, 1999). Most municipal waste incineration ashes contain portlandite ($\text{Ca}(\text{OH})_2$), anhydrite (CaSO_4), bassanite ($\text{CaSO}_4 \cdot 0.5 \text{H}_2\text{O}$), gypsum ($\text{CaSO}_4 \cdot 2 \text{H}_2\text{O}$), calcium silicates and calcium aluminates, which react as binding agents (Khorasani et al. 1989). Demolition waste mainly consists of crushed concrete and bricks, and thus contains binding agents as well.

pH-stat Test

The pH-stat test is a suitable technique to assess the pH-dependent leaching of inorganic contaminants. In this study, 50 g of the sample are leached with 500 mL of deionised water. A consistent suspension is guaranteed by continuous stirring. The pH is kept at a set value by stepwise addition of nitric acid (suprapure®) or sodium hydroxide (suprapure®). The consumption of the acid or base is used to calculate the acid neutralisation capacity (ANC) or the base neutralisation capacity (BNC), respectively. The time limit of the performance of the pH-stat test is 24 h. As this does not allow the calculation of this factor in absolute figures, the ANC and BNC are defined as ANC_{24} and BNC_{24} .

The chosen pH values (4, 5, 6, 7, 11) represent the various steps that are possible in the life cycle of a recycling material from exposure to acid rain (pH 4-5) to its utilisation in road construction (pH 7-11) or as landfill surface sealing material (pH 5-7).

Column Experiments

For both, the saturated and the unsaturated column experiments Perspex tubes 50 cm in length are employed. The inner diameters of the saturated and unsaturated column are 5 cm and 10 cm, respectively. The water used in the analysis is in both cases tap water diluted with deionised water down to a water hardness of 0.0895 mmol/L. This value corresponds to rainwater. The analysis water is pumped with an Ismatec IPC-12 ISM762 from the storage tank into a mixing cell. In the mixing cell, nitric and sulphuric acid suprapure® are added with an infusion pump (Braun Perfuso segura FT), to adjust the pH to a value of 4. Afterwards, the water is pumped into the columns. The main weathering agent that the recycled materials are exposed to is acid rain. We therefore decided on a worst -case value of pH 4. The sampling as well as the measurement of pH, E_H and electric conductivity at the outlet of the columns occurs continuously during the first few days and once a day in the following. The withdrawal of samples is carried out by means of an autosampler. The columns are designed to allow sampling along the flow-path. Hence, next to the loads, the pH- and E_H -dependent processes of mobilisation and fixation of the regarded contaminants can be investigated.

Saturated Column

The columns are sealed with a Perspex plate at the bottom and a polyethylene plug at the top. Inlet and outlet of the columns are connected to tygon® tubes (Fig. 3).

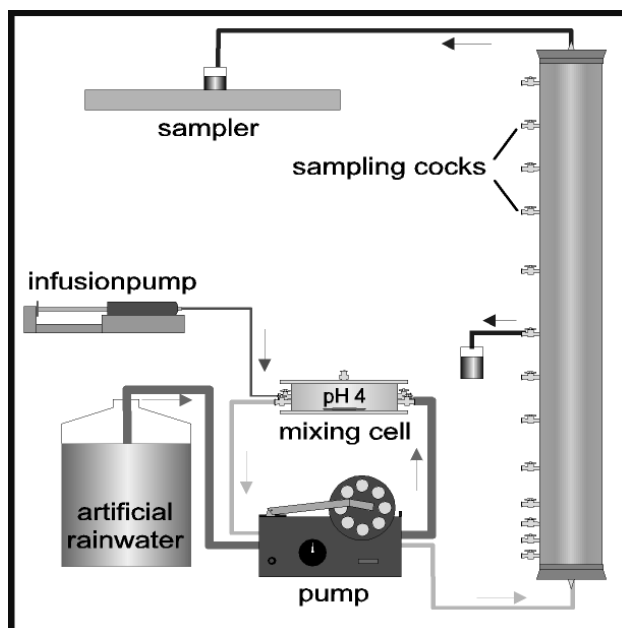


Figure 3: General structural design of the saturated column experiment

Filling the columns is done with water-saturated material, with a slight excess of water in order to avoid air inclusions and grain-size fractionation. The artificial rainwater flows from the bottom to the top of the column. The annual amount of precipitation in Germany averages at 825 mm, a value derived from the international standard period 1931-1960 (Mattheß, 1983). Depending on evaporation, transpiration, and on the type of soil, 200-300 mm comply with the renewal of groundwater. Based on the area of the column that the artificial rainwater is flowing through, the volume of water to be pumped is 1.0–1.6 mL per day, which would not yield a sufficient sample volume. Therefore, we decided on a pump rate of 0.1 mL/min, which is equivalent to a groundwater renewal rate of approximately 26,770 mm/a. This represents a compromise between the naturally prevailing conditions and the time- related and technical restrictions in a laboratory.

The columns are equipped with stopcocks situated at each centimetre over the first 10 centimetres and every second centimetre over the remaining length (Fig. 3), in order to allow sampling along the flow-path. At the end of each column experiment, a tracer experiment including lithium bromide (detected as electric conductivity) is carried out to describe the hydraulic regime of each column.

Unsaturated Column

The air-dried material is filled into the column. Irrigation is carried out through six stopcocks attached to a Perspex plate, which is attached to the top of the column (Fig. 4).

As mentioned above, the annual amount of precipitation in Germany averages 825 mm. Less than 2.5 L/(m²·h) of rain precipitate during a weak rain event, more than 7.5 L/(m²·h) during a strong rain event (Matteß, 1983). Considering the area of the column, this precipitation rate is equivalent to approximately 20-60 mL/h. In the experiment an irrigation of 50 mL/h is carried out daily which equals an annual precipitation rate of approximately 2,300 mm. Considering the evaporation which averages 20% in an air-conditioned laboratory at a constant temperature of 22°C +/-1, the sample volume will be sufficient for all required analyses. Within the first 24 h, the column is irrigated every second hour in order to enable initial sampling on the second day of the experiment.

A gauze (200 µm) is attached to the bottom of the column, on top of which there is a layer of coarse-grained quartz sand and a second layer of fine-grained quartz sand, each measuring 1.5 cm column length, to prevent retention of water seeping through the material in the column. A sample chamber with a volume of approximately 500 cm³ is mounted underneath the gauze. Samples collected in the sample chamber are taken once a day with a syringe. To enable

sampling along the seepage-path, soil moisture samplers (Eijkelkamp) are incorporated each three centimetres along the entire length of the column. A soil moisture sampler consists of a 10-cm long microporous polymeric tube with a diameter of 2.5 mm. It is stabilised with an internal stainless- steel rod. The microporous tube is connected to a 10- cm long PVC tube, with a Luer-lock connector at the end. Pore water can be sampled by applying a vacuum with a syringe.

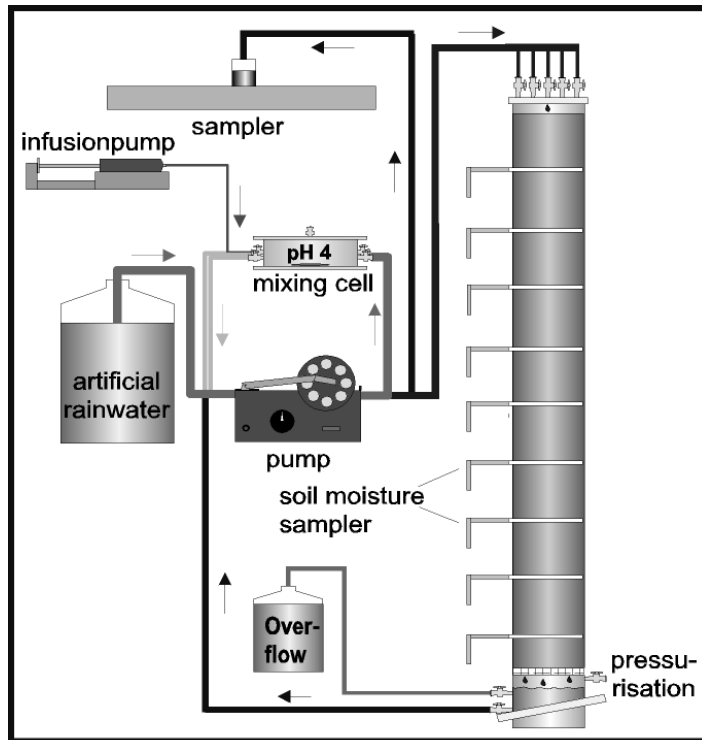


Figure 4: General structural design of the unsaturated column experiment

Dialysis

Dialysis technique is a method designed for the *in situ* collection of pore water samples. The principle is to let a certain volume of deionised water reach an equilibrium with the surrounding pore water by diffusion through a Teflon® membrane.

The dialysis pole employed in this project (Fig. 5) was developed by Dahmke et al. (1991). The advantages compared to other dialysis devices (e.g. Hesslein, 1976; Carignan, 1984, Schwedhelm, 1988) consist in its easy incorporation into the soil, the provision of a large chamber volume (24 mL), sampling with a resolution of 3 cm, two opposing chambers at one depth, and a sampling depth of 2 m. The dialysis pole has been successfully employed in water-saturated sediments of tidal flats, with an equilibrium time of about 8 weeks (Lücke, 1991).

Since the fine fraction of the harbour sediments is unsaturated, deionised water has to be added to the investigated sample until the water is saturated. The dialysis pole is first placed in the

middle of a bucket and the material is then filled around it, compacted, and stored in a cold-storage room at 3-5°C. Three parallel tests are performed lasting 6, 8 and 12 weeks, respectively.

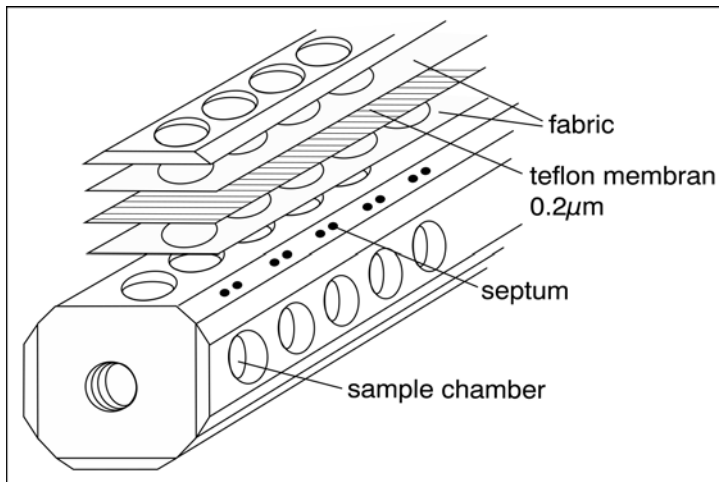


Figure 5: General structure of one dialysis pole segment

Centrifugation

In order to compare pore water sampling with the dialysis pole, the centrifugation-technique is used, a commonly applied ex situ sample technique to obtain pore water. It is difficult to obtain pore water from the fine fraction of harbour sediments with an ordinary centrifuge tube, since the material is not saturated with water. A modified approach for unsaturated sediments is used here (Fig. 6).

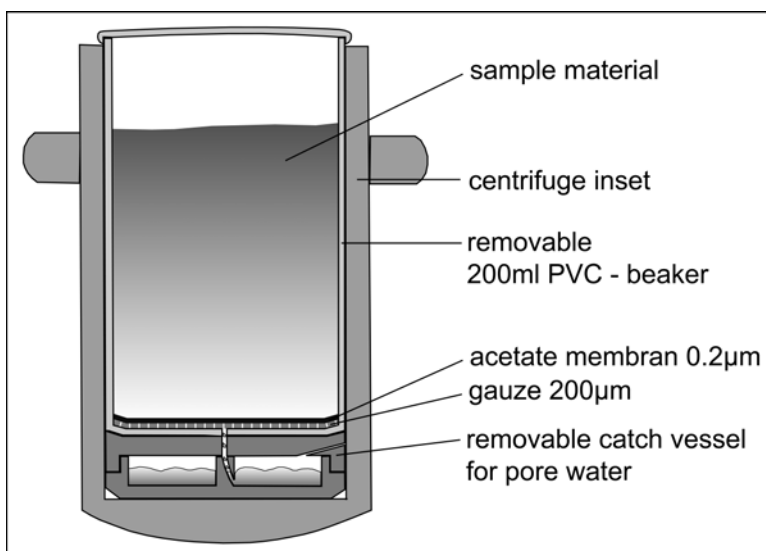


Figure 6: Centrifuge inset for pore water extraction from unsaturated material

The centrifuge tube has three little holes in the bottom, a filter on top, and a sample cup below the tube. The pore water is transported by rotation towards the bottom of the tube and can leave

the tube, filled with the sediment, through the holes (Fig. 6). These modifications yield a sample volume sufficient for all analyses. Centrifugation is performed for 20 min at 3,700 g.

Results and Discussion:

The preliminary results presented are the pH and E_H values as well as the electric conductivity of the applied batch experiments including the three materials. Furthermore, the analysis of the main elements from the batch experiments performed and the methods to extract pore water from the fine fraction of the harbour sediments are shown.

Tab. 2 demonstrates the pH and E_H values, and the electric conductivity of the fine fraction of harbour sediments after the application of several methods, in addition, the *in situ* values measured at the dredged material deposit. Apart from the pH-stat test, the pH values of all methods and the *in situ* measurements are in the neutral range. The E_H values range between +365 and +535 mV, unless measured under *in situ* conditions (+30 and +420 mV). The *in situ* values indicate that the fine fraction of the harbour sediments is banked up in the drainage field, where the material piles up in aggregates. The surface of these aggregates has contact to atmospheric oxygen, whereas the interior parts of the aggregates are almost free of oxygen.

Table 2: pH and E_H values, and electric conductivity of the fine fraction of harbour sediments after the application of various methods

Method	pH	E_H [mV]	Electric conductivity [mS/cm]
DEV S4 test	7.6	510	0.5
ammonium-nitrate extract	7.0	365	64.0
Soil saturation extract	7.2	450	3.4
pH 4 stat test	4.0	470	5.6
pH 5 stat test	5.0	510	5.0
pH 6 stat test	6.0	535	3.9
pH 7 stat test	7.0	485	1.8
Dialysis	7.0	470	2.2
Centrifugation (dialysis)	7.3	400	3.9
Centrifugation (original)	7.3	450	5.0
<i>in situ</i>	6.5 – 7.0	30 - 420	n.d.

n.d.: not detected

The electric conductivity depends on the concentration, the quality of the solution and hence on the leaching agent. Consequently, the electric conductivity by itself is not appropriate for the

comparison of the various methods, on account of the fact that the leaching agents are different. However, it reflects the trend seen in the calcium concentrations shown in Figure 7. The electric conductivity is appropriate to make a first comparison regarding the total ionic content of solutions gained by applying different methods (Tab. 2) and different materials (Tab. 3).

Table 3: pH and E_H values, and electric conductivity of the investigated materials in the DEV S4 test and ammonium-nitrate extract

Material	Sandy fraction of harbour sediments		Fine fraction of harbour sediments		Municipal waste incineration ash		Demolition waste	
	DEV S4	NH ₄ NO ₃	DEV S4	NH ₄ NO ₃	DEV S4	NH ₄ NO ₃	DEV S4	NH ₄ NO ₃
pH	8.3	7.1	7.6	7.0	11.3	9.1	12.0	8.5
E_H [mV]	580	38	510	365	265	310	215	220
El. conductivity [mS/cm]	1.5	80	0.54	64	1.2	81	3.4	76

The concentrations of calcium (exemplary for calcium, potassium, magnesium, strontium, barium) and sulphur (exemplary for sulphur, sodium) of some of the applied methods are shown in Figure 8. It is well known that the release of most elements is pH-dependent, e.g. calcium, and that for others it is not, e.g. sulphur. But even if only the results of the methods with pH values lying in the *in situ* range (DEV S4, SSE, pH-stat test at pH 7, centrifugation, dialysis) are regarded, the difference of the concentrations might be 100%.

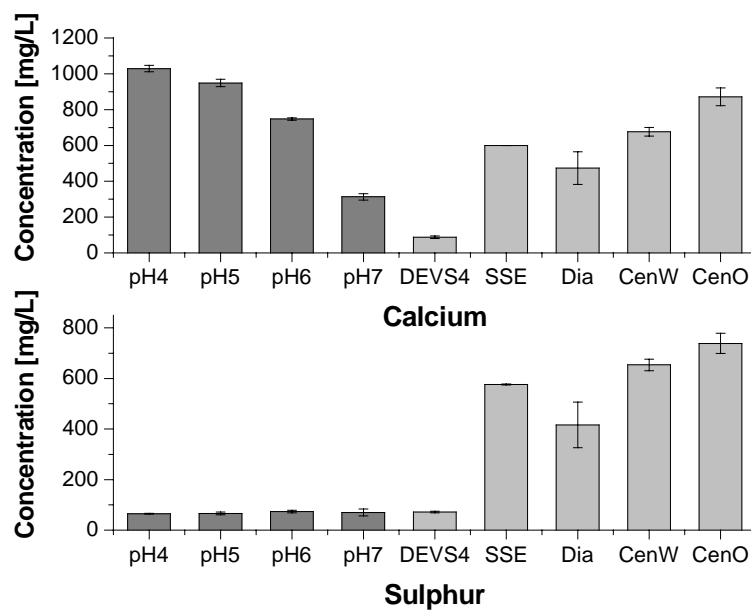


Figure 8: Concentration of calcium and sulphur in various leaching experiments including the fine fraction of the harbour sediments in comparison with its pore water concentration. Index: pH4, 5, 6, 7: pH-stat test with various pH values; Dia: dialysis; CenW: centrifugation of the fine fraction of harbour sediments used in the dialysis; CenO: centrifugation of the original fine fraction of harbour sediments

The liquid/solid ratio (L/S) is another important parameter (Fig. 8) for comparing the concentrations of calcium and sulphur derived from the DEV S4 test (L/S 10 L/kg) with those from the soil saturation extract (L/S 1 L/kg), dialysis (L/S 0.45 L/kg), and centrifugation (L/S 0.34 L/kg). Upon applying the DEV S4 test, the concentration of both elements is ten times lower than in the pore water samples extracted by centrifugation of the original harbour sediment fine fraction. One way to regard the liquid/solid ratio is to express the source strength not in terms of concentrations, but as leached amount based on the dry mass of the material (g/kg dry weight).

In order to calculate the probability of occurrence of certain pH and E_H conditions, and thus the concentrations of certain contaminants, either the acid (ANC) or the base (BNC) neutralisation capacity (van der Sloot et al., 1997) as well as the oxidation (OXC) and the reduction (REC) capacity (Heron et al., 1994a, b) must be known. The ANC_{24} or BNC_{24} can be calculated by inserting the volume (V_t) of either acid or base consumed in the pH-stat test, the concentration (c) of the acid or base, and the mass (m) of the solid material into the following equation:

$$ANC_{24} / BNC_{24} \left[\frac{meq}{kg} \right] = \frac{V_t [L] \cdot c \left[\frac{eq}{L} \right] \cdot 1000}{m [kg]}$$

The ANC_{24}/BNC_{24} can be described as a proxy for the variation of the source strength over time. Based on the ANC, the time period for a soil horizon with a certain thickness to be entirely acidified can be estimated (Hamer and Karius, 2002). The volumes of water needed to consume the buffer capacity (V_{ANC}) can be estimated by regarding the thickness of the sediment layer (s), the compactness (ρ), the H^+ activity of the infiltrating water ($[H^+]_{inf. water}$) volume and the ANC_{24} :

$$V_{ANC} \left[\frac{L}{m^2} \right] = \frac{ANC \left[\frac{mmol}{kg} \right] \cdot \rho \left[\frac{kg}{m^3} \right] \cdot s [m]}{H^+_{inf. water} \left[\frac{mmol}{L} \right]}$$

Divided by an annual infiltration rate (L), the time period (t) for total acidification to take place is obtained by:

$$t[a] = \frac{V_{ANC} \left[\frac{L}{m^2} \right]}{I \left[\frac{L}{a \cdot m} \right]}$$

Table 4 presents a list including ANC_{24} , V_{ANC} and t for the various materials employed, as well as the liquid/solid ratio resulting from infiltration, which were calculated by dividing the V_{ANC} by the compactness and the thickness of the individual horizon regarded.

Tab. 4: Time period until the ANC is consumed in a horizon of a thickness of 0.1 m and a compactness of 1.6 g/cm³ (average compactness for soils (Scheffer and Schachtschabel, 1998) by infiltration of water at pH 4

Material	ANC ₂₄ [meq/kg]	Infiltration until consumption of the ANC ₂₄ : V _{ANC} [L/m ²]	L/S [L/kg]	Time until pH 4 is reached [a]	
				Infiltration rate 300 [mm]	Infiltration rate 50 [mm]
Sandy fraction of harbour sediment	90	144000	900	480	
Fine fraction of harbour sediments	620	992000	6200		19800
MWIA	1170	1872000	11700	6250	
Demolition waste	1600	2560000	16000	8500	

The concentrations measured in the pH-stat tests are not released spontaneously as soon as pH 4 is reached in the seepage water. During the consumption of the buffer capacity in the soil horizon, a steadily increasing discharge of the contaminants will occur with decreasing pH values. However, as Hamer and Karius (2002) stated, this model to calculate ANC consumption represents a simplification of the acidification process. The leachate concentration in the field, i.e. the source strength is influenced by a number of other parameters, such as redox potential, liquid/solid ratio, ionic strength, presence of complexants and whether a monolithic or granular material is dealt with (van der Sloot, 1998).

The development of E_H conditions in a certain soil horizon, i.e. the prediction of the period required for complete oxidation or reduction of a layer can be made on the basis of the oxidation (OXC) and the reduction (REC) capacity. Heron et al. (1994b) compared different experimental methods to determine the OXC in relation to iron and manganese oxides and hydroxides. But, regarding the application to different materials, the results are quite inconsistent (Veehmayer, 2000).

OXC and REC are like the ANC and BNC parameters to describe the potential variations of the source strength over time in case of a change in the E_H or pH environment. Analogous to

estimating the capacity of potential acidification, the oxidation (OXC) and the reduction (REC) capacity can be calculated on the basis of the potential electron transfers (Bourg and Loch, 1995):

$$\text{OXC} = 4[\text{O}_2] + 5[\text{NO}_3] + 2[\text{Mn(IV)}] + [\text{Fe(III)}] + 8[\text{SO}_4^{2-}] + 4[\text{TOC}]$$

$$\text{REC} = 8[\text{NH}_4^+] + [\text{Fe(II)}] + 2[\text{Mn(II)}] + 8[\text{S(-II)}] + 4[\text{TOC}]$$

The main contributors to the OXC in a soil or sediment sample with a low liquid/solid ratio are iron oxyhydroxides, organic matter (if reducible), and, to a lesser extent, manganese oxides (Bourg and Loch 1995). Data required for the calculation of OXC and REC are not available yet.

In order to reflect the natural conditions and processes, i.e. to simulate natural reaction and transport conditions, laboratory- scale column experiments with sandy materials and diffusion tests (dialysis) with clayey soils can be devised, thus allowing for a verification of the results obtained from the batch experiments and the calculations mentioned above.

The main requirements of the methods applied in this project to determine the source strength need to be valid, practicable, and generally accepted (Oberacker and Eberle, 2002). In addition, a good reproducibility is necessary for the validity and the informative value of the individual methods. The method that fulfils all these requirements, and takes the natural conditions regarding the pH and E_H environment and the liquid/solid ratio into account, is the pore water analysis. This method produces results that are closest to the leachate concentration under natural conditions, i.e. the source strength. Further investigations are necessary in order to evaluate the validity of the various batch experiments, if pore water sampling is either not possible or pore water is not available, e.g. in case of demolition waste or municipal waste incineration ash.

2.2. Methodical comparison of the saturated and unsaturated column tests using the example of the sandy fraction of harbour sediments

In the following results of the saturated and unsaturated column tests with the sandy fraction of harbour sediments are presented with respect to the comparability of the two methods. Apart from the water saturation and the flow regime the two columns also differ in dimensions. But, although the hydraulic regimes are very different in the two methods, only silicon shows a different development of the concentrations in the outlet samples of saturated and unsaturated column. The wider distribution of E_H values and concentrations of organic carbon in the unsaturated column are due to the intrusion of oxygen. In the flow-path sampling of the saturated column the development of a reaction front is observable that is moving through the column with time. Due to the fact that the water saturation is inhomogeneous within the unsaturated column, meaning an increase along the seepage-path, the sampling with porous soil moisture sampler was only possible in the second half of the column. Thus no information can be given about the development of such a potential reaction front in the unsaturated column.

Hydraulic Regime

6 mL/h of the artificial acidic rainwater were constantly pumped into the saturated column, while the precipitation of the unsaturated column was 50 mL/h·d. The Darcy velocity in the saturated column was 8 cm/d, the total porosity 31 %. A coefficient of hydraulic conductivity (k_f) of $8.3 \cdot 10^{-5}$ was calculated according to Beyer (1964). By means of the first flush phenomenon the exchange of one water-saturated pore volume could be estimated. The initial strong leaching ended after approximately 1.8 days, leading to the presumption that the water-saturated pore volume was exchanged once in 1.8 days. With a length of the column of 48 cm this led to an effective velocity of 26 cm/d and an effective porosity of 30 %. The water saturation of the column was 99.8 %.

Table 1: Properties of the column tests

	Length	Diameter	Dry material incorporated	Flow conditions	Effective flow/seepage velocity	Saturation
Saturated column	48 cm	5 cm	1.7 kg	Bottom to top	8 cm/d	99.8 %
Unsaturated column	48 cm	10 cm	5.26 kg	Precipitation	3.4 cm/d	60 %

In contrast, in the unsaturated column the total porosity was 40 %. The seepage velocity in the unsaturated column could be estimated in the line of the saturated column, by means of the exchanged pore volume. This led to an effective seepage velocity of 3.4 cm/d. The water saturation was 60 % in average, though a distinct increase in the saturation was observable with depth. But, due to the incorporated filter quartz sand saturation was not reached. Similar effects were observed in unsaturated column experiments with overburden materials, where the air filled porosity (total porosity 44 %) decreased from 0.2 to 0.08 within 80 cm of the column (Hecht and Kölling, 2002).

Outlet samples

The leaching in the initial state of the unsaturated column test was mainly affected by the low pH value (Fig. 1), leading to an enhanced mobilisation of most elements. The E_H values showed a wider distribution in the unsaturated than in the saturated column. This is due to the varying intrusion of oxygen in the unsaturated column.

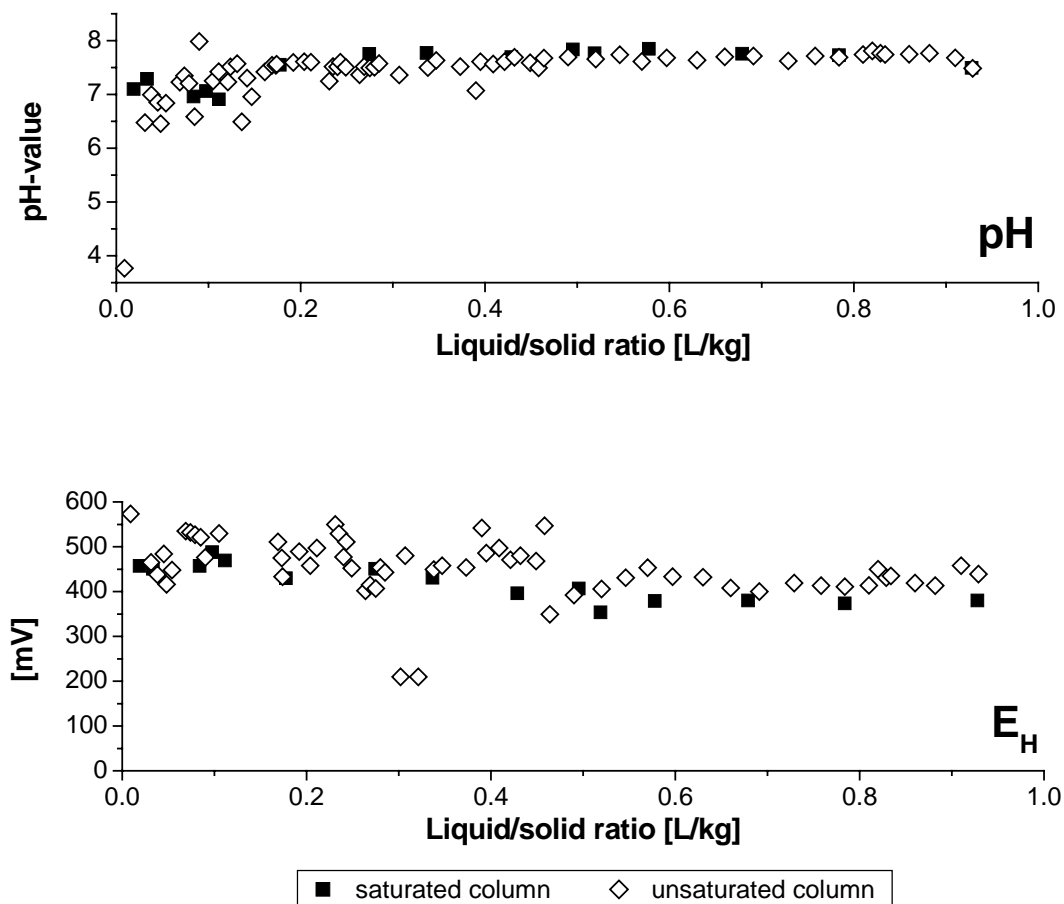


Figure 1: pH and E_H value in the saturated and unsaturated column

The DOC showed the same effect as the E_H value. But, the mean concentrations were higher in the unsaturated than in the saturated column (Fig. 2). Because of the subsequent delivery and intrusion of airborne oxygen under unsaturated conditions the degradation of organic matter was higher compared to saturated conditions leading to a high amount of dissolved organic matter (Guderian, 1999).

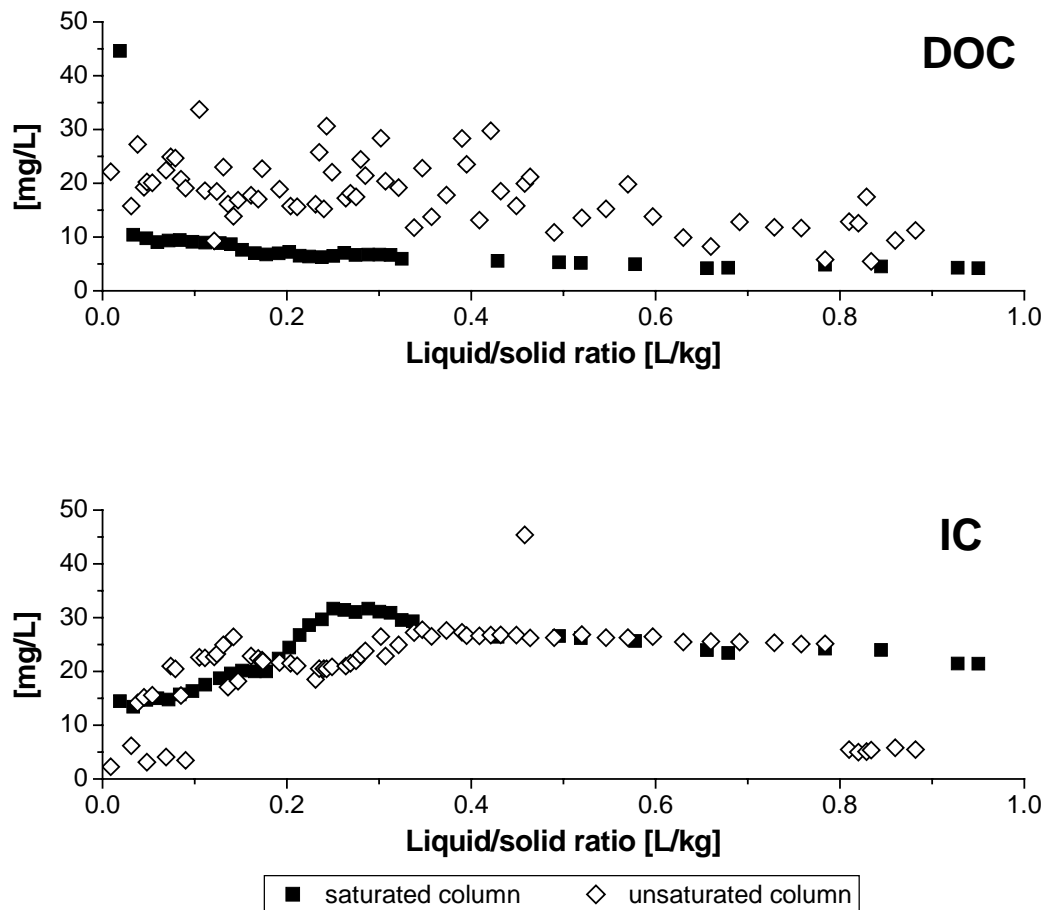


Figure 2: Concentration of dissolved organic carbon (DOC) and inorganic carbon (IC) in the saturated and unsaturated column

The water saturation was not homogeneous within the unsaturated column, it increased along the seepage-path from top to bottom. The withdrawal of a sufficient amount of sample with porous soil moisture sampler was only possible after a seepage-path of 26 centimetres. The inhomogeneous distribution of water also becomes apparent accounting for the oxygen saturation within the column. It was measured with an optical oxygen sensor (optode) developed and constructed by Hecht and Kölling (2001). The oxygen saturation decreased with depth according to the increasing water saturation (Tab. 1).

Table 1: Oxygen saturation in the unsaturated column

Seepage path [cm]	O ₂ -saturation %
9	24
25	23
37	23
49	19

With the exception of aluminium and silicon all main elements showed the same leaching behaviour. After a first flush a constant discharge until a liquid/solid ratio of 0.3 L/kg could be observed, after which the leaching decreased strongly (Fig. 3). In addition, good agreement was found between the two column tests, with the same exceptions of aluminium and silicon.

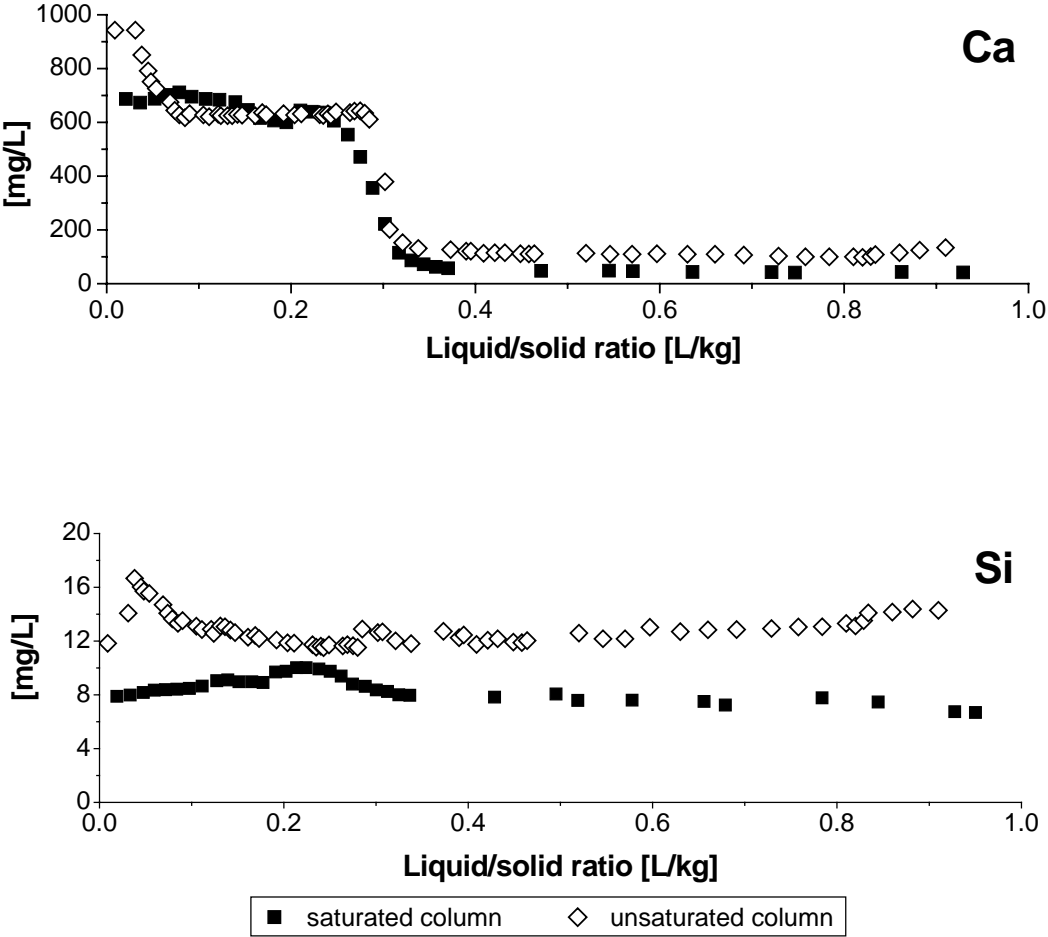


Figure 3: Concentration of calcium and silicon in the saturated and unsaturated column

Calculations of saturation indices with PHREEQC (version 2.8) (Parkhurst and Appelo, 1999) indicated that a quasi thermodynamic equilibrium evolved after the initial leaching until a liquid/solid ratio of 0.08 L/kg, as plagioclase, quartz and calcite showed saturation indices around zero (Fig. 4).

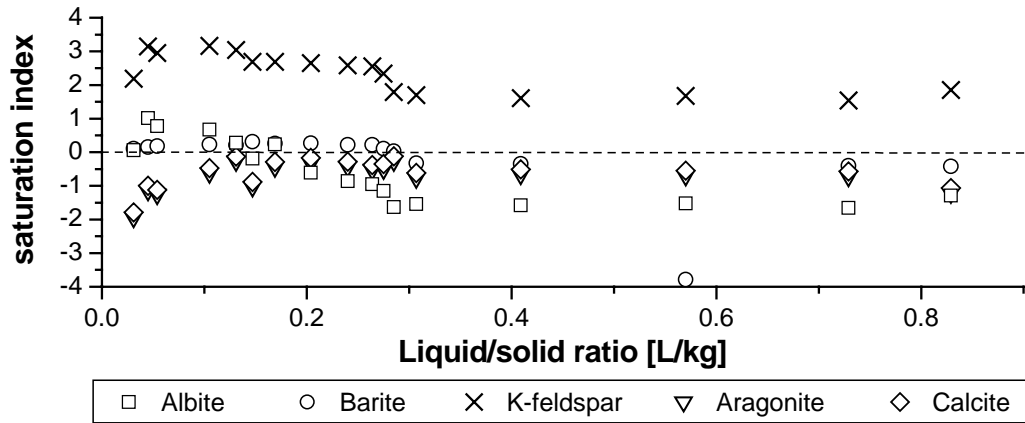


Figure 4: Calculated saturation indices of various phases in the outlet samples of the saturated column

Flow-path sampling

In Figure 4 the pH values and the concentrations of barium are displayed in the samples gained by two flow-path samplings in the saturated column after 90 and 282 days and one in the seepage-path sampling of the unsaturated column after 24 days. After 282 days of constant leaching a narrow zone evolved where the pH value increased from 4.2 to 7.8. This front moves according to the buffer capacity of the material (Karius et al., 2002). Along this reaction front dissolution reactions related to the low pH occur. Further down the flow-path at higher pH values phases can re-precipitate (Fig. 5 Ba).

Only one seepage-path sampling was conducted of the unsaturated column after 24 days. This time period is not sufficient for the visible development of a reaction front. Considering that only in the second flow-path sampling of the saturated column after 282 days at a liquid/solid ratio of 21 L/kg the reaction front was observable, it would take a couple of years until a front would have developed in the unsaturated column. Additionally, methodical limitations make it difficult to observe this phenomenon in the unsaturated column. Sampling was only possible in the second half of the column due to the inhomogeneous distribution of the water saturation. Thus no information can be given about the development of a reaction front in the unsaturated column.

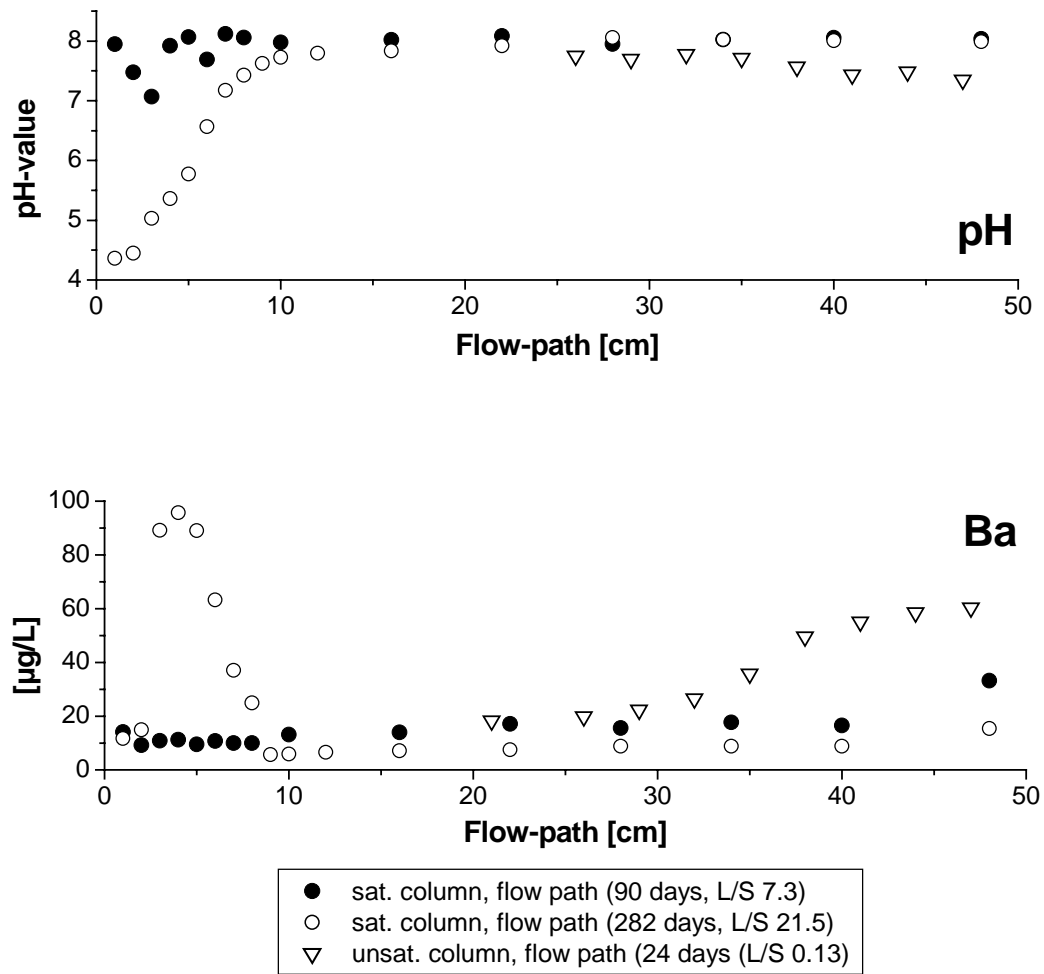


Figure 5: pH value and concentration of barium in the two flow-path sampling of the saturated column

3. Results

3.1. Mobility of heavy metals in harbour sediments - an environmental aspect for the reuse of contaminated dredged sediments

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Mobility of heavy metals in harbour sediments - an environmental aspect for the reuse of contaminated dredged sediments

Tanja Lager, Kay Hamer and Horst D. Schulz

Abstract

Different methods to determine the source strength of two harbour sediment fractions were applied and are discussed with a focus on As, Cd, Cr, Co, Cu, Mo, Ni, Pb, and Zn. Four different batch experiments were performed with both the sandy and the silty fractions of harbour sediments sampled at the disposal site for dredged material in Bremen Seehausen, North Germany. In addition to the batch experiments a modified centrifugation tube was used in order to obtain pore water from the unsaturated silty fraction. Column experiments were run with the sandy fraction of the material under saturated and unsaturated conditions, irrigated with artificial acid rain water. The results show that the legal threshold value applying to Mo and Pb is not exceeded in any of the methods applied to the sandy or the silty fractions. As for the sandy fraction, Cr and Co were also below this limit. Cd, Ni, Cu and Zn exceeded the threshold values in some of the batch experiments applied to both fractions, depending on the pH value and the elution agent used in the experiment. All results obtained from saturated column leaching were below the respective threshold values; however, it should be noted that the pH was between 7 and 8 throughout the entire experiment. This pH also applied to the unsaturated column, with the exception of the 'first flush'. This first sample had a pH value of 3.8, which was due to sulphide oxidation at the beginning of the experiment and led to strong leaching of all elements under study. As a consequence, arsenic, Cu and Ni values exceeded their respective threshold values.

Keywords: harbour sludge, contaminated dredged material, heavy metals, batch experiment, column experiment

Introduction

The sediments of harbours and waterways have to be dredged to maintain the water depth for shipping and naval traffic. In the Netherlands as well as in Germany, the responsible authorities have to deal with 30-50 million m³ annually. Due to contaminants some fractions of the dredged

material cannot be relocated to the river, the estuary or the sea, but have to be treated or deposited on land. In Germany, annually 2-3 million m³ are deposited on land each year.

The sediment management concept in Bremen is based on three main principles: Firstly, to identify the sources of pollution in the catchment area of the Weser River and reduce sedimentation in the harbour (Nasner, 1992) in order to minimise the amount of sediment which has to be managed; secondly, to deposit the material if environmentally justifiable (Biener et al., 1999); and thirdly, to investigate and compare treatment techniques as potential alternatives to land disposal. Technologies discussed are subaqueous disposal (Jacobs and Förstner, 1999), brick production (Hamer and Karius, 2002), the use as containment or cover layer in landfills (von Lührte et al., 1996; Tresselt et al., 1998), and the production of light-weight aggregates (Derman and Schlieper, 1999; Hamer et al., 2003).

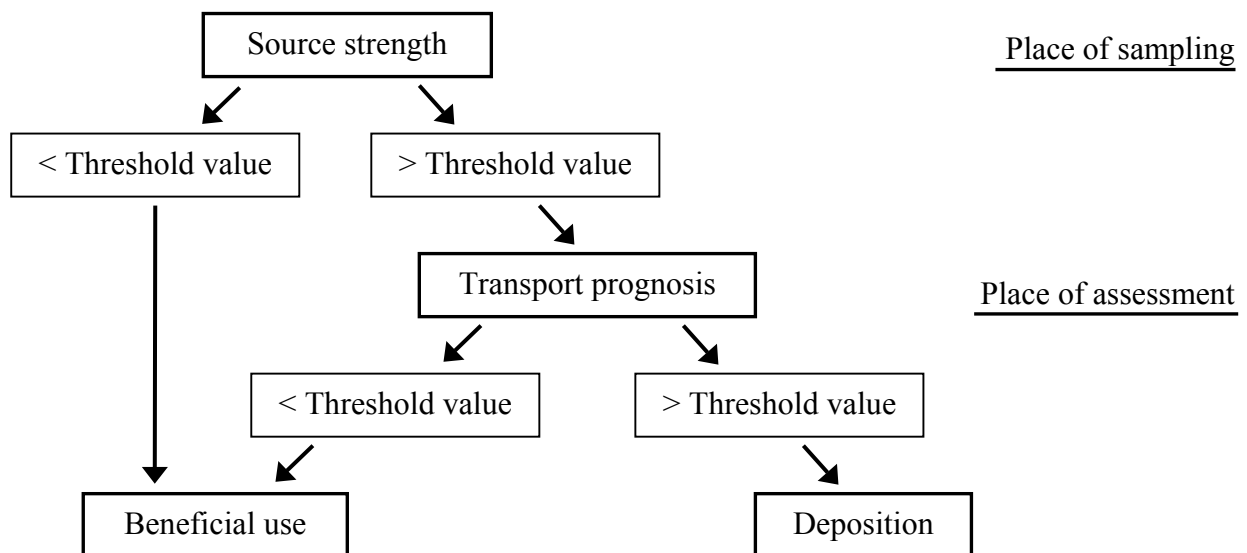


Figure 1: Beneficial use versus deposition: Process design as described in the German Federal Soil Protection Act (Federal Bulletin, 1998)

According to German law (Federal Bulletin, 1998) such reuse of dredged material requires an environmental assessment, which considers the mobility of pollutants. The investigation procedure according to the Federal German Soil Protection and Contaminated Site Ordinance (Federal Bulletin, 1999) considers both bulk and eluate concentrations. If the bulk concentrations allow a reuse of the sediments, the focus will lie on the mobility of the pollutants measured in various eluate tests. Within this context, the so-called "source-strength" is important. The Federal German Soil Protection Act (Federal Bulletin, 1998) defines the source strength as the total mass flow caused by natural leaching. If the threshold value defined for various inorganic and organic contaminants (Tab. 4) is exceeded, a transport prognosis shall have to be made. The aim of this prognosis is to ascertain that the concentration leached from the material under

investigation by natural precipitation does not exceed threshold values on its way into the groundwater. Ultimately, this prognosis leads to a calculated value of the contaminant concentration at the boundary between the vadose and the saturated zone. This interface between vadose and saturated zone is the 'place of assessment'. If threshold values are not exceeded in the place of assessment, reuse of dredged material will be possible (Fig. 1) (Oberacker and Eberle, 2003; Lager et al., 2003).

The seepage water is exposed to a variety of chemical and physical processes on its way through the contaminated deposit and the underlying soil to the boundary between the vadose and the saturated zones. The concentration of the seepage water at this point is strongly dependent on the type of contaminant and the composition and texture of both the regarded recycling material and the subsoil. On their way through the underground, the contaminants can be transformed or immobilised, and organic substances might become decomposed (van der Sloot et al., 1997; Scheffer and Schachtschabel, 1998). These processes can lead either to a delayed discharge or a reduction of the concentration of contaminants (Wild, 1993). Focussing on inorganic contaminants, the most important processes are sorption and precipitation (van der Sloot et al., 1997). Both strongly depend on the conditions of pH and E_H prevalent in the unsaturated zone. Therefore, it is crucial to take these conditions inclusive of their temporal development into account upon selecting the adequate method to determine the source strength.

The results of the different methods will be discussed with respect to the suitability of the respective methods for the determination of the source strength. Furthermore, conclusions will be drawn specifying the circumstances under which a reuse of the sandy and the silty fraction of the harbour sediments could be possible, considering soil and groundwater quality.

Material

The sediment samples investigated were collected at the disposal deposit site for dredged material located at Bremen Seehausen, northern Germany. Here, the sediment material dredged from the harbour basins of Bremen and Bremerhaven is deposited after a dewatering process of about one year. This is done by dredging ships pumping the material via pipelines onto drainage fields (Biener et al., 1999). A grain-size separation of the former dredged sediments takes place in the course of this field flushing process. The sandy fraction accumulates close to the outlet of the pipeline (von Lürthe et al., 1996). Aggregates of clayey material are found in the sandy fraction, whereas clayey, slightly sandy silt deposits are dispersed throughout the rest of the field.

The accumulated material was sampled by taking 20-30 kg of both fractions at five different locations. As far as the homogeneity the analysis of the elements is concerned calcium, potassium, magnesium, sodium, strontium, and zinc in the pore water displayed deviations of less than 10 %. However, five samples of both fractions were each homogenised to a single sample and used in all subsequent investigations. The grain-size distribution of the sandy fraction and the silty fraction was determined according to DIN 18123 (1996) either by sieving or with an areometer, respectively (Fig. 2). E_H and pH values were determined *in situ*, all other parameters were performed in the laboratory (Tab. 3).

Methods

Various batch experiments were employed to investigate both grain-size fractions of the investigated material:

- German standard method DEV S4, a leaching experiment with deionised water (DIN, 1984)
- Ammonium-nitrate extract (DIN, 1997b)
- Soil saturation extract (Federal Bulletin, 1999)
- pH-stat test at pH 4, 5, 6, 7, and 11 (Obermann and Cremer, 1992; Karius and Hamer, 2001b)

Table1: Analysis of the artificial rainwater

Element	Unit	Concentration	Element	Unit	Concentration
Al	[$\mu\text{g/L}$]	3	Mn	[$\mu\text{g/L}$]	<5
Ba	[$\mu\text{g/L}$]	3	Mo	[$\mu\text{g/L}$]	<2
Ca	[mg/L]	3	Na	[mg/L]	1
Cd	[$\mu\text{g/L}$]	<2	Ni	[$\mu\text{g/L}$]	1
Co	[$\mu\text{g/L}$]	<2	S	[mg/L]	3
Cr	[$\mu\text{g/L}$]	<2	Si	[$\mu\text{g/L}$]	450
Cu	[$\mu\text{g/L}$]	13	Sr	[$\mu\text{g/L}$]	9
Fe	[$\mu\text{g/L}$]	3	V	[$\mu\text{g/L}$]	<10
K	[$\mu\text{g/L}$]	270	Zn	[$\mu\text{g/L}$]	24
Mg	[$\mu\text{g/L}$]	320			

The acid consumption of each pH-stat test performed at pH 4, 5, 6 and 7 was recorded within the 24- hour test and used to calculate the acid or base neutralisation capacity (ANC_{24}).

A modification of the centrifugation technique for gaining pore water from unsaturated material was applied to the fine fraction of the harbour sludge, while the sandy fraction was applied in column experiments conducted under saturated and unsaturated conditions. The saturated column was made of Perspex, measured 48 cm in length and 5 cm in diameter. The wet material weighing 1.82 kg was saturated with water upon being loaded into the column. The flow rate was set to 0.1 mL/min. The unsaturated column, also made of Perspex, measured 48 cm in length and 10 cm in diameter of 5.88 kg of the wet material were incorporated and slightly compacted. The column was irrigated once a day over an hour with 50 mL artificial rainwater applied through 6 precipitation cocks. Tap water was diluted with deionised water down to yield a water hardness of 0.0895 mmol/L. Acid rain was simulated by application of an average pH of 4.1 (Tab. 2).

Table 2 shows the test conditions and several factors controlling the release of contaminants in the methods applied. A detailed description of the methods can be found in Lager et al. (2003).

Table 2: Test conditions and some release controlling factors of the applied methods (modified after Lager et al. 2003)

Method	Grain-size	pH value	Liquid-solid ratio [L/kg] / flow conditions	Elution time	Elution agent	parallel tests
DEV S4 test	< 1 cm	dependent on material	10 / overhead shaker	24 h	deionised water	3
Soil saturation extract (SSE)	original	dependent on material	~1 / kneading and stirring for 15 min respectively	48 h	deionised water	3
Ammonium-nitrate extract	< 2 mm	dependent on material and NH ₄ NO ₃	4 / overhead shaker	3 h	1 M NH ₄ NO ₃	3
pH-stat test	< 2 mm	4, 5, 6, 7, 11	10 / stirrer	24 h	deionised water	3
Saturated column	< 2 mm	4	variable / 0.1 mL/min	weeks to month	artificial rainwater	1
Unsaturated column	< 2 mm	4	variable / 50 mL/day in 1 h	weeks to month	artificial rainwater	1
Centrifugation	original	dependent on material	adequate to natural / none	1 h	pore water	3

Bulk concentrations of the sediments were determined by total digestion induced by microwave heating (DIN, 1983). All samples were filtered through a 0.2 µm cellulose- acetate membrane filter and acidified with nitric acid for conservation. The concentration of lead was determined by GF-AAS measurement (Unicam Solaar 989QZ). The detection limit for lead was 4 µg/L. The

concentrations of the main elements were determined by ICP-OES measurement (Perkin Elmer, Optima 3000), the apparatus being connected to a cross-flow nebulizer. The concentrations of arsenic, cadmium, chromium, cobalt, copper, molybdenum, nickel and zinc were also obtained by ICP-OES measurement, here however, an ultra sonic nebulizer (CETAC USN) was connected. The detection limit of arsenic was 10 µg/L, 4 µg/L in case of cadmium, and 2 µg/L for chromium, cobalt, copper, molybdenum, nickel and zinc. Measuring the artificial rainwater the detection limits were lower for some trace elements (Tab. 1). This is due to the fact that some of the samples derived from the experiments had to be diluted. The ICP-OES connected with the ultra sonic nebulizer was not able to measure higher bulk contents than 1 g/L.

PHREEQC (version 2.8) (Parkhurst and Appelo, 1999) is a programme for speciation, saturation-index calculations, one-dimensional (1D) transport calculations and inverse modelling. It was used in this study to calculate the speciation and saturation-indices of copper in the batch experiments. The LLNL (Lawrence Livermore National Laboratory) dataset included was therefore complemented with the Gibbs free energies of the known copper amino complexes (Smith and Martell, 1976). In order to calculate the pyrite weathering reactions in the unsaturated column experiment, the MINTEQA2 dataset was employed.

Results

Material characterisation

The main sandy harbour-sediment fraction is composed of quartz. Minor constituents are potassium-feldspar, plagioclase, muscovite, and zircon. Quartz is also the major constituents of the fine fraction of harbour sediments. Minor components are potassium-sodium-feldspar, muscovite, clinocllore, calcite, and dolomite. The soil physical properties (Tab. 3, Fig. 2) of the fine fraction allow for its utilisation as building material in noise- protection embankments and dikes or an application as a mineral sealing agent in landfill surface sealing (von Lührte et al., 1999; Biener et al., 2003). The sandy material could be used for road construction or as landfill drainage sand.

Table 3: Material properties of the sandy and fine fraction of harbour sediments

	Sandy fraction	Fine Fraction
Water content [% weight] (DIN, 1989)	~ 5	~ 34
Specific weight [g/cm ³] (DIN, 1997a)	2.61	2.42
Specific surface area [m ² /g]	2.4	16.2
pH (<i>in situ</i>)	~ 8.3	7.0 – 7.6
E _H [mV] (<i>in situ</i>)	~ 250	30 – 420
El. conductivity [mS/cm]	1.5	0.54
Total carbon [%]	0.3	4.6
Inorganic carbon [%]	0.07	0.46
Organic carbon [%]	0.23	4.14

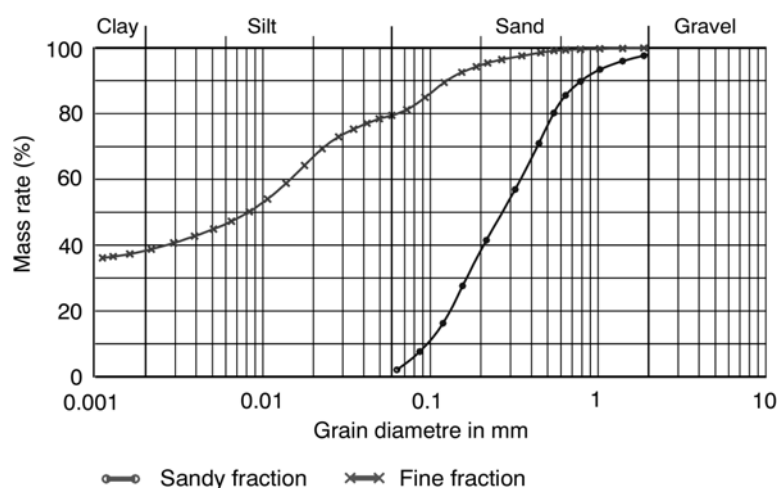


Figure 2: Grain- size distribution of the sandy and fine fractions of harbour sludge

In the following section, the concentrations of the heavy metals arsenic and molybdenum are compared with the threshold values as defined *inter alia* for the source strength by the Federal Soil Protection and Contaminated Sites Ordinance (Federal Bulletin, 1999) for each fraction of the harbour sediments.

Leaching tests

Sandy fraction

In none of the applied experiments with the sandy fraction of harbour sludge did cobalt, chromium (Fig. 3) and molybdenum exceed the threshold value of 50 $\mu\text{g/L}$, which is defined for these three elements. Neither did lead exceed the threshold value of 25 $\mu\text{g/L}$.

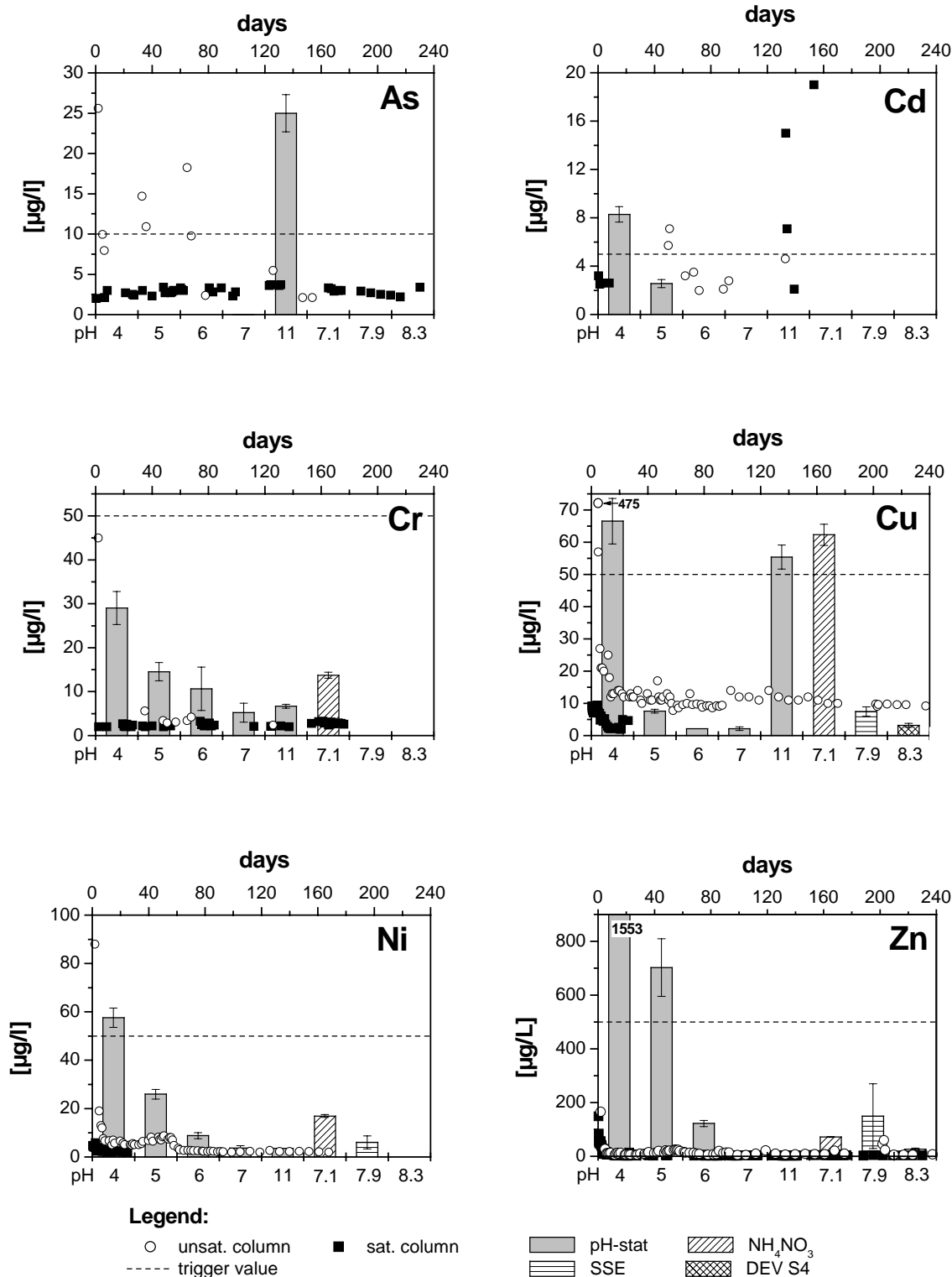


Figure 3: Concentrations of arsenic, cadmium, copper, nickel, and zinc in the sandy fraction of harbour sludge as determined by various batch experiments and at the outlet of the saturated and unsaturated column. The respective threshold values are shown (Federal Bulletin, 1999).

Cadmium, copper, nickel and zinc exceeded their respective threshold values (Fig. 3) in the pH4-stat test, zinc also in the pH5-stat test. Arsenic and copper showed strong leaching behaviour at pH 11 exceeding their respective threshold values. Enhanced leaching properties that led to

concentrations higher than the respective threshold value of copper could also be seen in the ammonium-nitrate extract.

Arsenic, copper and nickel exceeded the threshold value in the first flush of the unsaturated column. As for cadmium, only few values in the two column experiments were measured above the detection limit of 5 µg/L. Two concentrations in the saturated column distinctly exceeded the threshold value, one after 132 and the other after 151 days, whereas a concentration of 7 µg/L was reached in the unsaturated column after 50 days.

Fine fraction

The concentrations of lead and molybdenum were not found to be above their respective threshold values after application of all test methods (Tab. 4). An enhanced leaching at low pH values could be observed in the case of cadmium, cobalt, chromium, copper, nickel and zinc. Thus the respective threshold values of these elements were exceeded in the pH-stat test at low pH values (Tab. 4). Arsenic and copper also exceeded the threshold value at pH 11. In addition, the concentration of copper in pore water was higher than the threshold value. The same applied to cadmium in the ammonium-nitrate extract. Zinc showed very high concentrations in the pH-stat test at pH 4-6 which was a result of the very high bulk concentrations. The bulk concentrations were about six times higher than the highest background value measured in soils in Germany (Scheffer and Schachtschabel, 1998) (Tab. 5).

Table 4: Concentration of trace elements in the applied batch experiments, measured in the fine fraction of harbour sediments. The respective threshold values (Federal Bulletin, 1999) are shown for reasons of comparison in [µg/L]; concentrations underlined indicate that the threshold value was exceeded.

	As	Cd	Co	Cr	Cu	Mo	Ni	Pb	Zn
pH4-stat	<10.	<u>112</u>	<u>118</u>	<u>102</u>	<u>64</u>	<2	<u>381</u>	6.6	<u>21186</u>
pH5-stat	<10.	<u>35</u>	30	<u>53</u>	8.3	<2	<u>146</u>	<5	<u>7088</u>
pH6-stat	<10	<u>8.1</u>	<2	16	5.3	<2	33	<5	<u>1062</u>
pH7-stat	<10.	<4	<2	<2	4.8	<2	4.3	<5	68
pH11-stat	<u>170</u>	4.7	<2	8.0	<u>393</u>	3.3	29	<5	36
NH ₄ NO ₃ , pH 7.0	<10.	<u>6.0</u>	<2	7.0	40	<2	24	<5	164
SSE, pH 7.2	<10.	<4	<2	13	7.8	<2	8.9	<5	208
Cen, pH 7.3	<10	<4	<2	<2	<u>60</u>	<2	23	<5	299
DEV S4, pH 7.6	<10	<4	<2	<2	4.7	<2	2.3	4.6	79
Threshold value	10	5	50	50	50	50	50	25	500

Table 5: Bulk concentrations and background values determined in soils in Germany listed in Scheffer and Schachtschabel (1998) in [mg/kg]

	As	Cd	Co	Cr	Cu	Mo	Ni	Pb	Zn
Sandy fraction	1.4	0.9	4.2	11	9.3	1.5	<0.05	12	53
Fine fraction	12	4.1	20	90	65	1.5	41	75	490
Background values, soils	1-20	0.1-0.6	1-40	5-100	2-40	0.2-5	3-50	2-80	10-80

Discussion

Grain-size and soil mechanics:

Unlike former investigations (Hamer et al., 1995), the fine fraction of harbour sediments was found to be very homogeneous. The sandy fraction also displayed good homogeneity. Additionally, enough material is dredged annually to supply a construction project with building material. Homogeneity and capacity are important factors for the assessment of potential reuse and the substitution of primary resources. The beneficial use of harbour sediments has a lot of advantages. Primary resources like sand or clay can be saved and thus natural landscapes can. Secondly, the construction of new disposal sites including dewatering facilities, which consume space (Hamer and Karius, 2002) and are thus often accompanied with the destruction of natural landscapes, can be prevented. In addition to common deposition or relocation, silty dredged material could be used as a substitute raw material in the production of bricks (Hamer and Karius, 2002), be applied as a containment or cover layer in landfills (von Lührte et al., 1996; Tresselt et al., 1998), or used in the production of light weight aggregates (Derman and Schlieper, 1999; Hamer et al., 2003). A general overview of the current activities is given in Netzband et al. (2002).

Environmental aspects: mobility of heavy metals

Simple leaching tests

The release of most heavy metals increases with decreasing pH value (van der Sloot, 1996; Paschke et al., 1999; Karius and Hamer 2001a; van Herreweghe et al., 2002). Thus cadmium, copper, nickel and zinc exceeded their respective threshold values (Tab. 4, Fig. 3) in the eluates at pH values below 4. Arsenic and copper showed a strong leaching behaviour at pH 11 and exceeded their respective threshold limit values.

Arsenic(V) is the dominant dissolved arsenic-species in natural waters, i.e. under oxic and circum natural pH conditions. Arsenic(V) has a high adsorption affinity to oxides and hydroxides

of iron and aluminium (Langmuir et al., 1999; Sracek et al., 2004). Bowell (1994) found a maximum sorption of arsenic(V) on goethite and lepidocrochite at pH 6 and on hematite between pH 7-8. A mobilisation of arsenic in the acidic and alkaline pH range therefore mainly occurs by dissolution of the adsorbing matrix (Dankwarth and Gerth, 2002). The strong leaching of aluminium (1 mg/L) and iron (0.3 mg/L) in the pH- stat test at pH 11 as compared to the pH-stat test conducted in the neutral to slightly acidic range led to the mobilisation of formerly adsorbed arsenic.

A strong leaching effect of copper under alkaline conditions, which was probably due to the formation of hydroxo complexes, was also found by van Herreweghe et al. (2002). Copper is known to have a strong tendency to form hydroxo complexes (Tamura and Furuichi, 1997).

An enhanced leaching in the ammonium-nitrate extract was recognized for cadmium, chromium, copper, nickel and zinc. Cadmium and copper even exceeded the threshold values when this method was applied. Despite the fact that dissolved ammonium-nitrate has a pH of 5, the pH adjusted to 7.1 in the experiment including the sandy fraction of harbour sludge. Thus the enhanced leaching observed in comparison with the pH- stat experiment is not due to the pH value. In comparison with the pH-stat and the DEV S4 test, the liquid to solid ratio in the ammonium-nitrate extract was significantly lower (Tab. 2). However, if the leached amounts [in mg/kg] are compared, there is still an enhanced leaching effect to be observed in the case of copper. Thermodynamic modelling with PHREEQC (Parkhurst and Appelo, 1999) showed the formation of tetra- amino-complexes with copper ($[\text{Cu}(\text{NH}_3)_4]^{2+}$). Another reason why the concentrations measured in the ammonium-nitrate extract should be handled carefully as far as the validity of the source strength is concerned is that high ionic strength can lead to an increased retention of oxo-anion creators like arsenic and vanadium (Dankwarth and Gerth, 2002).

Breakthrough curves of column experiments

The breakthrough curves of all conducted column experiments showed a high initial concentration followed by a decrease. In general, this makes it obvious that an assessment based on simple leaching tests as previously discussed neglects plenty of information, which might be important. Consequently, column experiments seem to be essential for a detailed description of the leaching behaviour and thus for long- term risk assessment.

For example, the concentration of nickel in the unsaturated column was almost twice as high as the threshold value at the beginning of the leaching process that of copper was almost 10 times higher. The reason for the far higher values compared to those derived from the pH4-stat test is the even lower pH of 3.8 which prevailed in the initial state in the column.

The pH value at the outlet of both columns varied between 7 and 8, except for the first sample of the unsaturated column. The so-called ‘first flush’ showed a pH value of 3.8, which is even lower than the pH of the artificial rainwater used to irrigate the column. This is possibly due to iron(II)-oxidation, iron(III)-precipitation ($\text{Fe}^{2+} + 1/4\text{O}_2 + 3/2\text{H}_2\text{O} \Rightarrow \text{FeOOH} + 2\text{H}^+$) and the formation of sulphurous acid ($2\text{S}^- + 3.5\text{O}_2 + \text{H}_2\text{O} \Rightarrow 2\text{SO}_4^{2-} + 2\text{H}^+$). To verify this hypothesis, thermodynamic calculation were performed with the model PHREEQC (version 2) (Parkhurst and Appelo, 1999). The conditions of pH and E_H of the initial solution (pure water) corresponded to the conditions in the first flush of the unsaturated column (pH 4.1, pe 10.232). The phases $\text{Fe}(\text{OH})_3$ and kaolinite were allowed to react to equilibrium (saturation index equal to 0). O_2 and CO_2 partial pressures were maintained at $10^{-0.67}$ and $10^{-3.4}$ (atmospheric partial pressure). The weathering of 2.7 mmol pyrite resulted in a solution with a pH of 3.3. Buffering reactions with calcite were included in a second step. A pH of 3.8 was reached after adding 4.25 mmol calcite.

This phenomenon does not occur in the saturated column. Here, less O_2 is present and available due to the water-saturated conditions.

Table 6: Acid and base neutralisation capacities calculated from the acid or base consumption in the pH-stat tests

	ANC ₂₄			
	pH4	pH5	pH6	pH7
Sandy fraction	91	50	21	17
Fine fraction	624	525	375	137

Considering how sensitive the development of the pH is for the potential mobilisation of inorganic contaminants, the ANC should be accounted for. The ANC₂₄ against infiltration of water with a pH of 4 was 624 meq/kg in the fine fraction, and only ~91 meq/kg in the sandy fraction (Tab. 6). Thus, in case of the fine fraction, it takes much longer until the ANC is consumed in a horizon of material, and consequently until the material is fully acidified and the enhanced leaching of heavy metals commences (Lager et al., 2003). Anticipating an annual infiltration rate of 300 mm of water at pH of 4, it will take less than 500 years until a horizon of 0.1 m of sandy fraction is fully acidified by means of acidic rain- water infiltration. Under the assumption of an infiltration rate of 50 mm/a, complete acidification of the fine fraction will be due after about 20,000 years (Lager et al., 2003).

Conclusion and outlook

The effects that various elution agents may have on the mobilisation of contaminants have to be taken into account when a method for the determination of the source strength is to be defined. Ammonium-nitrate was shown to enhance the leaching of copper due to the formation of amino-complexes. In addition ammonium-nitrate, as compared to deionised water, which was used as the elution agent in all other batch experiments, is known to decrease the mobility of arsenic and vanadium.

With the exception of molybdenum and lead all other elements exceeded their threshold value in all applied methods. This is mainly depended on the pH value. An enhanced leaching at decreasing pH applied to all heavy metals. Arsenic and copper revealed an increased mobility at pH 11 as well. Arsenic is mainly adsorbed to oxides and hydroxides of aluminium and iron. The enhanced leaching of aluminium and iron in the pH-stat test at pH 11 therefore produced enhanced leaching of arsenic. Copper is more mobile at high pH values, probably due to the formation of hydroxo- complexes. These pH-dependent mobilisation mechanisms can be interpreted from the pH-stat tests. Furthermore the ANC contributes to develop a time frame applying to the acidification of a horizon.

The observed strong and rapid increase of the concentrations in the breakthrough of the column experiments, the so-called 'first flush', a phenomenon that also occurs under natural conditions of deposition, can only be revealed in a column test. It was shown that leaching in the first flush will be even potentially stronger under unsaturated conditions, if sulphide oxidation occurs and results in a decrease of pH.

A similar approach consisting of a combination of pH-stat and column test should be applied to organic contaminants, e.g. TBT. There are attempts within the BMBF –project to design a column test suitable for the examination of both inorganic and organic contaminants (e.g. North Rhine-Westphalian State Environment Agency (LUA NRW) in Essen). However, a sample volume of 1 L is necessary in order to measure organic substances in a range of 10-100 ng/L. The analytical requirement of huge sample volumes leads to a decrease in time resolution in the breakthrough curves. Consequently, the first flush phenomenon is not easy to observe.

It is clear that a reuse of both fractions of harbour sediments, i.e. the silty sand and the clayey, slightly sandy silt fractions, is only possible under conditions, in which the pH-dependent degradation can be retarded or prevented. Considering the ANC₂₄ and the various transport mechanisms within the two materials, it becomes obvious that the precautionary measure should be even more stringent for the sandy fraction as compared to the fine fraction. A possible

scenario for the reuse of the fine fraction could consist in the substitution of natural clays in cover systems for wastes under an overlying soil layer (Tresselt et al., 1998). As previously shown, the buffer capacity of the fine fraction itself will last for about 20,000 years, anticipating an infiltration with rain at pH 4. Assuming preferential flow mechanisms to be negligible, diffusion will be the main transport mechanism within such a deposition of the fine fraction of harbour sludge, whereas advection will predominate in the sandy fraction.

The seepage water prognosis is defined as the estimation of the concentration of pollutants that will reach the groundwater together with the leachate in the foreseeable future. Including this period in the risk assessment, the risk of polluting the underlying groundwater is much higher underneath a deposition of the sandy fraction. A potential scenario of reuse could consist in its implementation as a core in dikes, a situation in which advection is rather irrelevant, because such core would be enclosed in a very fine-grained material displaying a hydraulic conductivity in a range of less than 10^{-8} m/s (von Lührte, 1997). With such an overlying fine-fractioned material the surface discharge will even be enhanced. A layer consisting of the same material and situated underneath the embankment would inhibit the discharge into the underlying soil.

Next to the bulk concentration and the leachable concentration of the contaminants, the development of the pH conditions, the buffer capacity of the material, and the flow conditions in a deposition of this material need to be considered in reuse scenarios.

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3.2. Comparison of Leaching Tests to Quantify the Release of Inorganic Compounds from Demolition Waste

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Comparison of Leaching Tests to Determine and Quantify the Release of Inorganic Contaminants in Demolition Waste

Markus Delay, Tanja Lager, Horst D. Schulz and Fritz. H. Frimmel

Abstract

The changes in waste management policy caused by the massive generation of waste materials (e.g. construction and demolition waste material, municipal waste incineration products) led to an increased reuse and recycling of waste materials. For environmental risk assessments, test procedures are necessary to examine waste materials before they can be reused. In this paper, results of column and lysimeter leaching tests having been applied to inorganic compounds in a reference demolition waste material are presented. The work is part of the research project “Seepage Water Prognosis” funded by the Federal German Ministry for Research and Education (BMBF).

The results show a good agreement between the leaching behaviour determined with the lysimeter unit and the column units used in the laboratory. In face of less time and system requirements compared to lysimeter systems, laboratory column units can be considered as a practicable instrument to assess the time-dependent release of inorganic compounds under conditions similar to those encountered in a natural environment. The high concentrations of elements in the seepage water at the initial stage of elution are reflected by the laboratory column leaching tests. Especially authorities or laboratories might benefit and have an easy- to -use and inexpensive, but nevertheless reliable method to serve as the basis for decision-making.

Keywords: demolition waste material, leaching test, column leaching test, inorganic pollutants

Introduction

The increasing accumulation of waste and its management presents a worldwide problem. For example, the generation of construction and demolition waste (including road construction and building-site waste), and soil excavations in Germany amounted to about 230 million tonnes in 1997 (German Federal Environmental Agency, 2002), and can therefore be seen as major part of industrial waste (Tränkler et al., 1996).

This massive generation of waste materials led to changes in waste management policy. According to the German Recycling Management and Waste Act (1994), waste has to be avoided as much as possible, at least material recycling and reuse should have priority over deposition. For example, demolition waste materials can be used in road construction (Bilitewski et al., 1990; Winkler, 2001). Of course, the environmental risk caused by the reuse should be kept as low as possible. Therefore, it is necessary to previously assess the waste materials with regard to their release of organic and inorganic pollutants. The required test procedures should be practicable for authorities and laboratories on the one hand, but also reflect possible scenarios under natural deposition conditions on the other.

In the last decades, numerous leaching tests have been developed and compared in the scope of several national and international research programmes and projects, for example the Energy Research Centre of the Netherlands (ECN) (see van der Sloot et al., 1997; van der Sloot et al. 2003; van der Sloot, 1996), the Florida Centre for Solid and Hazardous Waste Management (Townsend et al., 2003), and the Washington State Department of Ecology (2003).

The Federal German Soil Protection and Contaminated Sites Ordinance (Federal Bulletin, 1999) offers different methods for the investigation of soils, soil material, and materials like demolition waste. As part of the research project “Seepage Water Prognosis” funded by the German Ministry for Research and Education (BMBF), column leaching tests were developed and performed. One of the aims of this project was to determine the source strength of porous waste materials as regards to inorganic pollutants. The source strength is defined as the total mass flow caused by natural leaching (Oberacker and Eberle, 2002).

Here, results of column leaching tests applied to a reference demolition waste material are presented. The results of two project working groups (Prof. Schulz/Bremen and Prof. Frimmel/Karlsruhe) are compared to lysimeter data that have already been published by the North Rhine-Westphalian State Environment Agency (LUA NRW) (Susset et al., 2004).

In addition, the results of the column leaching tests are compared with a common batch leaching test according to DIN 38414-4 (S4) (DIN, 1984) and soil saturation extract (SSE) according to the German Federal Soil Protection and Contaminated Sites Ordinance (Federal Bulletin, 1999).

It will be discussed whether the leaching behaviour of demolition waste material in laboratory column leaching tests shows similarities with large scale field lysimeter leaching tests. In this case, laboratory column units could be seen as an easy- to- use, practicable instrument to assess the source strength of waste materials under natural conditions.

Experimental

Material

In the column leaching tests, a reference material generated by the German Federal Institute for Materials Research (BAM) was employed. Selected material properties are given in Table 1. The reference material consists of a mixture of various components typical of a demolition waste material with a maximum grain-size of 4 mm.

Table 1: Selected properties of the reference demolition material used in the leaching tests (Berger et al., 2004).

Parameter	Dimension	
Residual humidity	Weight-%	2.4
pH value (H ₂ O) ¹	-	11.9
Grain-size distribution		
6.3 - 4 mm	Weight-%	0.5
4 - 2 mm	Weight-%	14.9
2 - 0.63 mm	Weight-%	28.9
0.63 - 0.2 mm	Weight-%	36.4
0.2 - 0.063 mm	Weight-%	17.4
< 0.063 mm	Weight-%	1.9
Element content in solid ²		
Cr	mg/kg dried matter	16.3
Cu	mg/kg dried matter	29.2

¹Eluate with liquid/solid ratio 2:1 L/kg.

²Determined by means of Aqua Regia digestion.

Column leaching tests, sampling and sample preparation

Table 2 gives an overview of the properties of the different column leaching units used and regarded in this study. Further details relating to column A are reported in Lager et al. (2003). Detailed information about the column leaching unit used for column B (3 parallel column leaching tests) are given in Delay et al. (2003).

The results obtained in the two column leaching units were compared with lysimeter data from the North Rhine-Westphalian State Environment Agency (LUA NRW) (Susset et al., 2004). The lysimeter consisted of a cover layer (90 cm of quartz gravel, 2342 kg), the demolition waste material layer (45 cm, 1266 kg), and a filter layer (10 cm of quartz gravel, 260 kg). A liquid/solid ratio of about 0.6 L/kg was reached after 400 days. The liquid/solid ratio is the

quotient of the total mass of effluent that passed the column at a particular time and the total mass of dry solid used in the leaching test. The time in which samples were collected for this study was the period from the end of December 2002 until the beginning of January 2004.

Table 2: Properties of the column leaching systems regarded.

	Column dimensions Length, Diameter	Mass of solid in the column	Average flow velocity	Flow direction, Water saturation	Eluent, pH value
Column A	0.48 m, 5 cm	1.7 kg	0.08 m/d	Upstream, Water-saturated	Artificial rainwater, 4
Column B	0.24 m, 4 cm	0.6 kg	2.3 m/d	Upstream, Water-saturated	Demineralised water, 5
Lysimeter LUA NRW ¹	1.45 m, 1.4 m	1194 kg	0.06 m/d	Downstream, Water- unsaturated	Natural precipitation, 5.5-5.8

¹Susset et al., 2004.

Column B was run in three parallel tests, in order to evaluate the reproducibility of the method. The eluates obtained from column B passed a polyester filter (pore size 22 μm) at the column outlet and were collected by means of an automatic sample collector (Sample Collector/Pharmacia Fine Chemicals). The samples were exposed to ambient air during sampling and were not filtered. After measuring the pH value, an aliquot was used for the determination of chloride and sulphate, the rest of the sample was acidified (matrix: 1 % HNO_3 , suprapur/Merck). In contrast to the eluates of column B, the samples of column A were filtered (0.2 μm) before preservation and storage for further analysis. The pH value was measured by stream gauging. These samples were also exposed to ambient air in the course of sampling.

The seepage water from the lysimeter was collected in a stainless steel reservoir, where one sample was taken once a month. The samples were exposed to the air during sampling and were not filtered.

Batch leaching tests

The demolition waste material was also examined in batch leaching tests according to DIN 38414-4 (S4) (DIN, 1984) in order to compare the element concentrations with those obtained in column and lysimeter leaching tests. Furthermore, soil saturation extracts were prepared according to the Federal German Soil Protection and Contaminated Sites Ordinance (Federal Bulletin, 1999). Deionised water was added to 250 g of the material until it was fully imbued.

After 24 hours of storage at room temperature, more deionised water was added until the yielding point was reached. The liquid/solid ratio was approximately 0.25 L/kg. After another 24 hours of storage at 5 °C, the sample was centrifuged (3700 g) and the pore water obtained was filtered (0.2 µm cellulose acetate).

Analysis

Chloride and sulphate were determined by means of ion chromatography (IC). The metal concentrations in the eluates were determined by optical emission spectrometry with inductively coupled plasma (ICP-OES). The detection limit of copper and chromium was shown to be at least at 10 µg/L.

Results and Discussion

Column leaching behaviour

The eluates obtained in the column leaching tests were strongly alkaline (Fig. 1), with pH values ranging between 11.5 and 12.5. The pH values slightly decreased with increasing liquid/solid ratio. They were much lower in the lysimeter eluates than in the column leaching tests. This was probably due to the prolonged time of contact with the ambient air during the collection of the seepage water sample, when airborne carbon dioxide was allowed to react with the alkaline eluate and decrease the pH.

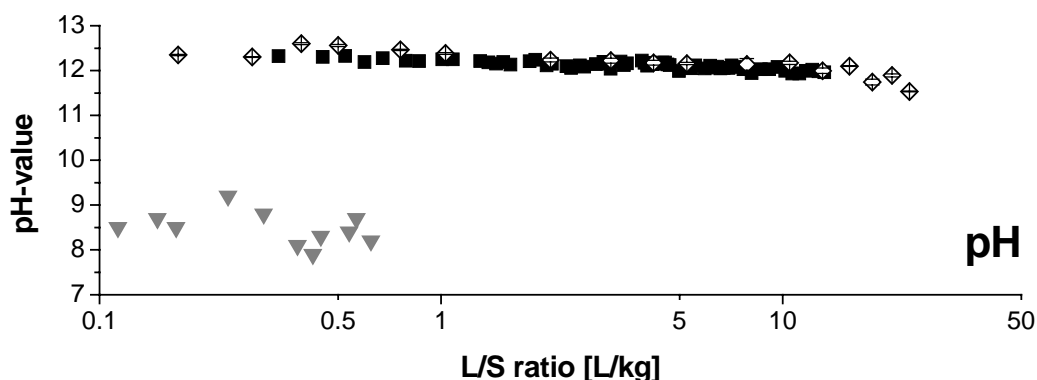


Figure 1: pH values in the column and the lysimeter leaching tests

As far as chloride, sulphate (Fig. 2), sodium and potassium (Fig. 3), chromium and copper are concerned (Fig. 4), the leaching behaviour in the two column leaching systems and the lysimeter showed good agreement despite the different column dimensions and flow conditions

(concentration differences less than factor 3) (Fig. 2-4). The comparison of the concentrations of calcium, aluminium, silicon, potassium, and sodium in the eluates obtained with the two column leaching systems also proved to be in good agreement upon applying these two methods (Fig 3, 5). Regarding all parameters investigated, the reproducibility of the column leaching tests B can be considered as appropriate for the reference demolition waste material.

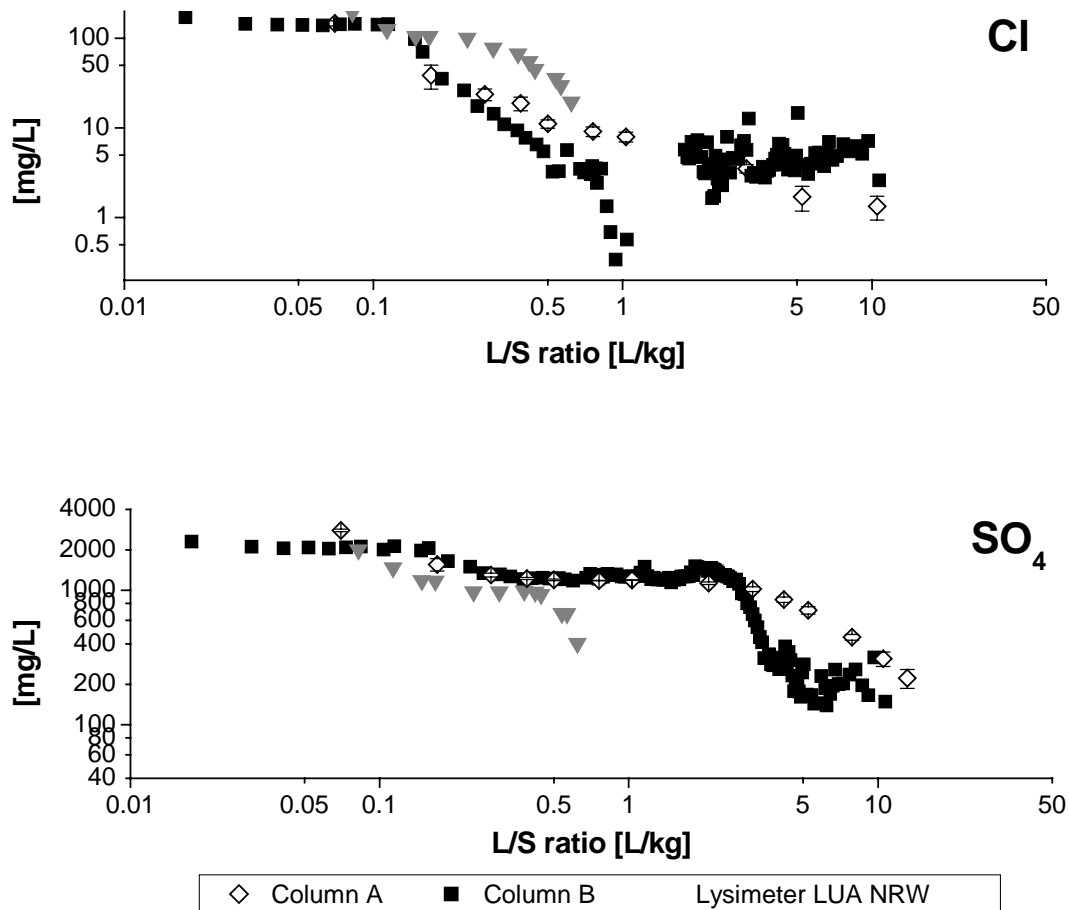


Figure 2: Comparison of leaching behaviour of chloride and sulphate in the column and the lysimeter leaching tests

In general, mobilisation of the inorganic components was highest in the initial stage of the elution. In this stage, dissolution and surface wash-off processes play a predominant role. The depletion of soluble or exchangeable components leads to lower concentrations in the eluates with increasing elution time. In view of environmental pollution, the high element concentrations of copper and chromium in the eluates have to be regarded critically, although the concentrations in the eluates decrease with the progression of leaching time and despite the fact that the overall mobility of the elements was low: after reaching a liquid/solid ratio of 20 L/kg, only a total amount of approximately 250 μg chromium was released in column B. Relative to the total

chromium content in the demolition waste material (16.3 mg/kg, Tab. 1), this is equivalent to a total mobilisation of roughly 2.5 %.

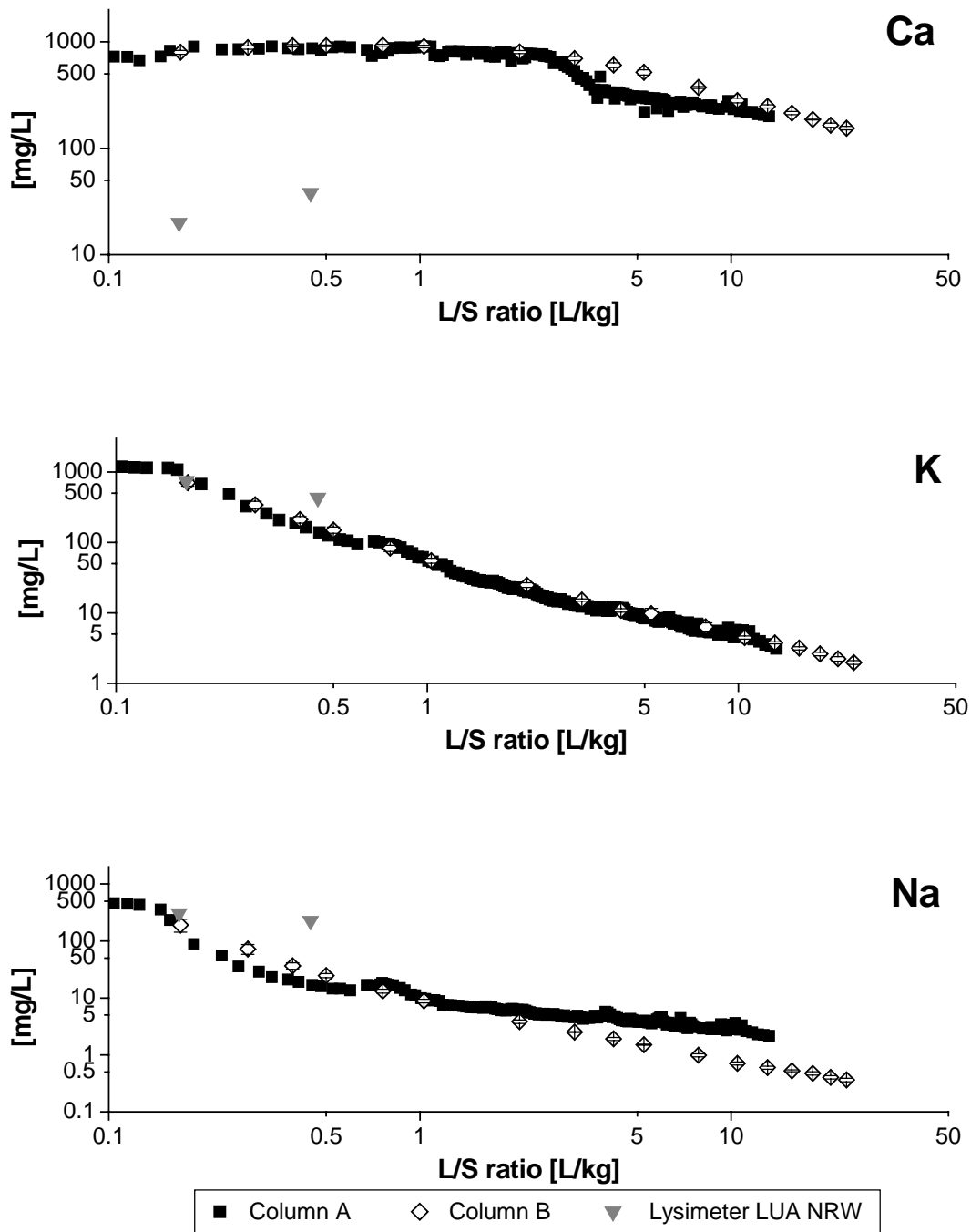


Figure 3: Comparison of the leaching behaviour of calcium, aluminium, silicon, potassium, and sodium in the column leaching tests

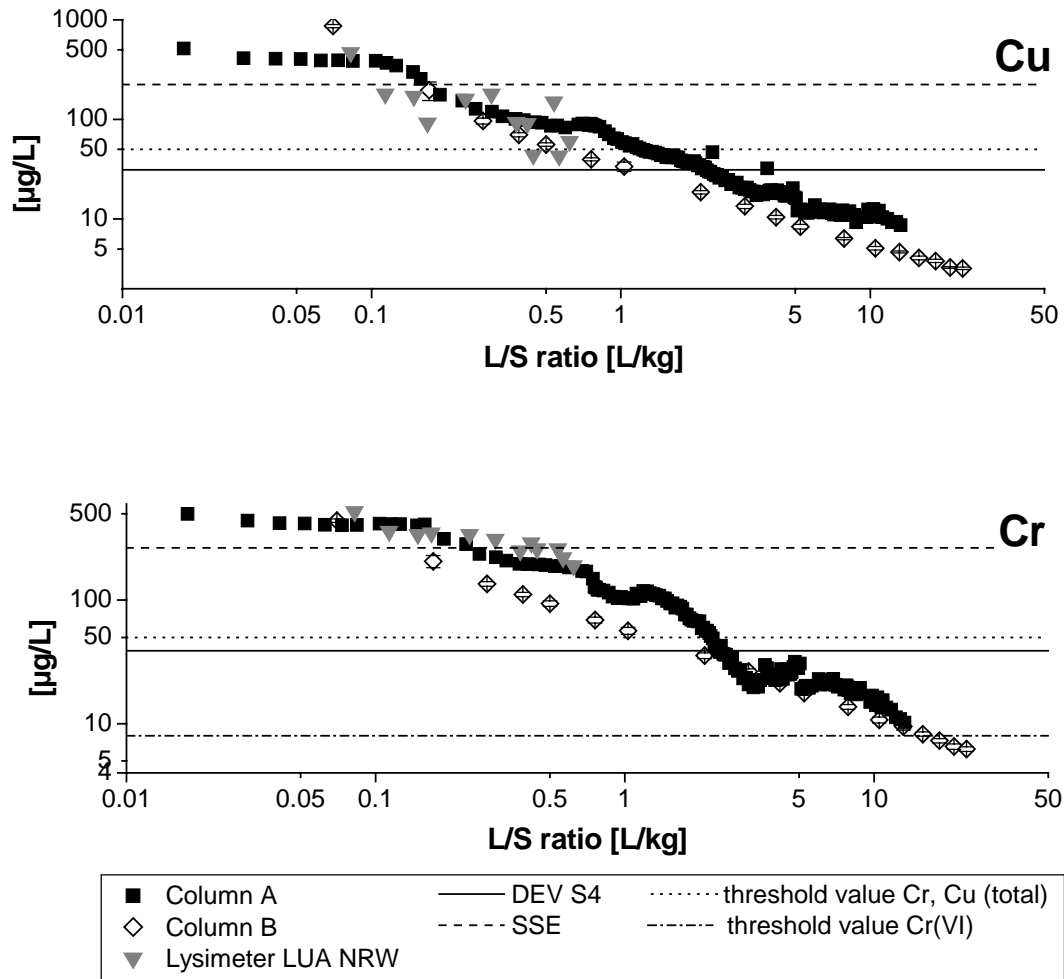


Figure 4: Comparison of leaching behaviour of copper and chromium in the column and the lysimeter leaching tests. For orientation, the concentrations in the DEV S4 and SSE eluates and the threshold values of the Federal German Soil Protection and Contaminated Sites Ordinance (Federal Bulletin, 1999) are shown.

The threshold values for chromium (total) and copper (total) were exceeded at least by a factor of 10 in the initial state of the elution. For chromium(VI), which is toxicologically relevant and likely to be the predominant chromium species at high pH values (Brookins, 1988), the threshold value was exceeded by factor 50.

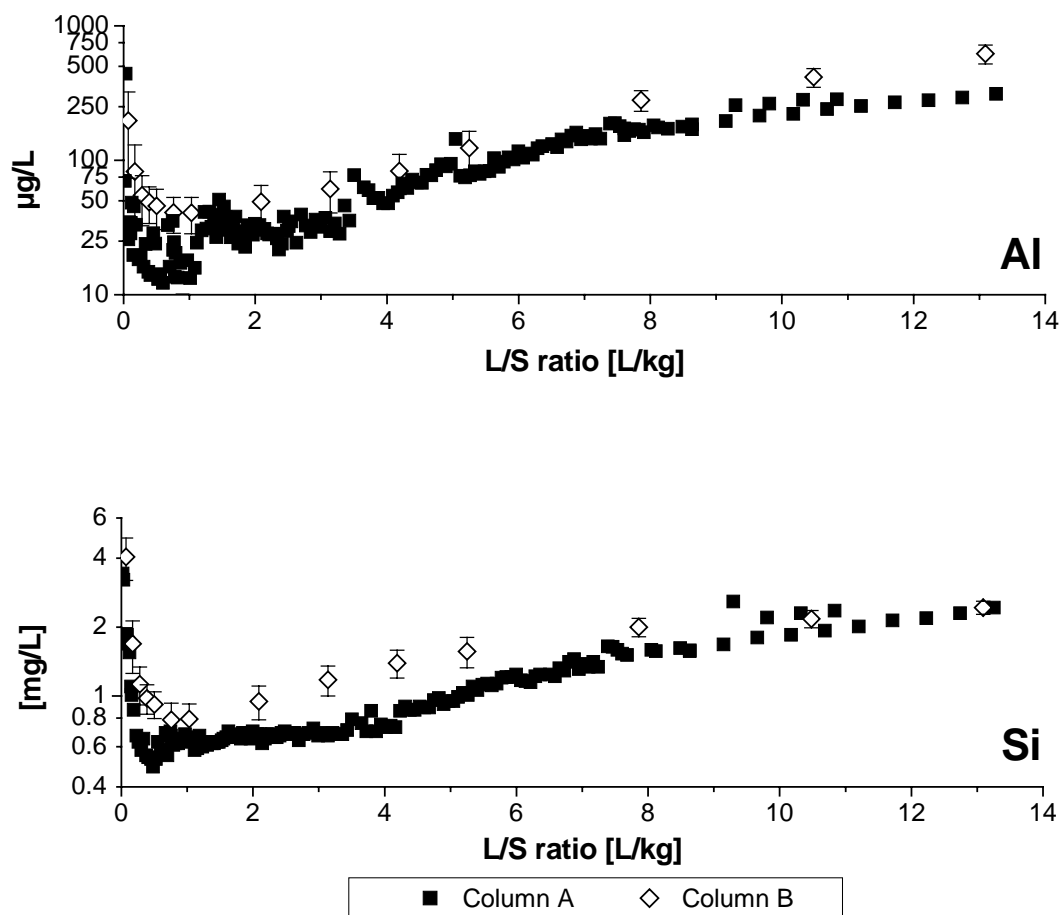


Figure 5: Comparison of the leaching behaviour of aluminium and silicon in the column and lysimeter leaching tests

Comparison with other leaching tests

Obviously, the concentrations in the S4 eluates do not reflect the concentrations occurring under natural conditions. In soils, lower liquid/solid ratios than 10 L/kg are present, since the total mass of water is much lower than the mass of the solid-phase. Therefore, the soil saturation extract with an average liquid/solid ratio of 0.3 L/kg represents a more realistic approach, but also does not reflect the high element concentrations appearing at the beginning of elution, as it can be seen in Figure 4. For example, the initial concentration of Cr in the soil saturation extract samples was at 266 µg/L, whereas in column B 440 µg/L were reached. For copper, the initial concentration in the soil saturation extract samples was 224 µg/L and 866 µg/L in column B.

However, a general comparison of concentrations determined by means of column leaching tests and batch experiments is not trivial, as the experimental conditions are different. In most approaches, the concentrations are compared with a defined liquid/solid ratio. In case of the release of chromium in column B, the concentration in the eluate was 136 ± 6 µg/L when the

liquid/solid ratio was at 0.3 L/kg, and 11 ± 1 $\mu\text{g/L}$ when the liquid/solid ratio was 10.5 L/kg. The concentration in the S4 eluate (liquid/solid ratio of 10 L/kg) was 39 $\mu\text{g/L}$, whereas in the soil saturation extract (liquid/solid ratio of 0.25) the chromium concentration reached a value of 266 $\mu\text{g/L}$ (Fig. 6). At first glance, batch experiments and soil saturation extract seem to overestimate the concentrations at a given liquid/solid ratio.

While the eluent is in constant contact with the examined material in the batch tests, the elution agent is constantly renewed in the column tests. This can lead to comparability difficulties when comparing batch and percolation tests. An alternative approach of comparing these two methods is relating the total amount of the released component at liquid/solid ratio x L/kg to the total amount of percolated elution agent at a liquid/solid ratio x L/kg and hence, to calculate an average concentration.

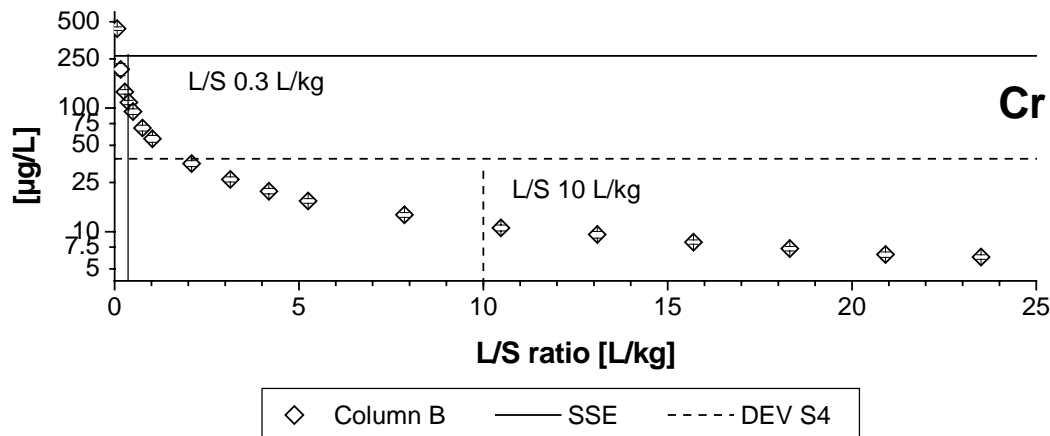


Figure 6: Comparison of concentrations obtained by means of a column experiment and batch experiments

If the total amount of eluted chromium in the column leaching test at a given liquid/solid ratio x L/kg ($m_{el,x}(\text{Cr})$) is considered by integrating elution curves (Fig. 7) and is related to the total elution volume ($V_{el,x}$), the average concentration ρ_{av} for chromium can be calculated as follows:

$$\rho_{av,x}(\text{Cr}) = \frac{m_{el,x}(\text{Cr})}{V_{el,x}}$$

In the column leaching test B, $\rho_{av,0.3}(\text{Cr})$ is 235 $\mu\text{g/L}$ and $\rho_{av,10.5}(\text{Cr})$ is 33 $\mu\text{g/L}$ (Fig. 7).

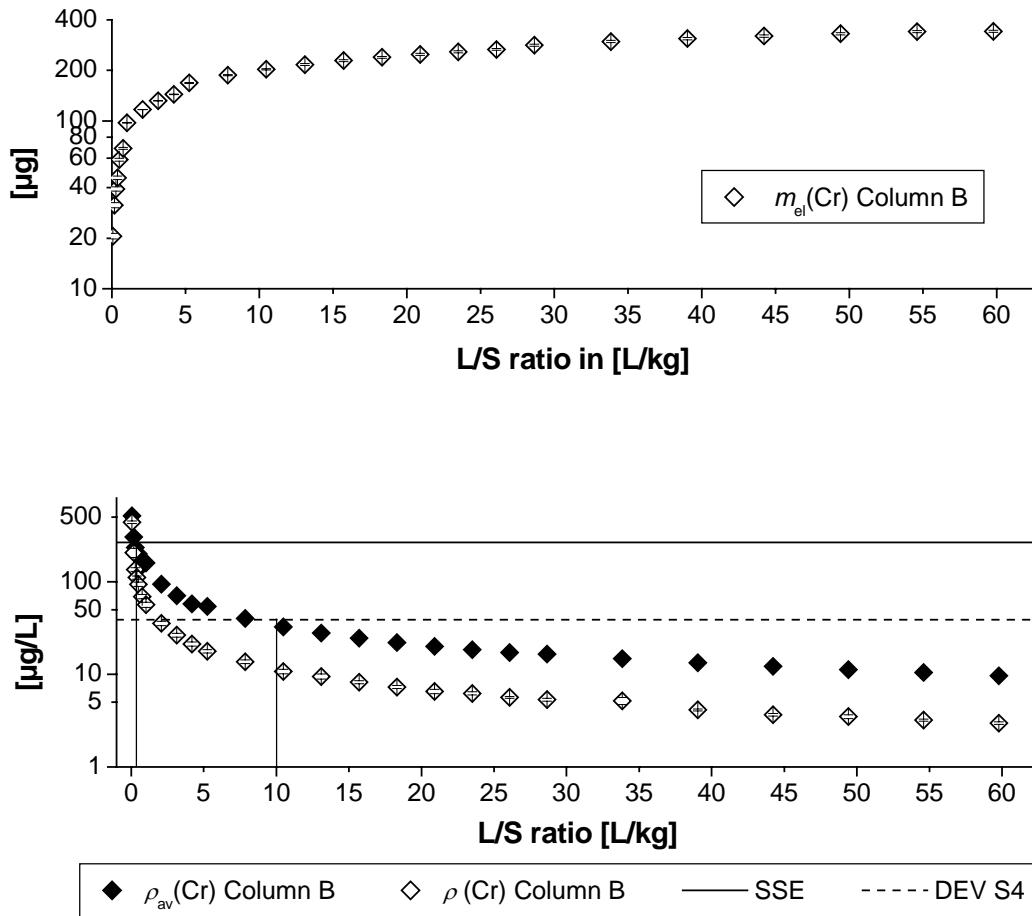


Figure 7: Cumulative release of chromium $m_{el}(Cr)$ (top) in column B and comparison of calculated average concentrations $\rho_{av}(Cr)$ and measured concentrations $\rho(Cr)$ with DEV S4 and SSE (bottom)

The calculated average concentrations ρ_{av} at a liquid/solid ratio of 0.3 and 10 L/kg are very close to the soil saturation extract and DEV S4 test results. This example shows that, not only the method question (column or batch experiment) has to be answered for risk assessment. It also has to be defined if the measured or the calculated average concentration at a certain liquid/solid ratio is the right concentration to determine the source strength and be applied in a reasonable assessment.

Conclusions

The results show that there is a good agreement between the leaching behaviour determined with the lysimeter unit of the LUA NRW and the column units used in the laboratory. In face of lower time and system requirements of laboratory column units, they can be regarded as a practicable tool to assess the time-dependent release of inorganic compounds under natural conditions. In the lysimeter unit, a liquid/solid ratio of about 0.6 L/kg is reached after 400 days; in comparison,

a liquid/solid ratio of 20 L/kg is reached after 240 days in column A and after 4.5 days with columns B. The low time requirement of column B corresponds especially with the needs of authorities or laboratories to have an easy to use and cheap, but nevertheless reliable method as basis for decisions. Low liquid/solid ratios reflect the initial state of the elution. Increasing liquid/solid ratios give information about the long-term leaching behaviour.

Due to the fact that the eluent is in constant contact with the material in batch tests, whereas the elution agent is constantly renewed in column tests, the question has to be answered whether the measured or the calculated average concentrations should finally be employed in risk assessments. As could be shown in the column experiments, there is no constant release of inorganic compounds. Their release changes in the course of time. In the worst case (no retardation and adsorption), the effluent concentrations might reach the groundwater. Generally, further strategies and decisions on the reuse of waste materials must clarify how to deal with the high element concentrations in eluates and the seepage water emerging in the initial stage of the elution.

If the decision on the reuse of waste materials is merely limited to a comparison of the source strength with threshold values, a simple DEV S4 test or soil saturation extract might be useful as a preliminary test. Figure 8 shows a potential risk assessment strategy regarding the source strength of a waste material. For detailed and time-resolved information, column leaching tests should be chosen in any case.

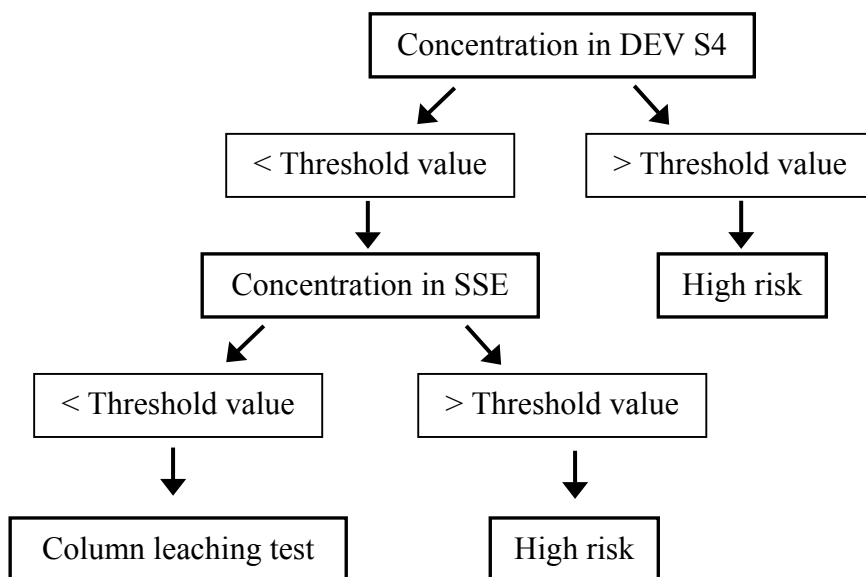


Figure 8: Potential strategy of a worst-case risk assessment.

3.3. Determination and Quantification of the release of Inorganic Contaminants from Municipal Waste Incineration Ash

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Determination and Quantification of the Release of Inorganic Contaminants from Municipal Waste Incineration Ash

Tanja Lager, Markus Delay, Volker Karius, Kay Hamer, Fritz. H. Frimmel and Horst D. Schulz

Abstract

Various leaching experiments applied to municipal waste incineration ash were compared in order to find the best available procedure to measure the total mass flow caused by natural leaching (source strength) of the material. The results of various batch experiments and two different laboratory column experiments run under water-saturated conditions were compared with water-unsaturated field-scale column experiments (lysimeter). The results of the batch experiments were not transferable to the column and field experiments. But, very good accordance could be observed between the breakthrough curves of the two column experiments conducted in the laboratory under water-saturated conditions and the water-unsaturated field-scale columns, with few exceptions. The results show that batch experiments are not sufficient to predict the source strength and its changes over time. Instead, column experiments are essential for a realistic risk assessment. For most inorganic parameters, a laboratory column experiment lasting less than 5 days is sufficient for a long-term risk assessment.

Keywords: municipal waste incineration ash, inorganic contaminants, source strength, leaching, leaching tests

Introduction

An estimated amount of more than 3 billion tonnes of waste are produced in Europe each year (EEA, 2003). Municipal waste makes up about 14 % of the total waste material (EEA, 2002). In 1999 and 2000, about 29% of the municipal waste in Western Europe was recycled, the main percentage (57%) was still deposited in landfills, and about 18% were admitted into incineration processes (EEA, 2003). Approximately 20 - 40 kg fly ash, 8 - 45 kg products from flue gas cleaning, and approximately 250 - 350 kg incineration ash are generated from one tonne of municipal waste (Thomé-Kozmiensky, 1994). Thus, between 105 and 147 million tonnes of municipal waste incineration ash accumulate in Europe each year.

In Germany, about 40 million tonnes of municipal waste are generated annually, 28 % of which are incinerated (German Federal Environmental Agency, 1999). Currently, there are 61 incineration plants operating as municipal waste treatment facilities in Germany. In addition, sewage sludge is thermally treated in 15 of them. These plants have an annual capacity of about 14 million tonnes of waste (German Federal Environmental Agency, 2003a). In spite of efforts to increase the amount of waste given into a recycling process, more incineration plants will be built in Germany. It is estimated that until 2005 about 73 incineration plants will be located in Germany, including those that are currently in planning stage. From the 1 June 2005 on, all disposal sites have to comply with the criteria of the Waste Disposal Ordinance (Federal Bulletin, 2002a) and the Disposal Site Ordinance (Federal Bulletin, 2002b). If a disposal site does not fulfil the requirements it has to be either upgraded or closed. In addition, the waste itself has to comply with certain requirements as well, which makes a pre-treatment of most of the municipal waste necessary. In 2005 capacities will therefore be needed for the treatment of more than 28 million tonnes of domestic waste and thus more municipal waste incineration ash will be produced (German Federal Environmental Agency, 2003b).

Reusing this industrial product of the waste incineration process has priority over its deposition. As far as the reuse of waste materials is concerned, methods have to be developed which allow an environmental risk assessment for different recycling scenarios. Batch tests, laboratory column tests and field-scale column tests were employed in the scope of a project funded by the federal Ministry for Education and Science (BMBF), called 'Seepage Water Prognosis'. One of the main objectives of these test procedures was to describe the source strength of the material, which is defined as the total mass flow caused by natural leaching (Oberacker and Eberle, 2002). As the source strength is a parameter, which changes with time, methods that allow a long-term risk assessment should be used.

In search of an appropriate long-term risk assessment method, the results of various batch methods and two differently dimensioned column experiments run under water-saturated conditions are presented and compared with field-scale experiments. A wide variety of parameters were taken into account in order to ascertain a detailed comparison: calcium, barium, strontium, sodium, potassium and silicon as the main elements, copper, chromium, molybdenum and nickel; the anions sulphate and chloride as trace elements, the pH value and the concentration of total organic carbon (TOC).

Material

Municipal waste incineration ash is a very heterogeneous material. The main parameters that influence the composition of the material are:

- The composition of the waste, which mainly depends on the time of year the waste is collected and the catchment area of the incineration plant
- the temperature and the duration of the incineration process

The material investigated in the scope of this work was produced by the Federal Department of Material Investigation (BAM) as reference material according to ISO Guideline 30 (1992). The guideline demands *“a sufficiently homogeneous material, of which one or more characteristic values are precisely defined, so that the material can be taken for calibration of measurement devices, evaluation of measurement methods and allocation of values for different substances”* (ISO Guideline 30, 1992). This includes the following requirements:

- low residual water content
- low content of fine particles
- maximum grain-size of 4 mm
- k_f -value of more than 10^{-6} m/s
- flowability.

In addition, the ash must have been deposited for 3 months. The material is a mixture of municipal waste incineration ashes collected from 4 different waste incineration plants in Germany. All laboratory methods required for the characterisation of the material including the determination of the bulk concentrations of both major and trace elements were conducted, the results are shown in Tables 3 and 4.

Methods

Bulk concentration

The bulk concentration of the material was determined by applying two different methods. The microwave digestion system (MLS Ethos 1600) uses a digestion solution composed of 3 mL HNO₃, 2 mL HF and 2 mL HCl. The system is temperature-controlled. At maximum temperatures of 240°C and a maximum pressure of 30 bar most the samples are digested within

1.5 hours. After another 2-3 hours of fuming off and homogenisation the sample is ready for analysis in the ICP-OES. Secondly, a solid- phase analysis was performed by applying energy dispersive polarization X-ray fluorescence analysis (EDP-XRF) (Spectro Xepos). The microwave digestion is the more precise method. However, it has the disadvantage that silicon cannot be measured on account of its volatility.

Laboratory column units

Two differently dimensioned column experiments were conducted. Both columns are made of Perspex and were run under saturated conditions, the flow direction was from bottom to top.

The first column (column A) had a length of 48.3 cm and a cross-sectional area of 78.5 cm². Loading of column A was carried out with water-saturated material, which for this purpose was gradually filled into the column under mild shaking in order to compact the material. A slight excess amount of water avoided the entrapment of air bubbles and a grain- size fractionation during the filling process. In total, 1.7 kg of dry material was loaded into the column. A polyester filter with a pore diameter of 63 µm was attached at the column inlet and outlet. Artificial rainwater adjusted to a pH value of 4.1 with a mixture of H₂SO₄ and HNO₃ was used as the eluent. The flow velocity was set to 6 mL/h. This led to a Darcy velocity of 7.3 cm/d. The absolute porosity was 31 %. The effective velocity and the effective porosity were determined by means of the estimated exchanged pore volume. The exchange of one water-saturated pore volume can be derived from the time the first flush ended. The water-saturated pore volume was exchanged once in 1.8 days. Given a column length of 48.3 cm this is equivalent to an effective velocity of 24 cm/d and an effective porosity of 30 %. According to Beyer (1964), this material is characterised by a coefficient of hydraulic conductivity (k_f) of $6.5 \cdot 10^{-5}$ m/s. A liquid/solid ratio of approximately 20 L/kg was reached after 240 days. All samples were collected by an autosampler and exposed to the air in the course of the sampling procedure, which afforded 3.5 hours per sample. The samples were filtered (0.2 µm, cellulose acetate) before preservation. Samples drawn for the analysis of cations were conserved at pH 1, samples for analysis of anions and total organic and inorganic carbon were stored at 4°C. The parameters pH, redox potential and electrical conductivity were gained by stream gauging. A technical description of column A can be found in Lager et al. (2003).

Three flow interruption experiments were conducted (Tab. 1) in order to study the reaction of the system to changes in the hydraulic regime. In addition, sampling along the flow-path was carried out after 83 days at a liquid/solid ratio of approximately 6.7 L/kg, and after 276 days at a

liquid/solid ratio of about 21 L/kg. The results give an indication of the processes behind the mobilisation and fixation reactions.

Table 1: Flow interruption experiment

L/S [L/kg]	Length of interruption [d]
1.8	2
8.2	2.5
21	26

Column B was run in three parallel tests. Each column had a length of 23.7 cm and a cross-sectional area of 50.2 cm². Columns B1-3 were filled with 0.6 kg of the air-dried municipal waste incineration ash and were then flooded. A polyester filter with a pore diameter of 20 µm was attached to the inlet and outlet of the column. The eluent consisted of deionised water adjusted to pH 5 (HNO₃ suprapur; Merck). The flow velocity was set to 120 mL/h, the hydraulic parameters were determined by a tracer test, i.e. Darcy velocity (2.3 m/d), effective velocity (7.7 m/d), and effective porosity (30 %). A liquid/solid ratio of 20 L/kg was reached after approximately 4.3 days. Sampling was conducted as with column A. The samples were not filtered. A technical description of the column experiment can be found in Delay and Frimmel (2003).

Field-scale column units (lysimeter)

In order to verify the transferability of the results of the laboratory methods, the results were compared to two field-scale column experiments carried out with the same municipal waste incineration ash. These lysimeter tests were conducted by the Research Centre for Environment and Health (GSF) in Neuherberg, southern Germany (Lysimeter A) and the North Rhine-Westphalian State Environment Agency (LUA NRW) in Essen (Lysimeter B). Both lysimeters were made of stainless steel and in both cases the only irrigation consisted in natural precipitation. The pH value of the rainwater measured over a period of two years (2002/2003) by the GSF in Neuherberg averaged at 6.5 and varied between 5.45 and 8.1, while the electric conductivity averaged at 65 µS/cm. The minimum value was 9, the maximum value 639 µS/cm. Evaporation and transpiration was prevented by the incorporation of a filter layer on top of the municipal waste incineration ash in both lysimeters and the absence of vegetation on the top layer.

Lysimeter A had a length of 85 cm and a cross-sectional area of 1 m². A 5-cm thick filter layer consisting of quartz sand at the bottom was followed by 50 cm of the municipal waste incineration ash (90 kg), which was covered by a 30-cm thick layer of quartz sand. The seepage water was collected in a reservoir underneath the lysimeter, and sampling was conducted once a week. The samples were exposed to air during sampling and were not filtered. The seepage velocity varied between 7 and 16 m/a. A liquid/solid ratio of about 1.2 L/kg was reached after 400 days. The sampling period started in January 2002. Until June 2003, 107 mm of rain fell and 107 L of sample were collected. For further information see Klotz et al. (2004) and Klotz and Schremm (2004).

Table 2: Test conditions and selected release controlling factors of the percolation tests applied

Method	Length / cross-sectional area	Mass of incorporated MWI ash	Water saturation / Flow direction	Average flow velocity (v _f)	Elution agent / pH	Elution time
Column A	48 cm / 19.6 cm ²	1.7 kg	Saturated / upwards	8 [cm/d]	Artificial rainwater / 4	Months
Column B	23.7 cm / 12.6 cm ²	0.6 kg	Saturated / upwards	2.3 [m/d]	Deionised water / 5	Days
Lysimeter A ¹	86 cm / 1 m ²	90 kg	Unsaturated / downwards	1 [cm/d]	Natural precipitation / 5.5 - 5.8	Years
Lysimeter B ²	1.45 m / 1.54 m ²	1266 kg	Unsaturated / downwards	5.7 [cm/d]	Natural precipitation / 5.5 - 5.8	Years

¹ Susset et al., 2004a, b

² Klotz et al., 2004a, b

Lysimeter B measured 1.45 m in height and had a cross-sectional area of 1.54 m². Above the filter layer, which consisted of 10 cm of quartz gravel, 45 cm of the municipal waste incineration ash (1266 kg) were loaded into the column. 90 cm of quartz gravel formed the cover layer on top. A perforated metal plate with holes measuring 5 mm in diameter was installed at the bottom of the column. The seepage water was collected in a stainless steel reservoir, where one sample was taken once a month. The samples were exposed to the air during sampling and not filtered. A liquid/solid ratio of about 0.6 L/kg was reached after 400 days. The time period of sampling documented in this paper lasted from late December 2002 until early January 2004. 570 mm of rain fell during these 12 months, which is equivalent to a theoretical irrigation of 875 litres of water in the lysimeter. The sample amount accumulated to 720 litres. A deviation of 18 % was due to the incorporation of dry municipal waste incineration ash. The 155 litres apparently unaccounted for were within the lysimeter, as they

constitute the field capacity. A detailed description of the lysimeter can be found in Susset et al. (2004a) and Susset et al (2004b).

Batch tests

Overview of the leaching tests that have been conducted with the material:

- German standard method DEV S4 (DIN, 1984); leaching with deionised water for 24 h with a liquid/solid ratio of 10 L/kg in an overhead shaker
- Ammonium-nitrate extract (DIN, 1997b); leaching with 1 M ammonium-nitrate solution for 3 h with a liquid/solid ratio of 2.5 L/kg in an overhead shaker
- Soil saturation extract (Federal Bulletin, 1999); leaching for 48 h with a liquid/solid ratio of approximately 0.25 L/kg stored at 4°C
- pH-stat test (Obermann and Cremer, 1992; Karius and Hamer, 2001b); leaching for 24 h with deionised water adjusted to a specific pH with HNO₃ or NaOH (pH 4, 5, 6, 7, and 11) with a liquid/solid ratio of 10 L/kg in a beaker with a stirrer

A detailed description of the batch methods as well as a Table with some release controlling factors of the methods applied can be found in Lager et al. (2003).

Analysis, phase analysis and thermodynamic calculation

Sulphate and chloride were measured using a HPLC system equipped with an anion separation column and indirect UV-detection. Total organic carbon was determined as the difference between total carbon and total inorganic carbon, both measured with a SHIMADZU TOC 5050. The CO₂ released from the sample by an addition of concentrated H₃PO₄ to the sample was measured as total inorganic carbon, CO₂ released from the pure sample at 680°C as total carbon.

The concentrations of the cations were determined by ICP-OES measurement (Perkin Elmer, Optima 3000), the apparatus being connected to a cross-flow nebulizer to measure the main elements, and to an ultrasonic nebulizer (CETAC USN) for the determination of heavy metals. The detection limit of heavy metal analysis was occasionally reached. Due to the fact that the ICP-OES connected with the ultrasonic nebulizer was not able to measure higher bulk contents than 1 g/L, samples withdrawn from the outlet of the columns possessing a liquid/solid ratio of up to 7 L/kg had to be diluted. The detection limit of the heavy metals was at 10 µg/L, when dilution was necessary, whereas the limit of detection was at 2 µg/L in undiluted samples, and at 5 µg/L for the batch test samples.

The main mineral constituents of the municipal waste incineration ash were investigated by means of powder diffraction (XRD). Data were collected with a Philips X'Pert PRO powder diffractometer including CuK α radiation and a secondary monochromator in the range between 5-80° 2 θ equipped with a X'Celerator detector.

PHREEQC (version 2.8) (Parkhurst and Appelo, 1999) was used in this study to calculate the speciation and saturation indices applying to the column experiments. It is a computer programme for the speciation and calculation of the saturation index, batch reactions, one-dimensional (1D) transport, and inverse modelling. Input parameters consisted in the complete respective analysis. In the municipal waste incineration ash investigated potassium-phosphate could be identified as minor constituent by XRD-analysis. As potassium-phosphate is likely to dissolve during the elution in the column and as it had not been analysed, a charge balance was accomplished with phosphate. All phosphate-phases were undersaturated in all samples calculated with PHREEQC.

Results and Discussion

Material Properties

A comparison of the data measured with the values submitted by the Federal Department of Material Investigation (BAM) reveals a very good agreement for most material parameters (Table 1-3).

Table 3: Characteristic material properties

	MWI ash	MWI ash (BAM) ¹
Water content [weight-%] (DIN, 1989)	3.7	3.7
Specific weight [g/cm ³] (DIN, 1997a)	2.62	2.60
Specific surface [m ² /g]	5.4	5.2
pH (<i>in situ</i>)	11.4	11.1
EH (<i>in situ</i>)	304	
Electrical conductivity [mS/cm]	1.2	2.0
TC [%]	0.90	0.86
IC [%]	0.46	0.33
TOC [%]	0.44	0.53

¹ (Berger et al., 2004)

Table 4: Grain- size distribution of the municipal waste incineration ash investigated (Berger et al. 2004)

[mm]	4 - 2	2 - 0.63	0.63 - 0.2	0.2 - 0.063	< 0.063
[weight-%]	19.5	26.8	31.0	17.6	5.0

The municipal waste incineration ash mainly consists of quartz. Furthermore minor amounts (less than 10 %) of calcite, plagioclase, magnetite, and potassium-phosphate were identified in the municipal waste incineration ash (Fig. 1). In addition the ash contains an amorphous fraction.

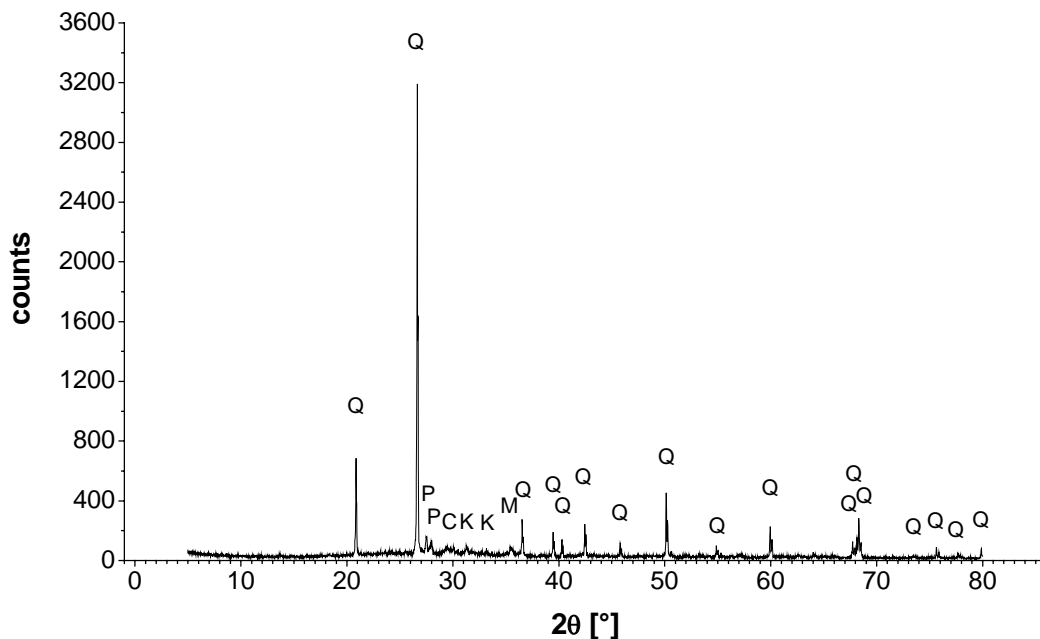


Figure 1: Observed powder diffraction patterns in the municipal waste incineration ash (Q = quartz, P = plagioclase, K = potassium-phosphate, M = magnetite, C = calcite)

The bulk concentrations gained by the two different methods, microwave digestion and XRF, are compiled in Table 5 and 6. The standard deviation was less than 10 %. The results show differences of up to 30 % between both methods. A comparison with the data gained by the Federal Department of Material Investigation (BAM) shows variations of about 10 %, with the exception of chromium and copper.

The bulk concentration of the municipal waste incineration ash exceeded all precautionary values defined for various elements in the Federal German Soil Protection Ordinance (Federal Bulletin, 1999). Exceeding this value indicates the risk of potentially harmful soil changes.

Table 5: Bulk concentration of the main elements, in [g/kg]

MWI ash	Al	Ca	Fe	K	Mg	Na	P	S	Si
Microwave digestion	37	72	57	9.5	9.9	23	—	4.3	
XRF	42	86	45	9.3	12	—	3.9	10	190
BAM ¹	—	70	50	—	—	—		—	

¹ (Berger et al., 2004)

Table 6: Bulk concentration of the trace elements in [mg/kg] and precautionary values for soils (Federal Bulletin, 1999)

MWI ash	As	Ba	Cd	Co	Cr	Cu	Mn	Mo	Ni	Pb	Sr	V	Zn
Microwave digestion	7.3	1161	4.3	18	237	1253	617	6.3	158	833	289	14	2214
XRF	5.7	—	5.9	26	312	1109	684	—	126	814	194	23	2179
BAM ¹	8.0	1300	5.8	—	67	1657	600	—	68	727	—	—	2073
Precaution value	—	—	0.4	—	30	20	—	—	15	40	—	—	60

¹ (Berger et al., 2004)

Laboratory column tests

General observations

In both columns, the release of all elements was very high during the initial phase of elution (“first flush”). The various concentration characteristics (e.g. Fig. 2) of this phenomenon observed in both columns were due to different column- loading procedures. As described above, loading of column A was carried out with water- saturated material and a slight excess of water in order to avoid the entrapment of air. Air bubbles might account for a lower effective pore volume, resulting in a larger effective flow velocity and thus a reduced time of contact with the material.

The frequency of sampling also had an effect on the concentrations measured. Samples were taken from column A every 3.5 hours. Thus, one sample (21 mL) equalled approximately one-fourteenth of the pore volume in column A. The first sample from column B analysed equalled one fully exchanged pore volume. This led to the measurement of higher concentrations in samples from column A and a greater resolution in time.

The pH value, the concentrations of major elements and trace elements as well as the concentration characteristics were similar, except for silicon and TOC, in the two column experiments. Figure 2 shows the concentrations and the amounts of total leached calcium. It is

obvious that not only the concentrations of the outlet samples of both columns were identical, but also the leached amount of calcium relative to the dry mass of material applied in the respective column. The various hydraulic regimes produced similar results. Despite the different flow velocities that have an influence on the reactions controlled by kinetic processes, the concentrations in the outlet samples of the two columns were in good agreement. This was only possible if the phases that showed fast dissolution kinetics under the prevailing conditions and were close to thermodynamic equilibrium at the end of each column had been dissolved. If slow dissolution kinetics had dominated the system, the released concentrations would have been higher in column A (lower flow rate) than in column B.

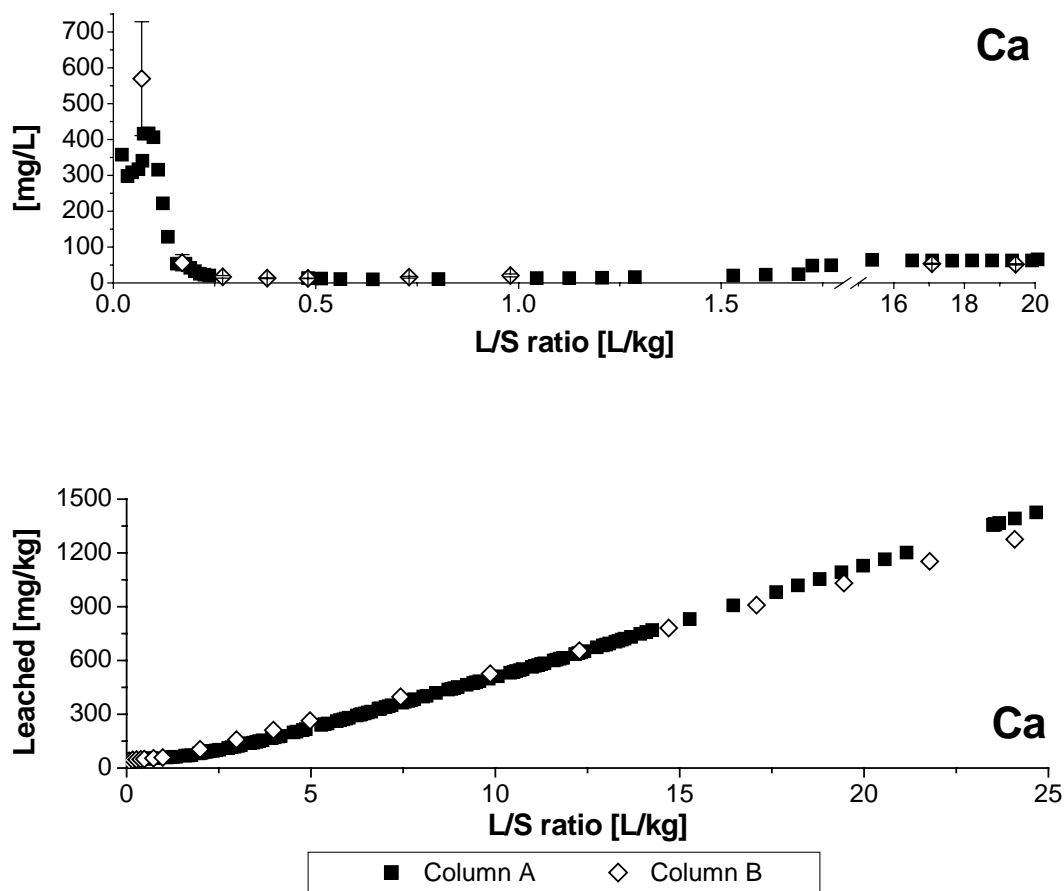


Figure 2: Concentration and leached amounts of calcium in the outlet samples of column A and B relative to the dry mass of the material

Calculations of the saturation indices of the phases identified by means of XRD show that the saturation indices of the main phases (quartz, feldspar, calcite) in column A barely varied within the last 38 centimetres of the column (Fig. 3). With only few exceptions, the elements demonstrated increasing mobilisation along the flow-path up to a certain level, after which no further release could be observed (Fig 4).

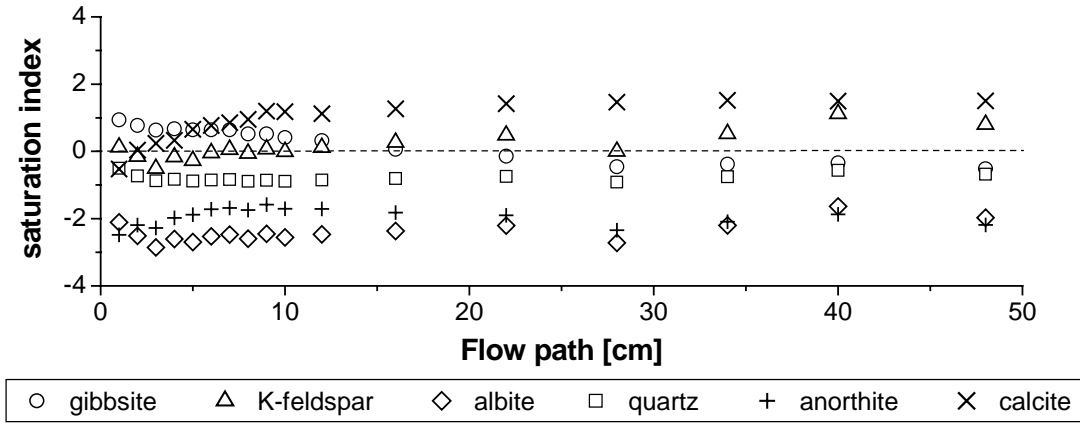


Figure 3: Saturation indices of the samples withdrawn from the flow-path of column A after 276 days, L/S 21 [L/kg]

Kinetics of calcite solution: Saturation length

Schulz (1988) established a method, which allows an easy and direct determination of the carbonate dissolution kinetics by measuring the length of saturation in sandy aquifer material. The basis for this method is a model for the dissolution kinetics of carbonates in natural systems, developed by Buhmann and Dreybrodt (1985a, b). The saturation length x_s (equation 2) is defined as the length of the flow-path at which the concentration in the pore water reaches 1-1/e of the concentration at equilibrium (63 % of C_{eq}) (Schulz, 1988) (Fig. 4).

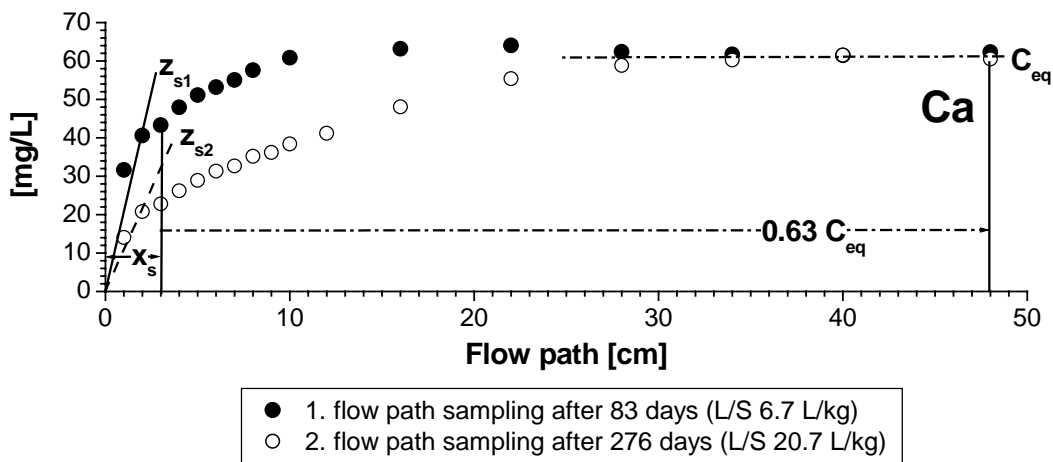


Figure 4: Concentration of calcium in the two flow-path samples taken from column A (C_{eq} [mol/L]= concentration at equilibrium, z [mol/L*cm] = initial gradient of the function of the dissolution rate, x_s [cm] = saturation length)

$$r \left[\frac{\text{mol}}{\text{L}} \cdot \text{sec} \right] = k \left[\text{sec}^{-1} \right] \cdot \left(C_{eq} \left[\frac{\text{mol}}{\text{L}} \right] - C \left[\frac{\text{mol}}{\text{L}} \right] \right) \quad (1)$$

$$x_s \left[\text{cm} \right] = \frac{v_a \left[\frac{\text{cm}}{\text{sec}} \right]}{k \left[\text{sec}^{-1} \right]} = \frac{v_a \left[\frac{\text{cm}}{\text{sec}} \right] \cdot v_p \left[\text{cm}^3 \right]}{\alpha \cdot S \left[\text{cm}^2 \right]} \quad (2)$$

$$k \left[\text{sec}^{-1} \right] = \frac{S \left[\text{cm}^2 \right] \cdot \alpha}{v_p \left[\text{cm}^3 \right]} = \frac{z \cdot v_a \left[\frac{\text{cm}}{\text{sec}} \right]}{C_{eq} \left[\frac{\text{mol}}{\text{L}} \right]} \quad (3)$$

with: $r \left[\text{mol/L} \cdot \text{sec} \right] =$	dissolution rate
$k \left[\text{sec}^{-1} \right] =$	material- dependant constant
$C_{eq} \left[\text{mol/L} \right] =$	concentration at equilibrium
$C \left[\text{mol/L} \right] =$	current concentration
$v_a \left[\text{cm/sec} \right] =$	effective flow velocity
$v_p \left[\text{cm}^3 \right] =$	effective pore volume
$\alpha =$	gradient of the specific function of the dissolution rate
$S \left[\text{cm}^2 \right] =$	reactive surface
$z \left[\text{mol/L} \cdot \text{cm} \right] =$	initial gradient of the function of the dissolution rate

The determination of the material-dependant coefficient k was accomplished following equation 3. The coefficient k is a composite parameter representing the effective pore volume, the reactive calcite surface, and the transport from the grain surface into the solution of the pore volume. By knowing this coefficient k , the effective flow velocity v_a , the effective pore volume v_p and the saturation length x_s , the gradient of the specific function of the dissolution rate α , hence, the kinetic reaction constant can be determined (equation 2).

Figure 4 shows the calcium concentrations in the pore water along the flow-path of the column after 83 and 276 days. The values for k are $2.9 \cdot 10^{-5} \text{ sec}^{-1}$ after 83 days, and $1.5 \cdot 10^{-5} \text{ sec}^{-1}$ after 276 days, the saturation length x_s was calculated as 2.9 and 3.1 cm respectively (equation 2). Thus 63 % of the equilibrium concentration was reached after approximately 3 cm. In order to determine the kinetic reaction constant α , the reactive surface of calcite had to be known, next to the effective pore volume v_p . Semi-quantitative XRD-analysis of the mineral phases identified calcite with a content of less than 10 %. The bulk concentration of calcium in the municipal waste incineration ash was 76 g/kg (mean value obtained by three different methods, Table 5). Presuming that half of the calcium was derived from calcite, the municipal waste incineration ash contained about 10 % calcite. Geometric calculation led to a surface of approximately 40

cm²/g, assuming a cube with an edge length of 0.6 mm—which was the mean grain diameter of the municipal waste incineration ash—as being representative of one calcite grain. Baumann (1986) calculated surfaces of 73 cm²/g for a grain diameter of 0.3 to 0.4 cm, and 58 cm²/g for a grain diameter of 0.4 to 0.5 cm assuming a rhombohedron for each calcite particle. The specific surface of the calcite within the first three centimetres of the column (saturation length) had to be regarded in the determination of the reactive calcite surface. A specific surface of 40 cm²/g and a calculated reactive calcite surface of 588 cm² led to a kinetic constant of $4.8 \cdot 10^{-5}$ cm/sec after 83 days of the column test, and $2.9 \cdot 10^{-5}$ cm/sec after 276 days. Thus the kinetic constant decreased with the progression of time. This is due to the dissolution of calcite within the first 3 centimetres of the column, resulting in a decrease of the reactive calcite surfaces. However, these values were in the range of theoretic and experimental values determined by Baumann et al. (1985), Baumann (1986) and Schulz (1988).

Silicon, total organic carbon and aluminium

Silicon is the only element that showed a different pattern concerning its concentration characteristics at the outlet of columns A and B (Fig. 5). The outlet concentrations of column A were up to five times higher than in column B. This indicates the dissolution of silicon containing phases that have rather slow dissolution kinetics. Karius et al. (2002) discovered the same phenomenon in column experiments with brick sand, which were part of a reactive transport modelling study. The time available for leaching, and thus the flow velocity are more important than the liquid/solid ratio (Karius et al., 2002).

The different pH values applied in the column tests (column A = pH 4, column B = pH 5) did not have any influence on the mobilisation of elements. Due to the high acid neutralisation capacity of the municipal waste incineration ash (Tab. 8), a pH of 9.8 developed within the first centimetre of the column (Fig. 5). In the course of the experiment (300 days) the mean pH value changed from 11.9 to 11.2.

TOC showed similar concentration values until a liquid/solid ratio of 2 L/kg was reached. While in column A the values increased again after a minimum at a liquid/solid ratio of 2 L/kg, the decrease continued in column B until the liquid/solid ratio was at a value of 10 L/kg. Afterwards a slight increase could be observed. It may be presumed that dissolution was the main release-controlling factor below a liquid/solid ratio of 2 L/kg. In the meantime the degradation of organic matter started. Consequently, the concentration of organic carbon increased. This process obviously depended on the flow velocity. Münch et al. (2002) investigated the influence of the effective pore water velocity in unsaturated column experiments with podsol and gleysol

soil samples. The release of dissolved organic carbon was linearly related to the flow velocity of the pore water. An increasing velocity and thus a shorter time of pore water residing in the column resulted in a decrease of dissolved organic carbon. The assumption that the longer overall duration of the experiments (of column A 300 days, column B 5 days) leads to the higher concentrations detected in column A is contradicted by the lysimeter A and B results. The concentrations of the total organic carbon were by a maximum factor of 4 below the concentrations of columns A and B, except for a few lysimeter B samples. Because of the longer overall duration (lysimeter A 470 days, lysimeter B 385 days) the concentrations should be higher as compared to the column tests.

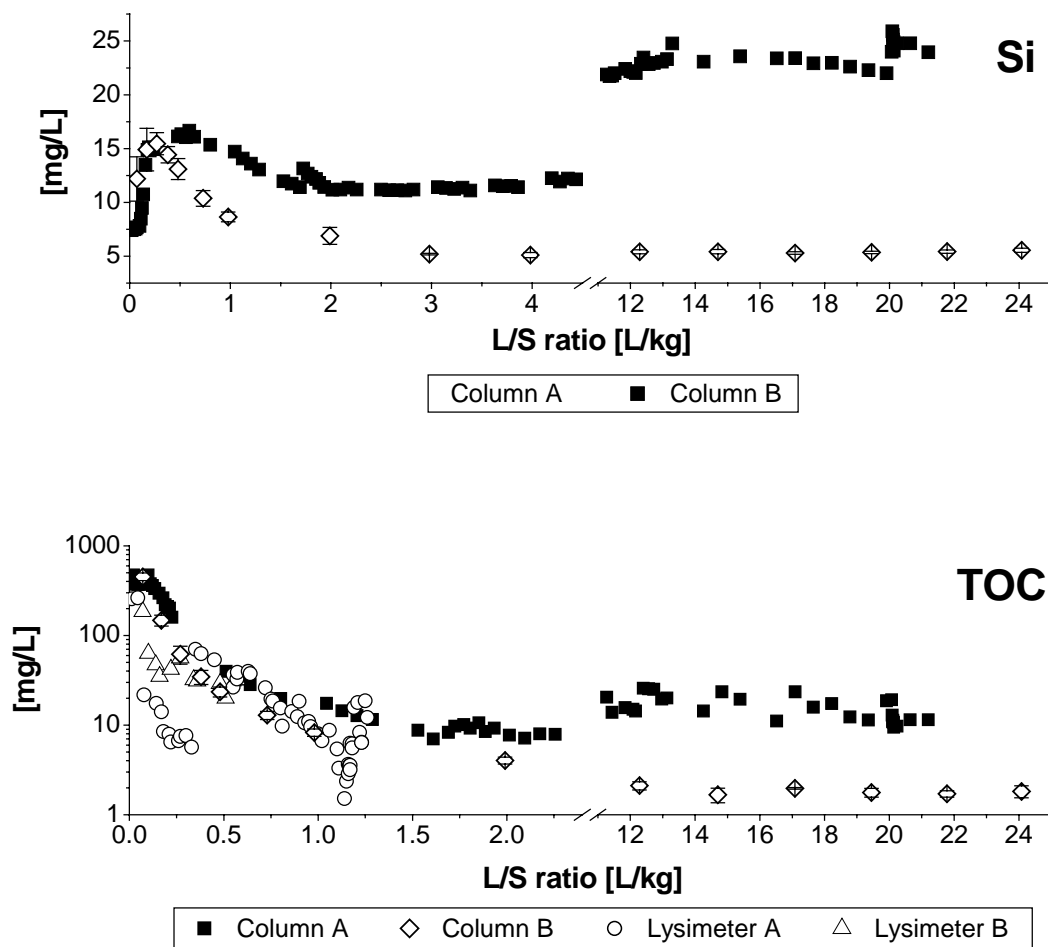


Figure 5: Concentration of silicon in the outlet samples of columns A and B and total organic carbon (TOC) in the outlet samples of columns A and B and lysimeters A and B

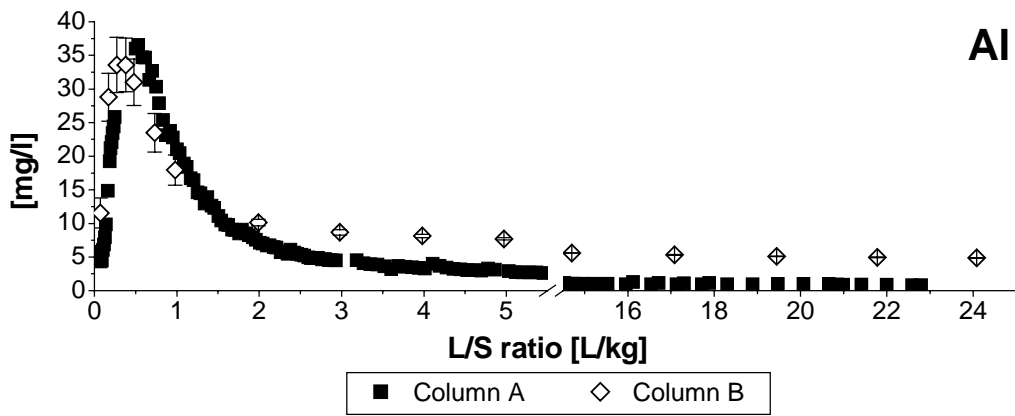


Figure 6: Concentration of aluminium in the outlet samples of the two columns

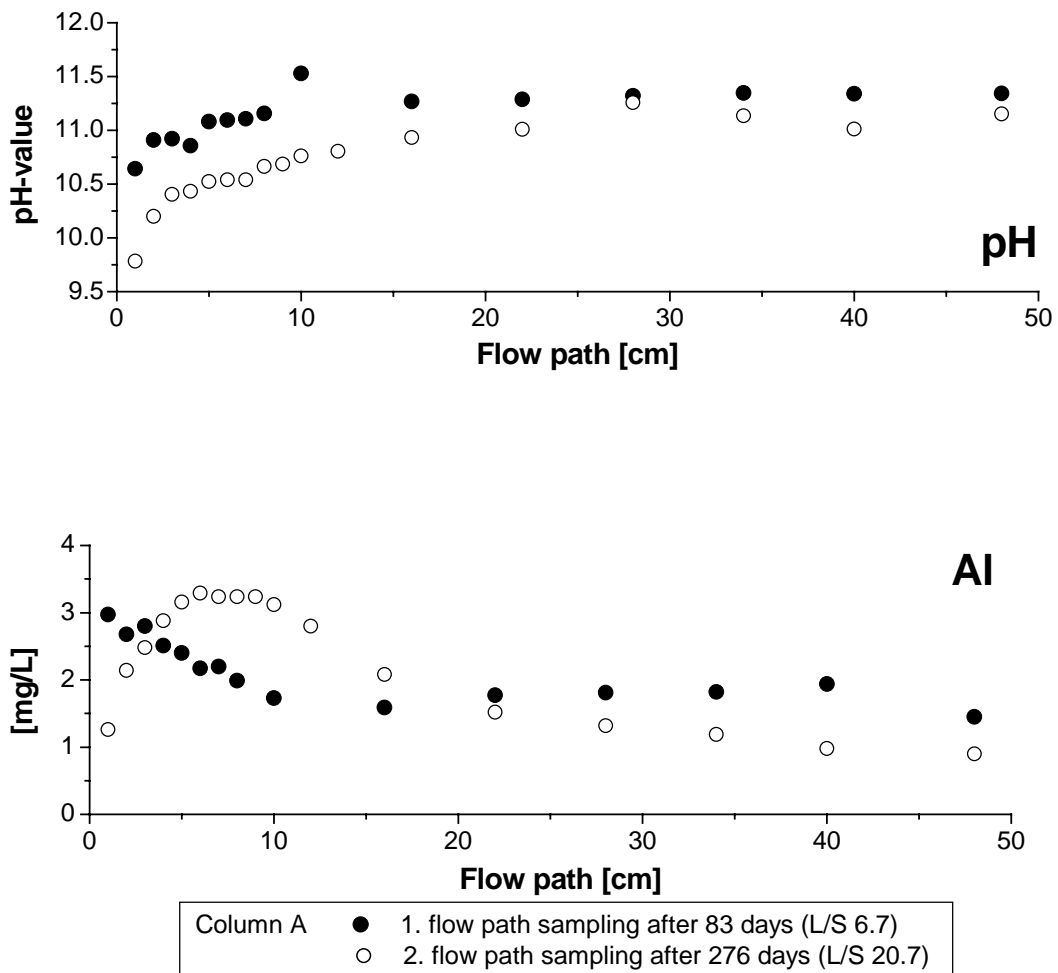


Figure 7: pH values and concentrations of aluminium in the two flow- path samples of column A

In general, aluminium and silicon showed a leaching behaviour, which was different from the other elements. Instead of a strong mobilisation in the initial phase of the experiment, aluminium and silicon displayed a maximum leaching after 5 days in column A, and 2.4 hours in column B, at a liquid/solid ratio of 0.5 L/kg respectively (Fig. 5, 6). In case of aluminium, the differences observed between the two columns were minor (Fig. 6). Upon comparing the two aluminium flow- path samples in Figure 7, it is obvious that a narrow zone in which the pH value changed began to develop within the first centimetres of column A. This reaction front migrated through the column in the course of time. The discharge of aluminium was delayed by precipitation reactions occurring along this reaction front.

Saturation indices for anorthite, kaolinite, gibbsite and calcium-montmorillonite calculated with the application of PHREEQC (version 2.8) (Parkhurst and Appelo, 1999) are displayed in Figure 8, shown in comparison with the release of aluminium along the flow-path of column A. As anorthite developed from undersaturation to a saturation index of zero, kaolinite, gibbsite and calcium-montmorillonite developed from oversaturation to a state of equilibrium. This happened within the first ten centimetres of the column and thus while the release of aluminium was at its maximum. Calcium-montmorillonite is the only phase that remained in a state of oversaturation along the whole flow-path. This is indicative of dissolution of plagioclase at the beginning of the column and a precipitation of calcium-montmorillonite.

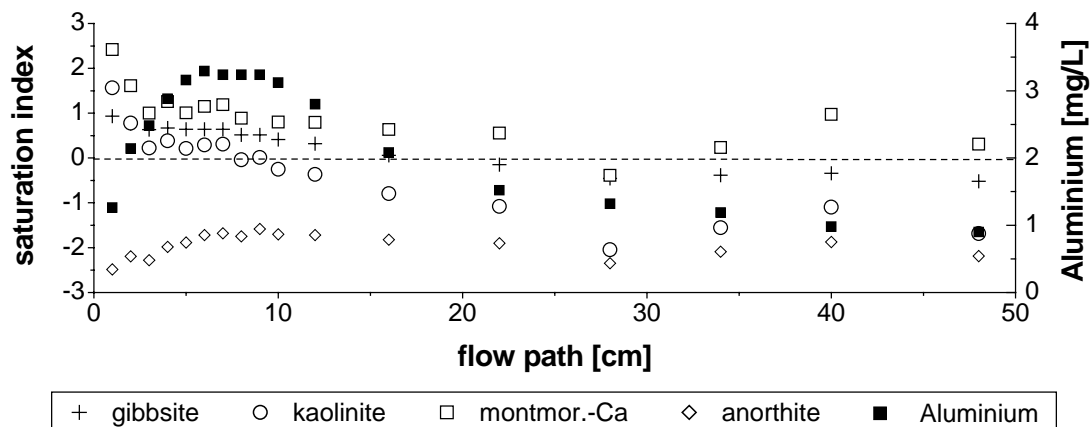


Figure 8: Concentration of aluminium along the flow-path of column A and calculated saturation indices

The activities $\log[\text{Ca}^{2+}]/[\text{H}^+]^2$ and $\log[\text{H}_4\text{SiO}_4]$ obtained upon calculating the saturation indices along the flow-path of the column were plotted into the stability field of anorthite and its possible weathering products gibbsite, kaolinite, and calcium-montmorillonite (Appelo and Postma, 1994). The activities lie in the field of calcium-montmorillonite. Dissolution/precipitation effects overlay the effect of dissolution kinetics. In spite of the by trend

slower dissolution kinetics of the primary aluminium phases both columns thus show similar results.

Flow interruption: Retention of chromium

Chromium is the only element which concentration slightly increased on account of the first two flow interruptions lasting 2 and 2.5 days. The third flow interruption lasting 26 days led to a strong retention (Fig. 9). Species and saturation index calculations with PHREEQC (version 2.8) (Parkhurst and Appelo, 1999) gave no indication as to the occurrence of any precipitation reactions. The speciation of chromium depends on the pH and the E_H conditions. Chromium is mainly present as chromium(VI) when alkaline and oxic conditions prevail in the column (Matteß, 1994; Kersten et al., 1998).

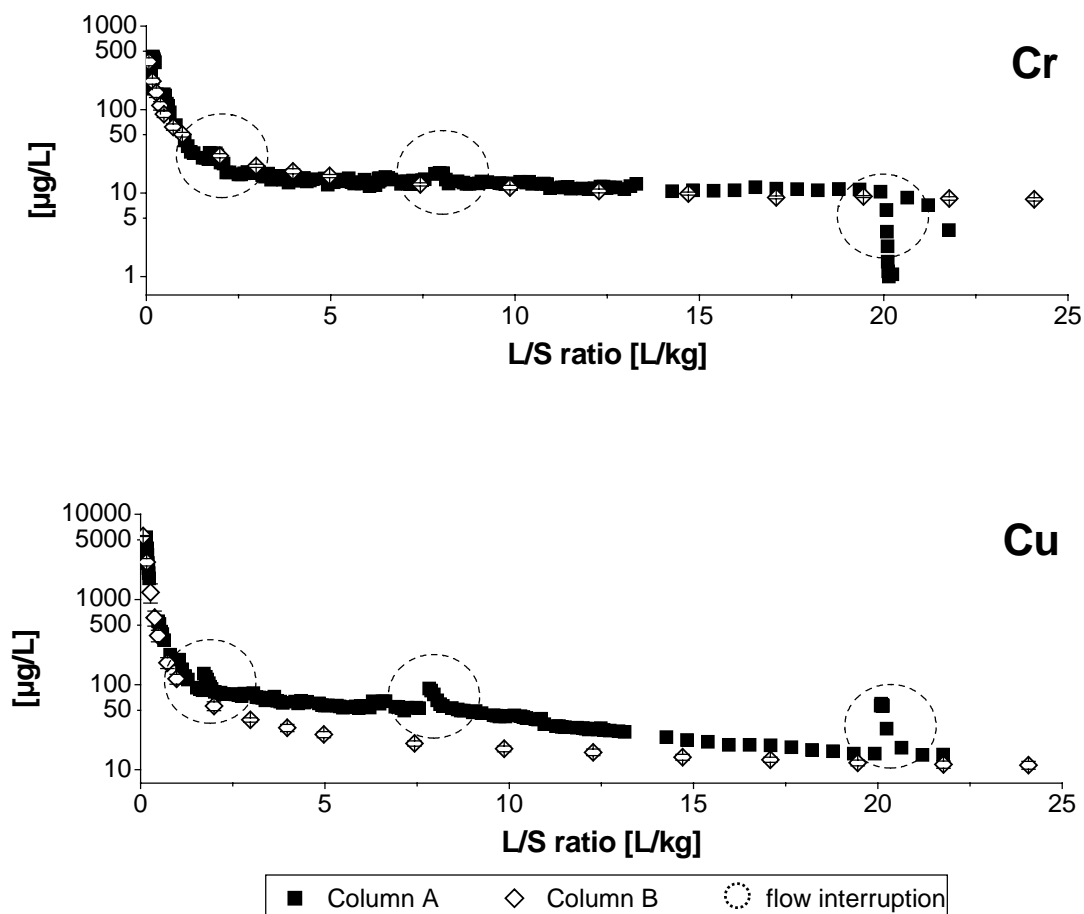


Figure 9: Concentrations of Cr and copper in the outlet samples of column A and B

This could be affirmed by thermodynamic calculations of the species distribution along the flow path as well as in the breakthrough of the column with PHREEQC (version 2.8) (Parkhurst and

Appelo, 1999). Under the prevailing alkaline conditions in the column more than 99 % of the chromium is present as chromium(VI). Thus pure electrostatic sorption is unlikely (Dzombak and Morel, 1990). The retention of chromium after the flow interruption is probably due to surface complexation processes on phases that have precipitated in the mean time, e.g. clay minerals. The fixation process of chromium also becomes apparent in the data derived from the flow- path samples (Fig. 10). After a maximum release at a flow-path of 40 cm, the concentrations of chromium decrease at the end of the column.

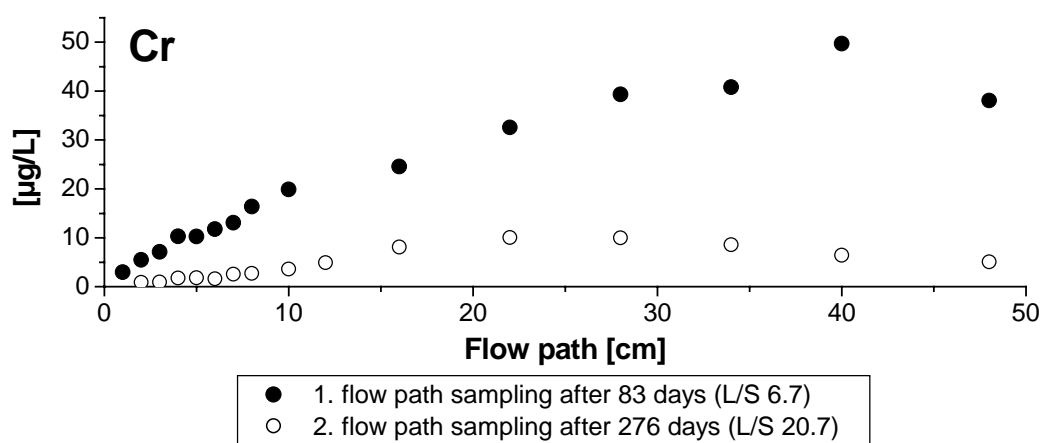


Figure 10: Concentration of chromium in the two flow- path samples of column A

Apart from the pH and the aluminium concentration (Fig 7), which appear to be unaffected by the flow interruptions, all elements except chromium showed an increased mobilisation after a flow interruption (Fig. 9). These findings are contrary to the results derived from the flow-path samples, which displayed either nearly constant or only slightly increasing concentrations for most elements and therefore indicated a state of equilibrium towards the end of the flow-path. The dissolution of phases with slower dissolution kinetics seems to be the most plausible explanation. During the normal operation of the column and at a flow velocity of the artificial rainwater of 7.3 cm/d the dissolution of the phases displaying slow dissolution kinetics was superimposed by the dissolving phases with faster dissolution kinetics.

After a liquid/solid ratio of 10 L/kg had been reached, the concentrations of copper were below the threshold value of 50 $\mu\text{g/L}$. The flow interruption after 276 days led to enhanced leaching which even resulted in concentrations exceeding the threshold value for copper (Fig. 9).

Lysimeter experiments

Not all elements underwent strong leaching in the initial phase of the lysimeter experiment as clearly as they did in the laboratory column experiments. Furthermore, differences between lysimeter A and B are visible. However, the differences between the two lysimeters or between the laboratory column experiments and the lysimeters were not greater than factor 5 as far as the ‘first flush’ concentration is concerned (Fig. 11, 12, 13).

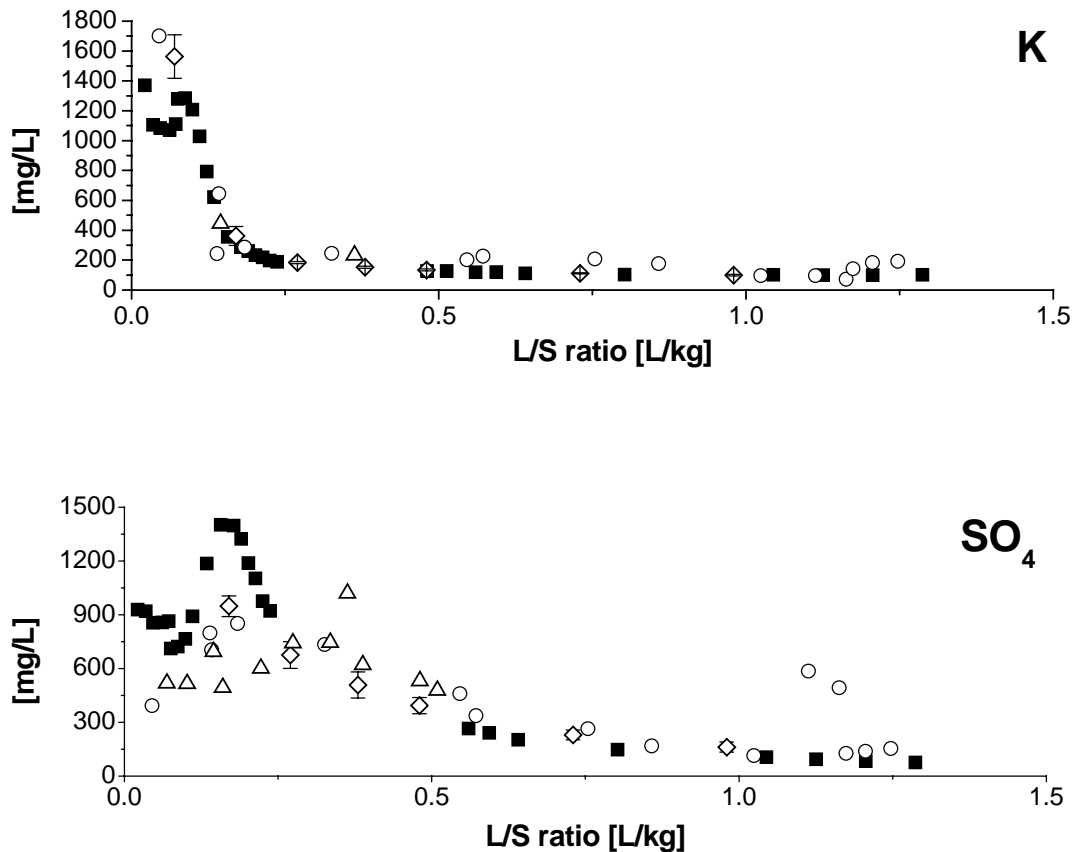


Figure 11: Concentrations of potassium and sulphate in the outlet samples of column A and B and lysimeter A and B

After a liquid/solid ratio of 0.1 L/kg, very good agreement could be observed between the laboratory and the field-scale experiments. Only the release of chromium was reduced by a factor of 4 in the lysimeter A (Fig. 12).

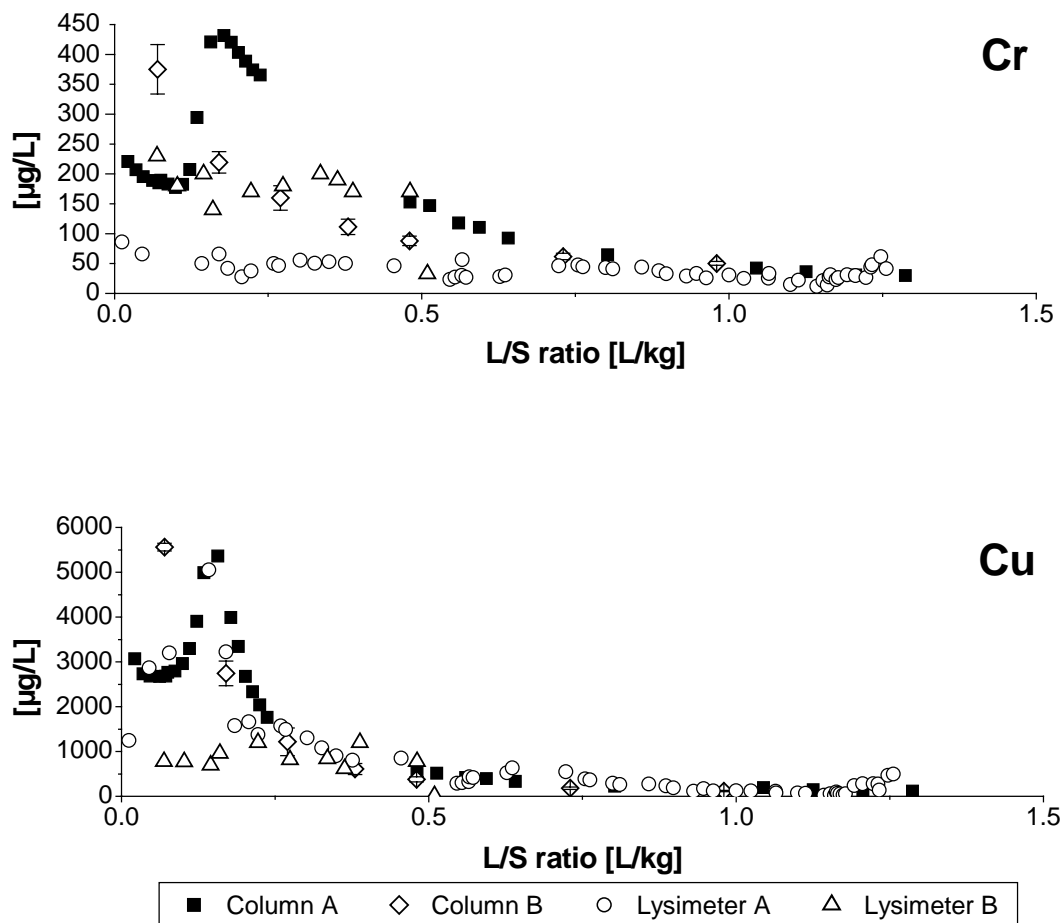


Figure 12: Concentrations of copper and chromium in the outlet samples of columns A and B and lysimeters A and B

The strong pH variations in the outlet samples of the two lysimeters, as compared to the laboratory column tests, were due to the different sampling methods (Fig. 13). The pH at the outlet of column A was measured by stream gauging, in case of column B directly after the sample was taken, a process which took about 30 minutes. In contrast, the samples of lysimeter A were exposed to air for one week, those of lysimeter B for one month. Thus carbon dioxide from the air dissolved in the samples and led to a decrease of pH.

The fluctuations of the calcium concentrations in the outlet samples of the lysimeters displayed in Figure 13 were also due to a prolonged exposure to the ambient air as compared to the column tests. The concentrations of carbon dioxide and calcium are linked to each other by the calcite-carbon dioxide equilibrium. A decrease in pH leads to a higher solubility of carbonates and hence of calcium. The calculated saturation indices indicated a supersaturation of calcite in column A (Fig. 3). Calcite was therefore likely to precipitate. Considering the similarities between the column tests and lysimeters, supersaturation of calcite was also likely to occur in the

lysimeters. Consequently, the variation of the calcium data could be due to re-dissolution processes of previously precipitated carbonates (Fig. 13).

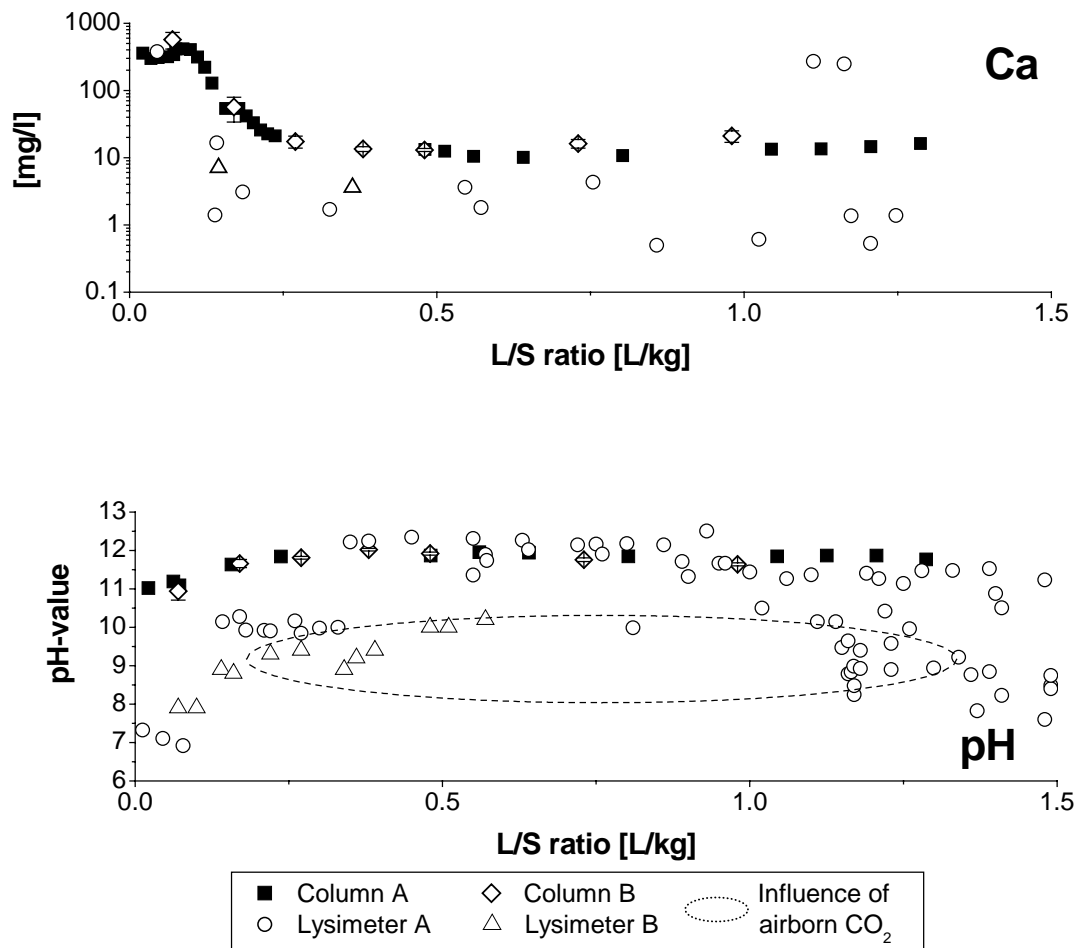


Figure 13: Concentrations of calcium and pH value in the outlet samples of columns A and B and lysimeters A and B

Batch tests

Very similar pH values were measured in the soil saturation extract (SSE) and the leaching test with deionised water (DEV S4 test) (Fig. 14). Due to the lower liquid/solid ratio of 0.25 L/kg in the soil saturation extract compared to 10 L/kg in the DEV S4 test, the element concentrations were much higher in the soil saturation extract than in the DEV S4. In order to account for the liquid/solid ratio, and to be able to describe the mobilisation and the leaching behaviour of the different contaminants, the leached amount of contaminant based on the dry mass of the material (mg/kg) should be regarded (van der Sloot, 1996; Lager, 2000).

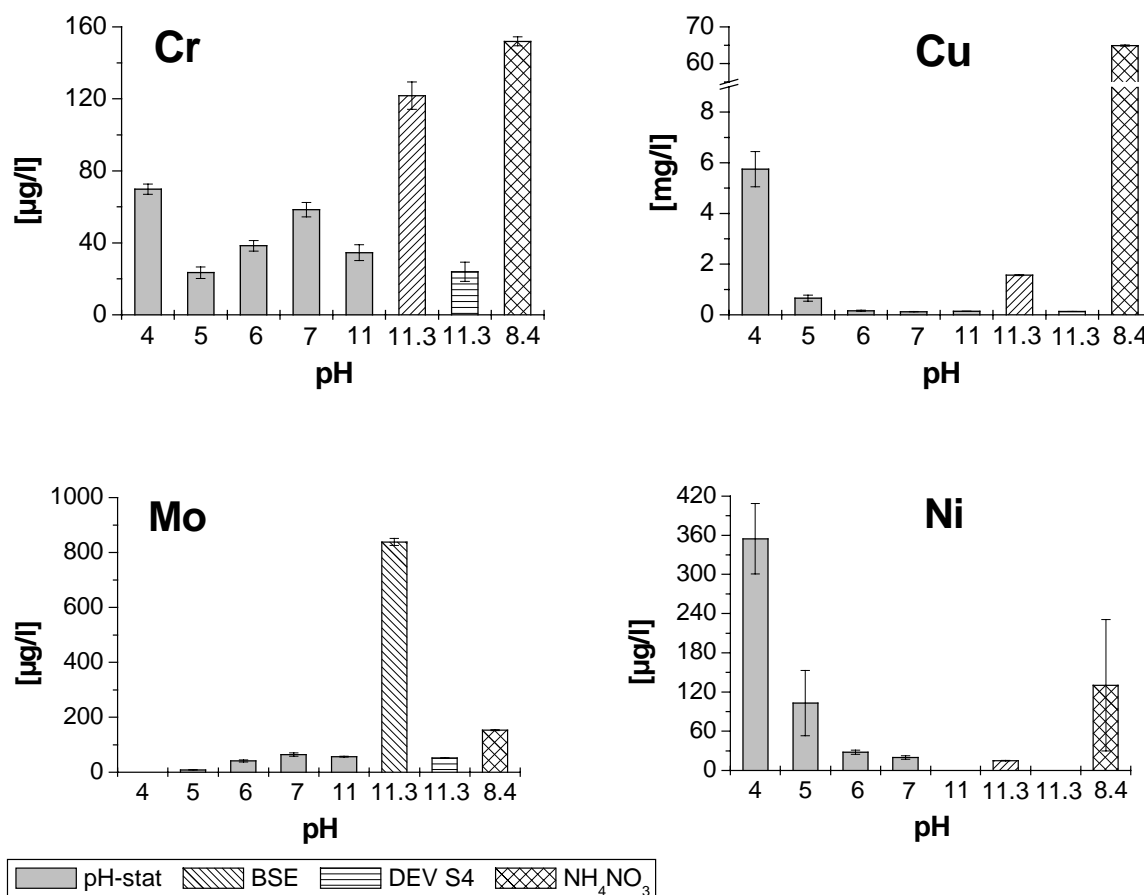


Figure 14: Concentrations of chromium, copper, molybdenum and nickel in the various batch experiments

Furthermore, the elution agent had a strong influence on mobilisation. Ammonium-nitrate is known to enhance the leaching of copper by forming hydroxo-complexes. In addition, investigations into soils contaminated with arsenic showed retention of arsenic and vanadium (Dankwarth and Gerth, 2003).

Table 7: Leached amounts based on the dry mass of the material (mg/kg) in the DEV S4 test, SSE and two column samples including their respective liquid/solid ratios

	Time [d]	L/S [L/kg]	pH	Al [mg/kg]	Ca [mg/kg]	Na [mg/kg]	Ba [$\mu\text{g/kg}$]	Cr [$\mu\text{g/kg}$]	Cu [$\mu\text{g/kg}$]	Mo [$\mu\text{g/kg}$]
Column A	2.9	0.25	11.0 - 11.9	3.1	48	839	26	72	801	385
Column B	0.06	0.27	11.8	7	49	347	35	71	793	307
SSE		0.25	11.3	0.42	43	742	21	30	393	210
Column A	128	10	11.0 - 11.9	60	542	1375	553	345	1664	649
Column B	2.1	9.9	11.4	91	528	671	574	263	1217	461
DEV S4		10	11.3	196	837	987	260	240	1319	515
pH4-stat		10	4	592	22865	1452	1727	698	57463	n.n.

Table 7 shows the leached amounts of the DEV S4 and the soil saturation extract of some elements in [mg/kg], in comparison with data derived from column A and B at the same liquid/solid ratio. A maximum factor of 2 lay between the batch test and column A at the respective liquid/solid ratio. This supports the previous assumption that both the columns and the batch experiments were in a quasi- thermodynamic state of equilibrium. With the exception of aluminium and sodium, the leached amounts of all elements in the two columns were nearly identical. In the first flush the concentration of sodium in column A was twice as high as in column B. Copper and molybdenum showed deviations of up to 30 %.

Conclusions

Batch tests can give valid information concerning the influence of various parameters, such as pH value, liquid/solid ratio, elution agent and elution time on the mobilisation of inorganic contaminants. However, a major disadvantage of all batch experiments is that they only represent a snapshot of the leaching history of the material investigated. Due to the quasi equilibrium in the column as well as in the batch tests the released amount based on the dry mass of the material (mg/kg) showed good agreement, with a deviation of less than 30 %. Yet, concentrations are relevant to assess the environmental risk of a waste material, as threshold values are defined as concentrations. The concentrations measured in the batch tests did not correlate with the results from the saturated column experiments.

Despite the different hydraulic regimes, the different pH, and composition of the elution agent in the two laboratory column tests, the results reproduced. This is only possible if the phases that show fast solution kinetics and nearly reach a state of saturation under the various hydraulic conditions are dissolved. In case of deviations, the data derived from flow-path samples can help to understand the processes within the column. Flow interruption experiments not only permit to identify a potential state of chemical non-equilibrium, but can demonstrate an enhanced leaching of some contaminants whose concentrations were initially below their respective threshold values on account of the repeated replacement of the pore volume. Thus flow interruption experiments are contributory in a complete risk assessment.

It is shown that the results of the saturated laboratory column tests were transferable to those of unsaturated field-scale lysimeters. The range of uncertainty was up to a factor of five for some elements. The overall duration of the experiments is obviously not crucial for the amount of mobilised constituents. Thus the natural conditions for depositing municipal waste incineration ashes can be simulated by a laboratory column experiment lasting less than 5 days. In general,

these findings are valid for major and trace elements that are bound to phases with fast dissolution kinetics. Materials containing phases with rather slow dissolution kinetics can only be assessed by column experiments with hydraulic regimes that are similar to natural conditions.

Acknowledgement

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4. Summarizing review and evaluation of the results

Motivation

The protection against harmful changes that can derive from the application of waste materials onto or incorporated into the soil has to be assured. In order to be able to evaluate the potential environmental risk the Federal German Soil Protection Act (Federal Bulletin, 1998) demands a seepage water prognosis. But, a practicable, valid, and generally accepted method applicable on different materials giving information for a long-term risk assessment has not yet been standardized. Reason enough for the Federal Ministry for Education and Science (BMBF) to fund a project called 'seepage water prognosis'. 41 institutions from all over Germany worked on the aim of either finding methods for the determination of the source strength, which is defined as the total mass flow caused by natural leaching or models to describe the contaminants transport from the contaminated material through the underlying unsaturated zone into the groundwater. Objective of the workings that led to this thesis was to find an applicable, valid and generally accepted laboratory method for the determination of the source strength.

Scope of workings

The four materials investigated in this project were the sandy and fine fractions of sediment dredged from the port of Bremen-City, municipal waste incineration ash and demolition waste. The Federal Department of Material Investigation (BAM) generated latter two as reference materials. Material parameters including mineral phase analysis were determined. Four batch tests, DEV S4 test, soil saturation extract, ammonium-nitrate extract and pH-stat test (at pH 4, 5, 6, 7, 11) were performed with all four materials. The pore water of the fine fraction of harbour sediments was gained with a modified centrifugation approach. Saturated column tests were run with the municipal waste incineration ash, the demolition waste and the sandy fraction of harbour sediments. The sandy fraction of the harbour sediments was also incorporated in an unsaturated column unit. Main elements, heavy metals including arsenic, molybdenum and vanadium, the anions chloride, nitrate and sulphate, inorganic carbon and dissolved organic carbon were measured in all samples taken. Additionally electrical conductivity, pH and E_H value were determined.

Source strength

When starting this project in 2001 it seemed almost impossible to find a method that is valid, meaning that can give sufficient information about the leaching of a material in the foreseeable future (Federal Bulletin, 1998) and thus, enabling a detailed risk assessment, and also be generally accepted and practicable (Oberacker and Eberle, 2002). Numerous scientists have applied various batch tests on various materials within the last 10 years (e.g. Obermann and Cremer, 1992; van der Sloot, 1998; van der Sloot et al., 2000; Wahlström et al., 2000; Townsend et al., 2003) in order to find a suitable method. The two main advantage of the project ‘seepage water prognosis’ compared to former investigations were the cooperation of 41 institutions throughout Germany, all applying at least two of the three provided reference materials. Secondly, the results of all laboratory methods could be compared with field-scale lysimeters performed by three of the participants. Thus reference values, meaning values for the source strength as well as the concentration at the place of assessment that represent the natural leached concentrations were available.

The leachate concentration in the field is influenced by a number of parameters, such as pH conditions, redox potential, liquid/solid ratio, ionic strength, and presence of complexants. For each parameter a batch test is available that can give information concerning the influence of the parameter on the mobilisation of inorganic contaminants. A major disadvantage of all batch experiments is that they only represent a snapshot of the leaching history of the material investigated. A simple batch test might be useful as a preliminary test. But, for time-resolved and detailed information, column leaching tests should be chosen.

Despite the different hydraulic regimes and time scales, the different pH values and compositions of the elution agent in the two saturated laboratory column tests applied, good agreement could be observed for both municipal waste incineration ash and demolition waste regarding most parameters. This is only possible, if phases are dissolved that show high dissolution kinetics and nearly reach the stage of saturation under the different respective hydraulic conditions. However, there are exceptions such as silicon and aluminium in the municipal waste incineration ash. Here data of flow-path samplings can help to understand the processes within the column. In addition, flow interruption experiments were shown to be a helpful instrument for a complete risk assessment. For some contaminants a flow interruption can lead to an enhanced leaching, resulting in an exceeding of the respective threshold value.

The strong mobilisation of almost all investigated elements in the breakthrough of the column tests in the initial stage (‘first flush’), a phenomenon that is also occurring under natural

conditions of deposition, can only be revealed in column tests. It was shown that the leaching in the first flush is potentially even stronger under unsaturated conditions, if sulphide oxidation occurs, which results in a decrease of pH. However, apart from the first flush saturated and unsaturated columns with the sandy fraction of harbour sludge showed good accordance for most parameters.

It is shown that the results of the saturated laboratory column tests were transferable to those of unsaturated field-scale lysimeters conducted with municipal waste incineration ash and demolition waste. The range of uncertainty was up to a factor of five for some elements as far as the initial leaching is concerned. However, the processes in a deposition of municipal waste incineration ash or demolition waste can be simulated by a laboratory column experiment lasting less than 5 days. In general, these findings are valid for major and trace elements that are bound to phases with a fast dissolution kinetic. The column test can be regarded as a practicable tool to assess the time-dependend release of inorganic compounds under natural conditions. The low time requirements correspond with the needs of authorities or laboratories to have an easy to use and cheap, but nevertheless reliable instrument for a sustainable environmental risk assessment.

5. Outlook

With the column test lasting less than five days the demands on the method for the determination of the source strength to be valid and practicable can be accomplished. In order to be generally accepted a standardisation of the column test is necessary. But, before a method can be standardised, it has to undergo a lot of test procedures and has to be applied to various materials, in order to gain information about the boundary conditions and the limitations of the method. A next step should be the performance of the short-term column test with other waste materials like the sandy fraction of harbour sediments, as results of long term saturated and unsaturated column tests are available. Furthermore, it should be investigated, if a simple batch test as a preliminary test, when the decision on the reuse of the material is merely limited to a comparison of the source strength with threshold values, can be considered for other material than demolition waste as well.

Next to inorganic contaminants a similar approach should be considered for organic contaminants. There are attempts within this BMBF-project e.g. from the North Rhine-Westphalia State Environment Agency (LUA NRW) in Essen to design a column test suitable for the examination of both inorganic and organic contaminants. However, to measure organic substances in a range of 10-100 ng/L a sample volume of 1 L is necessary. The analytical need for huge sample volumes leads to decrease of time resolution in the breakthrough curves. Further investigations are necessary.

Materials with a coefficient of hydraulic conductivity of less than 10^{-6} cm/s the performance of column experiments is not possible (DIN, 1998). The pore water of moist recycling material like the fine fraction of harbour sediments comes close to the source strength. For dry materials diffusion test should be employed.

As a next step for the risk assessment in the frame of the seepage water prognosis a transport prognosis has to be made with the source strength as the main input parameter. For this purpose my colleague Veith Becker developed a computer model called SEEPER. It is a tool for modelling reaction and transport processes in the unsaturated zone from the place of sampling (source strength) to the place of assessment, the transition area between unsaturated and saturated zone. By using transfer functions for most processes a practicable and valid model was developed.

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Abbreviations

n.d. not determined

A 1. Data for Chapters 2.1., 2.2. and 3.1.

A 1.1. Harbour sediments: Material description

A 1.1.1. Material parameter

		Sandy fraction	Fine fraction
Water content	[% weight]	~ 5	~ 34
Specific weight	[g/cm ³]	2.61	2.42
Specific surface	[m ² /g]	2.4	16.2
pH		~ 8.3	7.0 – 7.6
E _H	[mV]	~ 250	30 – 420
El. conductivity	[mS/cm]	1.5	0.54
Total carbon	[%]	0.3	4.6
Inorganic carbon	[%]	0.07	0.46
Organic carbon	[%]	0.23	4.14

A 1.1.2. Bulk concentration

Element	Harbour sediments			
	Sandy fraction		Fine fraction	
	Microwave digestion	RFA	Microwave digestion	RFA
Al	12676	13197	54758	58707
Ca	3452	7266	21976	22900
Fe	8358	9040	38872	38757
K	8819	8920	17580	17173
Mg	815	2730	8300	11187
Na	2329	n.d.	4016	n.d.
P	n.d.	1006	2290	3109
S	803	2517	4848	5165
Si	n.d.	337833	n.d.	223833
Cl	n.d.	n.d.	n.d.	179
As	1.4	1.0	12	15.3
Ba	219	239	490	386
Cd	0.91	n.d.	4.1	6.0
Co	4.2	n.d.	20	28
Cr	11	37	90	95
Cu	9.3	8.0	65	56
Mn	253	376	1136	1184
Mo	1.5	n.d.	1.5	n.d.
Ni	n.d.	4.4	41	37
Pb	12	13	75	95
Sr	40	40	150	132
V	n.d.	22	n.d.	146
Zn	53	42	487	446

A 1.2. Data of the batch experiments with the fine fraction of harbour sediments

A 1.2.1. Electric conductivity, pH- and E_H value and acid neutralisation capacity

Sample ID	el. cond. [mS/cm]	pH	E_H [mV]	ANC/BNC [meq/kg]
pH4-stat	8.08	4	214	612
pH5-stat	6.96	5	464	501
pH6-stat	5.48	6	451	362
pH7-stat	2.14	7	459	89
pH11-stat	1.60	11	221	231
NH ₄ NO ₃	64	7	365	n.d.
SSE	3.4	7.2	450	n.d.
Cen	5	7.3	450	n.d.
DEV S4	0.50	7.6	510	n.d.

A 1.2.2. Main elements

Sample ID	Al [µg/L]	St.Dev. [µg/L]	Ca [mg/L]	St.Dev. [mg/L]	Fe [µg/L]	St.Dev. [mg/L]	K [mg/L]	St.Dev. [mg/L]
pH4-stat	1776	158	1615	25	11996	1724	34	1.8
pH5-stat	67	11	1388	38	198	23	28	1.1
pH6-stat	27	3.6	1084	13	8.0	11	25	0.25
pH7-stat	4.21	0.48	381	40	92	154	19	2.1
pH11-stat	586	147	8.3	0.14	866	148	4.3	0.09
NH ₄ NO ₃	20	5.4	1897	21	25	3.3	141	1.8
SSE	36	14	624	4.6	33	23	22	0.40
Cen	113	15	904	25	18		37	2.2
DEV S4	17	10	91	4.7	6.8	3.0	7.3	0.06

Sample ID	Mg [mg/L]	St.Dev. [mg/L]	Na [mg/L]	St.Dev. [mg/L]	S [mg/L]	St.Dev. [mg/L]	Si [mg/L]	St.Dev. [mg/L]
pH4-stat	143	0.87	29	1.6	106	7.5	35	0.14
pH5-stat	116	2.7	27	0.37	119	5.3	22	0.88
pH6-stat	65	1.2	24	0.12	118	8.1	11	0.15
pH7-stat	41	2.0	23	0.21	120	1.3	7.7	0.10
pH11-stat	0.15	0.01	312	53	79	13	36	5.3
NH ₄ NO ₃	236	1.2	104	0.83	380	29	7.6	0.12
SSE	130	2.4	117	3.3	596	10	6.9	0.06
Cen	219	8.2	264	15	773	23	n.d.	
DEV S4	15	0.45	14	0.10	73	2.5	5.2	0.03

A 1.2.3. Trace elements

Sample ID	As [µg/L]	St.Dev. [µg/L]	Ba [µg/L]	St.Dev. [µg/L]	Cd [µg/L]	St.Dev. [µg/L]	Co [µg/L]	St.Dev. [µg/L]	Cr [µg/L]	St.Dev. [µg/L]
pH4-stat	<10		739	46	112	7.2	118	9.1	102	8.0
pH5-stat	<10		447	19	35	1.6	30	1.8	53	4.1
pH6-stat	<10		303	17	8.1	0.22	<2		16	2.7
pH7-stat	<10		119	12	<4		<2		<2	
pH11-stat	170	16	4.3	0.8	4.7	0.53	<2	0.18	8.0	0.72
NH ₄ NO ₃	<10		2138	89	6.0	0.08	<2		7.0	3.5
SSE	<10		31	1.7	<4		<2		13	
Cen	<10		46	2.7	<4		<2		<2	
DEV S4	<10		47	2.5	<4		<2		<2	

Sample ID	Cu [µg/L]	St.Dev. [µg/L]	Mn [µg/L]	St.Dev. [µg/L]	Mo [µg/L]	St.Dev. [µg/L]	Ni [µg/L]	St.Dev. [µg/L]	Pb [µg/L]	St.Dev. [µg/L]
pH4-stat	64	4.2	39287	95	<2		381	22	6.6	0.08
pH5-stat	8.3	1.6	17478	793	<2		146	5.4	<4	
pH6-stat	5.3	0.58	3784	317	<2		33	0.58	<4	
pH7-stat	4.8	0.27	46	16	<2		4.3	0.88	<4	
pH11-stat	393	47	7.0	1.3	3.3	0.11	29	2.2	4.6	0.40
NH ₄ NO ₃	40	1.3	457	10	<2		24	2.7	<4	
SSE	7.8	2.1	31	5.7	<2		8.9	0.90	<4	
Cen	60	34	94	24	<2		23	4.7	<4	
DEV S4	4.7	0.12	9.0	1.1	<2		2.3	0.12	<4	

Sample ID	Sr [µg/L]	St.Dev. [µg/L]	V [µg/L]	St.Dev. [µg/L]	Zn [µg/L]	St.Dev. [µg/L]
pH4-stat	5728	52	<2		21186	59
pH5-stat	4900	119	<2		7088	288
pH6-stat	3907	39	<2		1062	28
pH7-stat	1619	146	2	0.09	68	8.4
pH11-stat	21	3.1	343	48	36	4.0
NH ₄ NO ₃	11471	46	<2		164	1.7
SSE	2585	41	<2		208	86
Cen	4251	138	<2		299	117
DEV S4	442	20	2.9	0.08	79	46

A 1.3. Data of the batch experiments with the sandy fraction of harbour sediments

A 1.3.1. Electric conductivity, pH and E_H value and acid neutralisation capacity

Sample_ID	el. cond. [mS/cm]	pH	E _H [mV]	ANC/BNC [meq/kg]
pH4-stat	1.1	4	n.d.	91
pH5-stat	0.72	5	494	50
pH6-stat	0.41	6	436	21
pH7-stat	0.38	7	436	40
pH11-stat	0.77	11	248	55
NH ₄ NO ₃	80	7.1	38	n.d.
SSE	n.d.	7.9	n.d.	n.d.
DEV S4	1.5	8.3	580	n.d.

A 1.3.2. Main elements

Sample_ID	Al	St.Dev.	Ca	St.Dev.	Fe	St.Dev.	K	St.Dev.
	[µg/L]	[µg/L]	[mg/L]	[mg/L]	[µg/L]	[mg/L]	[mg/L]	[mg/L]
pH4-stat	643	24	184	9.9	936	84	12	1.2
pH5-stat	51	6.8	112	19	50	4.5	14	0.31
pH6-stat	26	10.0	61	3.1	17	2.5	14	0.70
pH7-stat	24	5.0	60	5.7	23	3.8	14	0.25
pH11-stat	1052	34	3.1	0.24	332	44	12	1.9
NH ₄ NO ₃	25	0.89	320	9.1	32	10	8.2	
SSE	12	8.6	571	7.2	15	2.5	16	0.08
DEV S4	21	9.3	22	2.2	19	4.4	1.2	

Sample_ID	Mg	St.Dev.	Na	St.Dev.	S	St.Dev.	Si	St.Dev.
	[mg/L]	[mg/L]	[mg/L]	[mg/L]	[mg/L]	[mg/L]	[mg/L]	[mg/L]
pH4-stat	9.9	1.0	1.6	0.06	12	0.48	4.9	0.23
pH5-stat	6.0	1.5	1.4	0.21	15	1.0	3.4	0.10
pH6-stat	3.4	0.12	1.3	0.03	15	0.06	2.4	0.14
pH7-stat	3.0	1.5	2.1	2.3	15	11	2.0	0.16
pH11-stat	0.19	0.13	128	9.6	16	0.13	7.1	0.10
NH ₄ NO ₃	12.13	0.52	<0.1		55	2.0	2.5	0.02
SSE	61	1.8	49	4.3	486	19	5.6	0.25
DEV S4	1.6	0.09	1.1	0.02	12	0.70	1.3	0.35

A 1.3.3. Trace elements

Sample_ID	As	St.Dev.	Ba	St.Dev.	Cd	St.Dev.	Co	St.Dev.	Cr	St.Dev.
	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]
pH4-stat	<10		481	33	8.3	0.64	21	1.8	29	3.8
pH5-stat	<10		231	29	<4	0.33	3		15	2.1
pH6-stat	<10		97	3.8	<4		<2		11	4.9
pH7-stat	<10		67	3.3	<4		<2		5.2	2.2
pH11-stat	25	2.3	1.9	0.01	<4		<2		6.7	0.46
NH ₄ NO ₃	<10		1728	29	<4		<2		12	1.6
SSE	<10		36	8.3	<4		<2		2.9	
DEV S4	<10		21	1.5	<4		<2		<2	

Sample_ID	Cu	St.Dev.	Mn	St.Dev.	Mo	St.Dev.	Ni	St.Dev.	Pb	St.Dev.
	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]
pH4-stat	67	7.1	7082	600	<2		58	4.0	13	1.2
pH5-stat	7.6	0.59	3044	348	<2		26	2.0	<4	
pH6-stat	2.1		779	12	<2		8.8	1.2	<4	
pH7-stat	2.1	0.57	76	22	<2		3.7	0.94	<4	
pH11-stat	55	3.7	12	2.6	2.2		3.6		<4	
NH ₄ NO ₃	62	3.3	2131	133	4.7	0.16	17	0.58	<4	
SSE	7.4	1.5	16	0.21	<2		6.0	2.7	<4	
DEV S4	3.2	0.6	6.8	3.4	<2		<2		<4	

Sample_ID	Sr [µg/L]	St.Dev. [µg/L]	V [µg/L]	St.Dev. [µg/L]	Zn [µg/L]	St.Dev. [µg/L]
pH4-stat	533	22	<2		1553	132
pH5-stat	381	44	<2		702	107
pH6-stat	235	8.8	<2		122	12
pH7-stat	212	24	<2		20	1.2
pH11-stat	10	0.70	50	1.3	11	3.8
NH ₄ NO ₃	1388	43	<2		72	1.6
SSE	1854	15	<2		150	120
DEV S4	82	7.5	<2	0.21	26	3.8

A 1.4. Data of the saturated column experiments with the sandy fraction of harbour sediments

A 1.4.1. Characteristic data of the column

Length	[cm]	48
Diameter	[cm]	5
Volume	[cm ³]	942
Amount material original	[kg]	1.822
Amount material dry	[kg]	1.703
Amount water in column	[L]	0.29
Vol. inlet/outlet	[L/d]	0.16
Specific weight	[g/cm ³]	2.61
Absolut porosity		0.31
Specific porosity		0.3
Watersaturation	[% abs. porosity]	99.8
kf-value	[m/s]	$8.3 \cdot 10^{-5}$
Vf (flow rate 0.11 mL/min)	[cm/d]	8
Va (flow rate 0.11 mL/min)	[cm/d]	26

A 1.4.2. Outlet

A 1.4.2.1. Liquid/solid ratio, electric conductivity, pH and E_H value, inorganic carbon, dissolved organic carbon and anions

Time [d]	L/S [L/kg]	el. cond. [mS/cm]	pH	E _H [mV]	IC [mg/L]	DOC [mg/L]	Cl [mg/L]	NO ₃ [mg/L]	SO ₄ [mg/L]
0.23	0.02	29	7.1	457	14	45	98	681	2016
0.40	0.03	29	7.3	451	13	10	74	518	1871
0.57	0.05	n.d.	n.d.	n.d.	15	9.8	79	566	1988
0.72	0.06	n.d.	n.d.	n.d.	15	9.1	84	590	2015
0.86	0.07	n.d.	n.d.	n.d.	15	9.3	90	646	2037
1.01	0.08	28	7.0	457	16	9.4	81	570	1971
1.17	0.10	28	7.1	488	16	9.1	70	513	1928
1.34	0.11	27	6.9	469	18	8.9	61	487	2029
1.53	0.13	n.d.	n.d.	n.d.	19	8.9	47	324	1823
1.67	0.14	n.d.	n.d.	n.d.	20	8.6	16	167	2153
1.82	0.15	n.d.	n.d.	n.d.	20	7.6	<0.1	48	2227
1.99	0.17	n.d.	n.d.	n.d.	20	7.0	<0.1	7.2	2187
2.13	0.18	24	7.6	430	20	6.8	<0.1	<0.1	2122

Time [d]	L/S [L/kg]	el. cond. [mS/cm]	pH	E _H [mV]	IC [mg/L]	DOC [mg/L]	Cl [mg/L]	NO ₃ [mg/L]	SO ₄ [mg/L]
2.30	0.19	n.d.	n.d.	n.d.	22	6.9	<0.1	<0.1	2220
2.43	0.20	n.d.	n.d.	n.d.	24	7.2	<0.1	<0.1	2183
2.57	0.21	n.d.	n.d.	n.d.	27	6.5	<0.1	<0.1	2007
2.69	0.22	n.d.	n.d.	n.d.	29	6.4	<0.1	<0.1	1918
2.86	0.24	n.d.	n.d.	n.d.	30	6.3	<0.1	<0.1	1669
3.01	0.25	n.d.	n.d.	n.d.	32	6.5	n.d.	n.d.	n.d.
3.15	0.26	n.d.	n.d.	n.d.	31	7.1	<0.1	<0.1	964
3.30	0.27	7.4	7.8	451	31	6.7	n.d.	n.d.	n.d.
3.47	0.29	n.d.	n.d.	n.d.	32	6.7	0.24	<0.1	218
3.61	0.30	n.d.	n.d.	n.d.	31	6.7	0.65	<0.1	134
3.76	0.31	n.d.	n.d.	n.d.	31	6.7	1.4	<0.1	85
3.90	0.32	n.d.	n.d.	n.d.	30	5.9	1.2	<0.1	79
4.05	0.34	3.2	7.8	431	29	n.d.	0.86	<0.1	55
5.15	0.43	2.9	7.7	396	26	5.6	0.76	0.31	32
5.95	0.50	2.8	7.8	407	27	5.3	1.1	0.21	33
6.24	0.52	2.8	7.8	354	26	5.2	1.8	0.52	31
6.94	0.58	2.9	7.8	379	26	5.0	1.2	<0.1	33
7.88	0.66	n.d.	n.d.	n.d.	24	4.2	1.1	<0.1	27
8.16	0.68	3.5	7.8	380	23	4.3	1.6	<0.1	24
9.42	0.78	2.4	7.7	374	24	4.9	3.0	<0.1	26
10.15	0.84	n.d.	n.d.	n.d.	24	4.5	1.5	<0.1	27
11.15	0.93	7.1	7.5	380	21	4.3	1.5	<0.1	23
11.42	0.95	n.d.	n.d.	n.d.	21	4.2	1.5	<0.1	24
12.15	1.01	2.5	7.6	290	20	4.0	1.4	<0.1	24
12.57	1.05	n.d.	n.d.	n.d.	21	3.9	2.0	<0.1	24
13.15	1.09	2.2	7.6	288	20	3.9	2.6	<0.1	26
13.57	1.13	n.d.	n.d.	n.d.	21	4.0	2.1	<0.1	25
14.15	1.18	2.2	7.6	234	19	3.8	2.1	<0.1	23
16.15	1.34	2.1	7.5	214	19	3.8	1.8	<0.1	24
19.15	1.59	2.1	7.4	202	20	n.d.	1.4	<0.1	24
20.15	1.68	2.0	7.3	207	23	n.d.	1.3	<0.1	26
21.15	1.76	2.0	7.3	218	18	4.3	1.1	<0.1	21
22.15	1.84	1.9	7.3	218	18	4.5	1.2	<0.1	17
25.15	2.09	1.8	7.2	200	16	3.6	1.8	<0.1	17
26.15	2.18	1.8	7.3	236	17	3.7	1.6	<0.1	17
27.15	2.26	1.8	7.3	238	16	3.5	2.1	0.11	15
29.15	2.43	1.7	7.4	244	n.d.	n.d.	n.d.	n.d.	n.d.
32.15	2.67	1.7	7.4	406	15	3.7	1.3	<0.1	16
33.15	2.76	1.7	7.3	323	17	4.0	1.4	<0.1	16
34.15	2.84	1.6	7.2	369	15	4.0	1.5	<0.1	17
35.15	2.92	1.6	7.2	371	14	3.8	1.6	<0.1	18
36.15	3.01	1.5	7.2	383	14	3.5	1.9	<0.1	18
39.15	3.26	1.5	7.2	405	14	3.6	1.9	<0.1	18
40.15	3.34	1.5	7.2	401	14	3.7	0.99	<0.1	18
41.15	3.42	1.5	7.2	401	16	3.8	1.5	<0.1	17
42.15	3.51	1.5	7.1	403	13	3.5	1.1	<0.1	17
43.15	3.59	1.5	7.1	396	13	3.3	0.84	<0.1	17
46.15	3.84	1.3	7.2	411	13	3.4	1.5	<0.1	16
47.15	3.92	1.5	7.2	420	13	3.4	1.6	<0.1	16
48.15	4.01	1.4	7.2	420	14	3.4	1.9	<0.1	15
49.15	4.09	1.5	7.2	412	13	3.5	1.1	<0.1	17
50.15	4.17	1.5	7.4	400	13	3.3	1.5	<0.1	16
53.15	4.42	1.4	7.2	422	13	3.2	0.99	<0.1	15
54.15	4.51	1.4	7.1	388	2.5	3.4	0.89	<0.1	17
55.15	4.59	1.4	7.4	405	11	3.3	0.96	<0.1	15
56.01	4.66	1.4	7.1	356	11	3.2	1.1	<0.1	17
60.15	5.00	1.4	7.1	378	11	3.6	1.1	<0.1	19
61.15	5.09	1.3	7.1	422	12	n.d.	0.76	<0.1	15

Time [d]	L/S [L/kg]	el. cond. [mS/cm]	pH	E _H [mV]	IC [mg/L]	DOC [mg/L]	Cl [mg/L]	NO ₃ [mg/L]	SO ₄ [mg/L]
62.15	5.17	1.3	7.1	454	n.d.	6.4	0.94	<0.1	14
63.15	5.25	1.3	7.3	387	12	3.4	1.0	<0.1	14
64.15	5.34	1.3	7.3	446	12	3.5	0.81	<0.1	14
68.15	5.67	1.3	7.2	486	17	3.3	0.87	<0.1	15
69.15	5.75	1.3	7.0	407	12	3.4	0.82	<0.1	15
70.15	5.84	1.3	7.1	458	12	3.4	0.68	<0.1	14
71.15	5.92	1.3	7.0	413	12	3.2	1.1	<0.1	14
74.15	6.17	1.4	7.6	439	12	3.9	0.78	<0.1	14
75.15	6.25	1.3	7.5	400	11	3.1	0.99	<0.1	15
76.22	6.34	1.3	7.5	435	11	2.9	0.73	<0.1	14
77.15	6.42	1.3	7.8	368	11	2.9	1.2	<0.1	14
78.01	6.49	1.3	7.5	438	11	3.1	0.30	<0.1	8
80.57	6.70	1.3	7.5	392	11	3.1	0.72	0.13	13
82.15	6.83	1.3	7.6	400	11	3.0	1.0	<0.1	14
83.15	6.92	1.3	7.8	401	11	2.9	0.71	<0.1	13
84.15	7.00	1.3	7.5	401	11	3.3	0.78	<0.1	14
85.15	7.08	1.2	7.5	397	11	3.3	0.73	<0.1	13
89.13	7.30	1.4	7.4	405	9.8	3.3	0.90	<0.1	15
95.15	7.80	1.2	7.4	398	11	2.6	0.78	<0.1	14
96.15	7.88	1.2	7.3	444	10	3.3	0.80	<0.1	15
97.26	7.98	1.2	7.3	416	10	2.8	0.68	<0.1	14
98.15	8.05	1.2	7.4	438	9.9	2.7	1.3	<0.1	15
99.17	8.14	1.2	7.7	423	10	2.9	0.57	<0.1	13
102.15	8.38	1.3	7.3	n.d.	11	2.4	0.87	<0.1	15
103.15	8.47	1.2	7.3	n.d.	10	2.8	0.73	<0.1	14
105.15	8.63	1.2	7.4	n.d.	10	2.5	0.82	<0.1	12
106.17	8.72	1.2	7.3	n.d.	11	2.5	0.84	<0.1	12
109.15	8.97	1.2	7.3	n.d.	10	2.4	1.0	<0.1	14
110.15	9.05	1.2	7.3	n.d.	2.7	2.7	0.74	<0.1	12
111.15	9.13	1.2	7.4	n.d.	10	2.4	0.77	<0.1	12
112.15	9.22	1.2	7.2	n.d.	10	2.5	1.0	<0.1	15
113.17	9.30	1.2	7.4	n.d.	10	2.4	1.5	<0.1	15
116.15	9.55	1.2	7.4	n.d.	10	2.8	1.0	<0.1	14
118.15	9.71	1.2	7.1	450	10.3	3.6	1.2	n.d.	14
119.15	9.80	1.3	7.3	383	10	2.3	1.0	<0.1	16
120.17	9.88	1.2	7.5	377	10	2.6	1.2	<0.1	16
123.15	10.13	1.2	7.3	413	10	2.3	1.2	<0.1	16
124.15	10.21	1.2	7.1	412	10	2.5	n.d.	n.d.	n.d.
126.15	10.38	1.2	7.2	420	10	2.3	1.4	<0.1	16
130.15	10.71	1.2	7.0	n.d.	10.0	2.6	1.3	<0.1	14
131.15	10.80	1.2	7.1	431	9.7	2.4	0.84	<0.1	6
132.15	10.88	1.2	7.1	435	9.8	2.2	0.81	<0.1	13
133.15	10.96	1.2	7.1	443	10	2.6	0.70	<0.1	13
134.17	11.05	1.2	7.2	374	10	4.0	1.6	<0.1	14
137.15	11.30	1.2	7.1	336	9.5	2.6	0.79	<0.1	13
138.15	11.38	1.2	7.1	342	9.5	3.0	2.9	<0.1	15
139.15	11.46	1.2	7.2	333	10.0	3.4	1.4	<0.1	14
140.15	11.55	1.2	7.1	370	10	3.0	1.7	<0.1	15
141.15	11.63	1.2	7.1	358	9.8	2.9	0.60	<0.1	14
145.15	11.96	1.3	7.7	335	7.6	2.5	1.2	<0.1	16
151.15	12.46	1.2	7.7	394	9.6	3.1	4.6	<0.1	14
153.15	12.63	1.5	7.6	391	10	3.3	10	<0.1	14
158.15	13.04	1.5	7.5	378	10	2.7	9.1	<0.1	14
161.15	13.29	1.5	7.5	397	11	2.7	8.7	<0.1	15
162.15	13.38	1.5	7.5	396	10	3.1	9.5	<0.1	17
165.15	13.62	1.5	7.5	416	10	4.8	8.2	<0.1	15
167.15	13.79	1.5	7.4	406	10	9.2	8.0	<0.1	14
169.15	13.96	1.4	7.4	395	10	3.5	8.7	<0.1	14

Time [d]	L/S [L/kg]	el. cond. [mS/cm]	pH	E _H [mV]	IC [mg/L]	DOC [mg/L]	Cl [mg/L]	NO ₃ [mg/L]	SO ₄ [mg/L]
172.15	14.21	1.4	7.4	400	10	3.2	7.9	<0.1	15
174.15	14.37	1.4	7.4	418	10	4.6	7.9	<0.1	16
176.15	14.54	1.4	7.4	348	10	3.9	7.7	<0.1	15
182.15	15.04	1.4	7.3	334	9.9	3.2	7.7	<0.1	14
188.15	15.54	1.4	7.4	357	11	2.1	6.2	<0.1	15
195.15	16.12	1.3	7.3	354	9.9	4.3	n.d.	n.d.	n.d.
202.15	16.70	1.2	7.2	367	10	3.5	4.3	<0.1	15
209.15	17.29	1.2	7.4	368	10	5.0	3.0	<0.1	14
216.15	17.87	1.2	7.1	344	9.8	2.9	2.0	<0.1	15
223.15	18.45	1.2	7.3	309	n.d.	n.d.	2.5	<0.1	15
230.15	19.03	1.1	7.2	258	n.d.	n.d.	2.7	<0.1	15
237.15	19.61	1.4	7.0	275	n.d.	n.d.	n.d.	n.d.	n.d.
244.15	20.20	1.1	7.2	312	9.7	2.8	2.6	<0.1	12
251.15	20.78	1.2	7.3	310	10	2.6	2.3	<0.1	13
258.15	21.36	1.1	7.2	301	2.0	2.4	2.2	<0.1	13
286.15	21.53	2.6	7.9	308	5.6	6.5	7.0	<0.1	7
286.32	21.54	n.d.	n.d.	n.d.	4.6	6.5	23	<0.1	16
286.47	21.55	n.d.	n.d.	n.d.	3.9	5.4	16	<0.1	17
286.61	21.57	n.d.	n.d.	n.d.	3.7	5.0	14	<0.1	20
286.76	21.58	n.d.	n.d.	n.d.	3.7	6.0	11	<0.1	21
286.90	21.59	2.5	7.8	380	3.7	6.5	10	<0.1	23
288.15	21.69	2.5	7.8	347	4.8	3.3	8.6	<0.1	13
293.15	22.11	1.3	7.6	364	1.8	2.5	5.5	<0.1	12
300.15	22.69	1.2	7.5	327	1.7	2.6	4.9	<0.1	15
307.15	23.27	1.1	7.5	363	n.d.	n.d.	3.8	<0.1	15

A 1.4.2.2. Main Elements

Time [d]	Al [µg/L]	Ca [mg/L]	Fe [µg/L]	K [mg/L]	Mg [mg/L]	Na [mg/L]	Si [mg/L]
0.23	26	687	10	27	87	81	7.9
0.40	21	674	<5	26	79	64	8.0
0.57	28	688	8.9	25	75	68	8.2
0.72	24	702	8.6	25	76	68	8.3
0.86	23	711	<5	25	79	69	8.4
1.01	25	695	n.d.	25	78	72	8.4
1.17	28	686	12	25	77	71	8.5
1.34	28	684	<5	25	76	67	8.6
1.53	25	675	11	26	75	66	9.0
1.67	32	646	10	25	72	62	9.1
1.82	22	617	8.1	23	69	53	9.0
1.99	21	607	<5	23	69	44	9.0
2.13	39	600	<5	22	69	33	8.9
2.30	15	644	<5	23	74	22	9.7
2.43	27	639	8.1	22	72	15	9.8
2.57	28	636	8.7	22	69	9.8	10
2.69	31	606	11	21	63	6.7	10
2.86	20	554	11	19	54	4.6	9.9
3.01	18	471	11	17	42	3.2	9.8
3.15	30	356	16	13	27	2.5	9.4
3.30	29	222	22	10	14	1.9	8.8
3.47	44	116	31	8.2	6.8	1.8	8.6
3.61	44	87	44	7.4	5.2	1.8	8.4
3.76	63	72	58	7.0	4.3	2.0	8.3
3.90	79	63	66	6.6	3.8	2.0	8.0

Time [d]	Al [µg/L]	Ca [mg/L]	Fe [µg/L]	K [mg/L]	Mg [mg/L]	Na [mg/L]	Si [mg/L]
4.05	79	57	77	6.4	3.6	2.1	8.0
5.15	117	48	108	5.9	3.1	2.1	7.8
5.95	117	48	127	6.0	3.2	2.0	8.1
6.24	18	47	<5	5.8	3.1	1.9	7.6
6.94	157	44	124	5.4	2.8	1.7	7.6
7.88	93	43	109	5.5	2.7	1.6	7.5
8.16	93	41	110	5.3	2.6	1.5	7.2
9.42	106	44	140	5.7	2.6	1.6	7.8
10.15	95	42	130	5.4	2.5	1.5	7.5
11.15	93	37	118	4.8	2.2	1.3	6.7
11.42	106	37	123	4.8	2.2	1.3	6.7
12.15	107	36	120	4.7	2.1	1.3	6.7
12.57	92	36	122	4.6	2.1	1.3	6.7
13.15	91	35	115	4.4	2.1	1.3	6.6
13.57	83	35	118	4.4	2.1	1.3	6.7
14.15	76	34	106	4.1	2.0	1.2	6.3
16.15	91	33	117	3.9	1.9	1.3	6.4
19.15	55	34	135	3.7	2.0	1.3	6.6
20.15	47	32	134	3.4	1.9	1.3	6.2
21.15	46	31	152	3.2	1.8	1.3	6.2
22.15	15	30	62	2.9	1.7	1.5	6.1
25.15	38	28	313	2.4	1.6	1.2	6.0
26.15	38	28	354	2.4	1.6	1.2	6.0
27.15	43	27	409	2.3	1.5	1.2	6.0
29.15	37	27	252	2.1	1.5	1.2	6.0
32.15	42	27	632	1.9	1.3	1.3	6.0
33.15	52	26	526	1.7	1.3	1.2	5.9
34.15	44	25	413	1.9	1.2	1.2	6.1
35.15	51	25	294	1.8	1.2	1.2	6.1
36.15	50	25	249	1.7	1.2	1.1	6.0
39.15	50	25	126	1.7	1.2	1.1	6.1
40.15	51	25	94	1.6	1.2	1.1	6.2
41.15	62	24	76	1.6	1.1	1.1	6.1
42.15	53	24	60	1.5	1.1	1.1	6.1
43.15	71	23	54	1.4	1.1	1.1	6.0
46.15	30	24	39	1.3	1.2	1.1	6.3
47.15	31	24	39	1.3	1.1	1.1	6.3
48.15	31	24	40	1.3	1.1	1.1	6.3
49.15	30	24	39	1.3	1.1	1.1	6.3
50.15	38	24	37	1.3	1.1	1.1	6.3
53.15	52	23	52	1.1	1.1	1.1	6.2
54.15	49	24	43	1.2	1.1	1.1	6.5
55.15	47	24	41	1.1	1.1	1.1	6.5
56.01	31	24	39	1.1	1.1	1.1	6.4
60.15	59	23	52	1.0	1.0	1.1	6.4
61.15	38	23	43	0.91	1.0	1.1	6.2
62.15	49	22	49	0.89	0.96	1.0	6.0
63.15	54	22	47	0.91	0.97	1.1	6.3
64.15	38	22	43	0.86	1.0	1.1	6.3
68.15	21	23	32	0.82	0.98	1.0	6.3
69.15	51	22	36	0.78	0.96	1.0	6.3
70.15	36	22	31	0.80	0.96	1.0	6.2
71.15	44	23	34	0.78	0.94	1.0	6.3
74.15	28	23	46	0.74	0.99	1.0	6.4
75.15	26	22	43	0.73	0.98	1.0	6.3
76.22	25	22	36	0.72	0.94	1.0	6.3
77.15	25	22	37	0.69	0.93	1.0	6.2
78.01	23	16	30	0.48	0.67	0.76	4.5

Time [d]	Al [µg/L]	Ca [mg/L]	Fe [µg/L]	K [mg/L]	Mg [mg/L]	Na [mg/L]	Si [mg/L]
80.57	34	21	41	1.2	0.88	1.0	6.2
82.15	25	21	38	0.64	0.88	0.96	6.1
83.15	37	21	28	0.63	0.89	1.0	6.3
84.15	38	20	40	0.59	0.83	0.97	5.9
85.15	30	20	41	0.57	0.85	0.99	5.9
89.13	28	23	27	0.62	0.96	1.1	6.5
95.15	44	21	71	0.55	0.80	1.0	6.8
96.15	46	21	79	0.54	0.81	1.0	6.8
97.26	n.d.	21	n.d.	0.53	n.d.	1.0	6.8
98.15	67	21	93	0.55	0.81	1.1	6.8
99.17	81	21	89	0.54	0.79	1.1	6.9
102.15	25	23	20	0.56	0.74	1.2	7.4
103.15	24	23	20	0.56	0.74	1.2	7.4
105.15	46	20	78	0.50	0.71	1.1	6.7
106.17	48	20	95	0.55	0.72	1.1	6.5
109.15	64	20	117	0.48	0.69	1.1	6.5
110.15	63	20	117	0.50	0.69	1.0	6.3
111.15	65	20	122	0.47	0.69	1.1	6.4
112.15	102	20	134	0.48	0.68	1.0	6.3
113.17	49	20	118	0.48	0.70	1.1	6.4
116.15	61	20	129	0.47	0.68	1.1	6.3
119.15	<10	21	44	0.49	0.70	1.1	6.5
120.17	36	20	66	0.49	0.69	1.1	6.4
123.15	36	20	94	0.46	0.67	1.1	6.4
124.15	42	20	103	0.50	0.70	1.1	6.6
126.15	<10	20	97	0.47	0.67	1.1	6.4
130.15	39	20	95	0.47	0.66	1.1	6.2
131.15	45	19	99	0.46	0.64	1.1	6.2
132.15	50	20	110	0.48	0.65	1.1	6.4
133.15	39	20	90	0.48	0.64	1.1	6.4
134.17	67	20	102	0.46	0.71	1.1	6.3
137.15	51	20	101	0.45	0.68	1.1	6.2
138.15	52	20	102	0.45	0.67	1.1	6.3
139.15	71	20	106	0.46	0.67	1.1	6.3
140.15	62	19	105	0.46	0.66	1.1	6.2
141.15	63	19	103	0.44	0.66	1.1	6.2
145.15	77	21	57	0.47	0.71	1.1	6.5
151.15	38	n.d.	100	n.d.	0.66	n.d.	n.d.
153.15	41	n.d.	151	n.d.	0.65	n.d.	n.d.
158.15	37	n.d.	102	n.d.	0.64	n.d.	n.d.
161.15	40	n.d.	107	n.d.	0.65	n.d.	n.d.
162.15	40	n.d.	108	n.d.	0.64	n.d.	n.d.
165.15	38	n.d.	108	n.d.	0.67	n.d.	n.d.
167.15	40	n.d.	110	n.d.	0.67	n.d.	n.d.
169.15	37	n.d.	104	n.d.	0.67	n.d.	n.d.
172.15	39	n.d.	109	n.d.	0.70	n.d.	n.d.
174.15	37	n.d.	105	n.d.	0.69	n.d.	n.d.
176.15	38	n.d.	108	n.d.	0.69	n.d.	n.d.
182.15	29	20	81	8.1	0.72	1.1	6.3
188.15	16	21	45	5.7	0.80	1.3	6.5
195.15	32	19	116	4.6	0.76	1.1	5.9
202.15	44	19	96	3.3	0.78	1.2	5.9
209.15	56	18	129	1.9	0.77	1.1	5.5
216.15	56	19	119	1.1	0.80	1.2	5.5
223.15	57	19	139	1.4	0.86	1.1	5.4
230.15	57	19	155	1.2	0.88	1.1	5.3
244.15	62	18	185	1.6	0.84	1.0	5.4
251.15	63	18	207	1.2	0.84	1.1	5.1

Time [d]	Al [µg/L]	Ca [mg/L]	Fe [µg/L]	K [mg/L]	Mg [mg/L]	Na [mg/L]	Si [mg/L]
258.15	105	18	259	1.3	0.88	1.1	5.0
286.15	<10	51	418	7.1	2.2	1.7	9.1
286.32	<10	42	231	26	1.6	1.6	8.7
286.47	11	37	173	18	1.5	1.5	8.2
286.61	11	36	144	15	1.4	1.5	7.9
286.76	10	36	124	12	1.5	1.5	7.7
286.90	<10	36	104	11	1.5	1.6	7.5
288.15	<10	43	68	8.7	1.8	1.8	7.3
293.15	42	18	125	5.1	0.79	0.99	5.7
300.15	56	17	140	3.9	0.79	1.1	5.1
307.15	454	n.d.	342	n.d.	0.83	n.d.	n.d.

A 1.4.2.3. Trace Elements

Time [d]	As [µg/L]	Ba [µg/L]	Cd [µg/L]	Co [µg/L]	Cr [µg/L]	Cu [µg/L]	Mn [µg/L]	Mo [µg/L]	Ni [µg/L]	Sr [µg/L]	V [µg/L]	Zn [µg/L]
0.23	2.0	43	3.2	<2	<2	9.2	19	<2	4.7	2479	<2	149
0.40	<2	41	<2	<2	<2	8.6	18	<2	4.6	2421	<2	84
0.57	<2	42	<2	<2	<2	8.2	20	<2	4.3	2460	<2	55
0.72	<2	43	<2	<2	<2	7.9	21	<2	4.1	2492	<2	49
0.86	<2	44	<2	<2	<2	8.2	22	<2	4.4	2496	<2	55
1.01	<2	43	<2	<2	<2	7.7	23	<2	4.3	2383	<2	49
1.17	<2	44	2.5	<2	<2	7.4	26	<2	4.5	2384	<2	49
1.34	<2	43	<2	<2	<2	7.1	29	<2	4.5	2384	<2	51
1.53	<2	43	<2	<2	<2	7.8	33	<2	4.1	2371	<2	52
1.67	<2	41	<2	<2	<2	7.7	35	<2	4.1	2286	<2	48
1.82	<2	39	<2	<2	<2	7.8	36	<2	4.0	2125	<2	56
1.99	<2	38	<2	<2	<2	7.1	38	<2	3.8	2117	<2	43
2.13	<2	37	<2	<2	2.0	9.1	42	<2	5.9	2111	2.5	43
2.30	<2	39	2.6	<2	<2	7.5	62	<2	4.8	2250	<2	40
2.43	<2	39	<2	<2	<2	8.1	77	<2	4.4	2176	<2	40
2.57	<2	38	<2	<2	<2	7.3	93	<2	4.1	2171	<2	36
2.69	<2	38	<2	<2	<2	<2	106	<2	<2	2048	<2	<2
2.86	<2	38	<2	<2	<2	7.0	122	<2	3.6	1855	<2	28
3.01	<2	41	<2	<2	<2	7.2	131	<2	3.6	1574	<2	24
3.15	<2	47	<2	<2	<2	7.5	132	2.1	3.2	1169	<2	24
3.30	<2	57	<2	<2	<2	7.7	107	2.5	2.9	737	<2	29
3.47	<2	79	<2	<2	<2	<2	85	<2	<2	424	<2	<2
3.61	<2	87	<2	<2	<2	8.4	84	3.3	2.3	347	2.0	10
3.76	<2	90	<2	<2	<2	8.9	84	3.6	2.5	300	2.2	9.7
3.90	<2	85	<2	<2	<2	9.5	83	4.4	3.8	268	2.4	11
4.05	<2	81	<2	<2	<2	8.7	85	4.2	2.5	254	2.3	8.8
5.15	<2	60	<2	<2	<2	7.2	89	4.9	2.5	220	2.3	7.8
5.95	<2	60	<2	<2	<2	6.9	118	5.4	2.6	226	2.3	16
6.24	2.1	49	<2	<2	<2	4.8	5.2	5.8	n.d.	222	<2	5.7
6.94	<2	52	<2	<2	<2	6.1	155	5.7	2.2	205	<2	6.9
7.88	<2	52	2.6	<2	2.0	5.3	160	7.0	2.5	206	<2	7.5
8.16	3.0	50	<2	<2	<2	4.5	158	5.6	2.3	199	<2	5.4
9.42	<2	53	<2	<2	<2	5.3	191	5.9	2.9	211	<2	6.8
10.15	<2	50	<2	<2	<2	3.6	191	5.2	2.3	202	<2	5.5
11.15	<2	45	<2	<2	<2	<2	197	<2	<2	182	<2	<2
11.42	<2	43	<2	<2	<2	2.9	196	4.8	2.1	174	<2	3.6
12.15	<2	43	<2	<2	<2	2.4	207	4.6	2.0	172	<2	3.8
12.57	<2	43	<2	<2	<2	2.4	222	4.5	2.2	175	<2	3.5
13.15	<2	42	<2	<2	<2	2.2	227	4.2	2.1	170	<2	2.4

Time [d]	As [µg/L]	Ba [µg/L]	Cd [µg/L]	Co [µg/L]	Cr [µg/L]	Cu [µg/L]	Mn [µg/L]	Mo [µg/L]	Ni [µg/L]	Sr [µg/L]	V [µg/L]	Zn [µg/L]
13.57	<2	43	<2	<2	<2	2.2	249	4.4	2.3	172	<2	3.5
14.15	<2	40	<2	<2	<2	2.2	249	4.4	2.1	162	<2	5.7
16.15	<2	42	<2	<2	<2	<2	295	4.1	2.3	158	<2	6.3
19.15	<2	40	<2	<2	2.7	2.6	449	6.3	2.5	162	<2	2.6
20.15	<2	39	<2	<2	2.1	2.0	409	4.6	2.3	155	<2	2.5
21.15	2.7	37	<2	<2	2.0	2.0	391	4.3	2.1	144	<2	2.6
22.15	<2	35	<2	<2	2.6	5.0	388	4.0	2.1	139	<2	14
25.15	<2	36	<2	<2	2.0	<2	400	3.1	1.8	136	<2	7.6
26.15	2.5	35	<2	<2	2.4	4.7	392	3.2	<2	130	<2	2.2
27.15	2.4	36	<2	<2	<2	<2	397	2.8	<2	130	<2	5.2
29.15	<2	35	<2	<2	<2	<2	368	<2	<2	129	<2	<2
32.15	<2	35	<2	<2	<2	<2	431	<2	<2	126	<2	<2
33.15	3.0	33	<2	<2	2.2	<2	432	2.2	<2	124	<2	<2
34.15	<2	33	<2	<2	<2	<2	399	<2	<2	117	<2	<2
35.15	<2	31	<2	<2	2.0	<2	387	<2	<2	116	<2	5.0
36.15	<2	31	<2	<2	<2	<2	375	<2	<2	116	<2	2.9
39.15	<2	29	<2	<2	<2	<2	295	<2	<2	122	<2	2.2
40.15	2.3	28	<2	<2	2.2	<2	282	<2	<2	116	<2	<2
41.15	<2	26	<2	<2	<2	<2	271	<2	<2	112	<2	<2
42.15	<2	25	<2	<2	<2	<2	236	<2	<2	109	<2	<2
43.15	<2	24	<2	<2	<2	<2	217	<2	<2	108	<2	<2
46.15	<2	24	<2	<2	<2	<2	249	<2	<2	112	<2	<2
47.15	<2	24	<2	<2	<2	<2	256	<2	<2	111	<2	<2
48.15	3.4	23	<2	<2	2.0	<2	258	<2	<2	109	<2	2.9
49.15	2.7	24	<2	<2	<2	<2	278	<2	<2	111	<2	3.4
50.15	<2	24	<2	<2	<2	<2	292	<2	<2	109	<2	<2
53.15	2.7	25	<2	<2	2.2	<2	273	<2	<2	107	<2	<2
54.15	2.8	26	<2	<2	<2	<2	301	<2	<2	109	<2	<2
55.15	3.0	26	<2	<2	<2	<2	293	<2	<2	108	<2	<2
56.01	3.0	27	<2	<2	<2	<2	306	<2	<2	109	<2	<2
60.15	3.3	26	<2	<2	<2	<2	313	<2	<2	107	<2	<2
61.15	3.1	26	<2	<2	<2	<2	278	<2	<2	106	<2	<2
62.15	3.0	24	<2	<2	<2	<2	258	<2	<2	101	<2	<2
63.15	<2	24	<2	<2	<2	<2	262	<2	<2	103	<2	<2
64.15	<2	24	<2	<2	<2	<2	247	<2	<2	104	<2	<2
68.15	<2	23	<2	<2	<2	<2	297	<2	<2	106	<2	2.5
69.15	<2	23	<2	<2	<2	<2	287	<2	<2	104	<2	<2
70.15	<2	23	<2	<2	<2	<2	273	<2	<2	104	<2	<2
71.15	<2	22	<2	<2	<2	<2	278	<2	<2	103	<2	<2
74.15	<2	29	<2	<2	3.3	<2	732	<2	<2	109	<2	5.5
75.15	<2	29	<2	<2	2.5	<2	707	<2	<2	108	<2	2.7
76.22	<2	28	<2	<2	2.7	<2	655	<2	<2	104	<2	2.7
77.15	<2	28	<2	<2	2.2	<2	743	<2	<2	103	<2	<2
78.01	<2	21	<2	<2	2.3	<2	512	<2	<2	75	<2	<2
80.57	3.3	26	<2	<2	2.9	<2	637	<2	<2	98	<2	2.6
82.15	<2	25	<2	<2	2.2	<2	581	<2	<2	98	<2	2.6
83.15	2.8	21	<2	<2	<2	<2	439	<2	<2	99	<2	<2
84.15	<2	23	<2	<2	2.4	<2	454	<2	<2	95	<2	2.4
85.15	<2	22	<2	<2	<2	<2	403	<2	<2	96	<2	2.6
89.13	3.3	20	<2	<2	<2	<2	208	<2	<2	107	<2	2.9
95.15	<2	14	<2	<2	<2	<2	5.3	<2	<2	90	<2	2.3
96.15	<2	14	<2	<2	<2	<2	8.0	<2	<2	88	<2	<2
98.15	<2	14	<2	<2	<2	<2	19	<2	<2	87	<2	<2
99.17	2.8	13	<2	<2	<2	<2	18	<2	<2	84	<2	2.4
102.15	<2	11	<2	<2	<2	<2	85	<2	<2	72	<2	<2
103.15	<2	11	<2	<2	<2	<2	115	<2	<2	75	<2	<2
105.15	<2	13	<2	<2	<2	<2	170	<2	<2	77	<2	<2
106.17	<2	14	<2	<2	<2	<2	256	<2	<2	78	<2	3.1

Time [d]	As [µg/L]	Ba [µg/L]	Cd [µg/L]	Co [µg/L]	Cr [µg/L]	Cu [µg/L]	Mn [µg/L]	Mo [µg/L]	Ni [µg/L]	Sr [µg/L]	V [µg/L]	Zn [µg/L]
109.15	<2	15	<2	<2	<2	<2	266	<2	<2	77	<2	3.1
110.15	<2	15	<2	<2	<2	<2	274	<2	<2	77	<2	3.0
111.15	<2	15	<2	<2	<2	<2	286	<2	<2	77	<2	3.1
112.15	<2	15	<2	<2	2.1	<2	299	<2	<2	76	<2	3.1
113.17	<2	15	<2	<2	<2	<2	318	<2	<2	78	<2	3.4
116.15	<2	16	<2	<2	<2	<2	277	<2	<2	79	<2	<2
119.15	<2	13	<2	<2	<2	<2	323	<2	<2	80	<2	<2
120.17	<2	14	<2	<2	<2	<2	297	<2	<2	80	<2	<2
123.15	3.6	15	<2	<2	<2	<2	187	<2	<2	78	<2	3.6
124.15	3.7	16	<2	<2	2.1	<2	148	<2	<2	81	<2	2.9
126.15	<2	15	<2	<2	<2	<2	71	<2	<2	78	<2	<2
130.15	3.6	13	<2	<2	<2	<2	7.3	<2	<2	77	<2	2.4
131.15	3.7	13	<2	<2	2.2	<2	4.7	<2	<2	76	<2	2.4
132.15	<2	13	15	<2	<2	<2	3.8	<2	<2	77	<2	2.7
133.15	<2	13	7.1	<2	<2	<2	3.1	<2	<2	77	<2	2.5
134.17	<2	13	<2	<2	<2	<2	3.2	<2	<2	85	<2	2.4
137.15	<2	13	<2	<2	2.0	<2	3.2	<2	<2	82	<2	2.6
138.15	<2	13	2.1	<2	<2	<2	3.1	<2	<2	81	<2	2.9
139.15	<2	13	<2	<2	<2	<2	3.2	<2	<2	80	<2	2.5
140.15	<2	13	<2	<2	<2	<2	3.2	<2	<2	81	<2	2.5
141.15	<2	13	<2	<2	<2	<2	3.2	<2	<2	81	<2	2.5
145.15	<2	11	<2	<2	<2	<2	2.9	<2	<2	84	<2	<2
151.15	<2	12	19	<2	<2	<2	<2	<2	<2	75	<2	3.7
153.15	<2	19	<2	<2	2.8	<2	580	<2	<2	73	<2	4.6
158.15	<2	18	<2	<2	3.2	<2	558	<2	<2	72	<2	3.8
161.15	<2	18	<2	<2	3.3	<2	555	<2	<2	73	<2	3.9
162.15	<2	19	<2	<2	2.9	<2	566	<2	<2	74	<2	3.7
165.15	3.3	19	<2	<2	2.4	<2	587	<2	<2	78	<2	3.5
167.15	3.2	19	<2	<2	3.1	<2	565	<2	<2	75	<2	3.6
169.15	2.9	18	<2	<2	2.7	<2	553	<2	<2	74	<2	3.3
172.15	3.0	18	<2	<2	2.9	<2	557	<2	<2	75	<2	6.8
174.15	3.0	18	<2	<2	2.8	<2	547	<2	<2	75	<2	3.5
176.15	<2	18	<2	<2	2.6	<2	504	<2	<2	72	<2	3.7
182.15	<2	14	<2	<2	<2	<2	126	<2	<2	72	<2	<2
188.15	2.9	9	<2	<2	<2	<2	195	<2	<2	75	<2	2.7
195.15	2.7	12	<2	<2	<2	<2	58	<2	<2	69	<2	3.7
202.15	2.5	11	<2	<2	<2	<2	47	<2	<2	68	<2	3.0
209.15	2.4	13	<2	<2	<2	<2	56	<2	<2	72	<2	3.0
216.15	2.2	13	<2	<2	<2	<2	81	<2	<2	74	<2	3.5
223.15	<2	16	<2	<2	<2	<2	111	<2	<2	80	2.0	3.6
230.15	3.4	14	<2	<2	<2	<2	169	<2	<2	77	<2	3.5
244.15	3.3	14	<2	<2	<2	<2	188	<2	<2	70	<2	3.7
251.15	3.3	14	<2	<2	<2	<2	240	<2	<2	71	<2	3.6
258.15	3.1	15	<2	<2	2.1	<2	296	<2	<2	70	2.2	4.0
286.15	9.0	34	<2	<2	5.3	<2	1690	<2	<2	181	<2	33
286.32	6.7	26	<2	<2	3.9	<2	1063	<2	<2	128	<2	50
286.47	6.8	24	<2	<2	3.3	<2	922	<2	<2	116	<2	21
286.61	6.5	23	<2	<2	3.1	<2	867	<2	<2	112	<2	16
286.76	6.5	22	<2	<2	3.0	<2	835	2.0	<2	110	<2	12
286.90	6.6	23	<2	<2	3.0	<2	848	<2	<2	113	<2	9.7
288.15	5.8	28	<2	<2	3.2	<2	960	<2	<2	138	<2	3.9
293.15	4.0	16	<2	<2	<2	<2	371	<2	<2	61	<2	3.6
300.15	3.0	17	<2	<2	<2	<2	320	<2	<2	59	<2	4.1
307.15	n.d.	20	<2	<2	<2	2.2	279	<2	<2	64	2.8	5.9

A 1.4.3. Flow-path sampling

A 1.4.3.1. Liquid/solid ratio, electric conductivity, pH and E_H value, inorganic carbon, dissolved organic carbon and anions

1. Flow-path sampling after 90 days

flow-path [cm]	el. cond. [mS/cm]	pH	E_H [mV]	IC [mg/L]	DOC [mg/L]	Cl [mg/L]	NO ₃ [mg/L]	SO ₄ [mg/L]
1	0.75	7.9	331	4.7	3.4	n.d.	n.d.	n.d.
2	0.78	7.5	295	5.8	4.4	n.d.	n.d.	n.d.
3	0.80	7.1	431	6.2	5.1	n.d.	n.d.	n.d.
4	0.89	7.9	589	6.7	6.2	n.d.	n.d.	n.d.
5	0.81	8.1	410	7.4	8.6	n.d.	n.d.	n.d.
6	0.88	7.7	416	6.7	7.2	n.d.	n.d.	n.d.
7	0.88	8.1	393	7.3	11	n.d.	n.d.	n.d.
8	0.90	8.1	379	7.1	13	n.d.	n.d.	n.d.
10	1.7	8.0	390	7.5	7.6	n.d.	n.d.	n.d.
16	1.2	8.0	435	9.5	7.1	n.d.	n.d.	n.d.
22	1.2	8.1	408	11	8.7	n.d.	n.d.	n.d.
28	1.3	8.0	461	12	8.3	n.d.	n.d.	n.d.
34	1.6	8.0	488	11	10.2	n.d.	n.d.	n.d.
40	1.3	8.1	393	12	11.3	n.d.	n.d.	n.d.
48	1.4	8.0	108	13	9.2	n.d.	n.d.	n.d.

2. Flow-path sampling after 282 days

flow-path [cm]	el. cond. [μS/cm]	pH	E_H [mV]	IC [mg/L]	DOC [mg/L]	Cl [mg/L]	NO ₃ [mg/L]	SO ₄ [mg/L]
1	562	4.4	568	2.3	23	1.8	2.0	11
2	502	4.4	549	2.3	14	2.1	2.1	11
3	463	5.0	535	2.6	12	1.8	2.5	11
4	438	5.4	544	2.6	9.9	1.6	1.9	12
5	458	5.8	507	3.7	21	1.5	2.2	12
6	507	6.6	487	4.3	18	1.8	2.0	12
7	588	7.2	458	4.2	18	1.7	2.0	12
8	660	7.4	447	5.1	14	1.6	2.3	13
9	788	7.6	437	6.3	19	1.7	2.0	13
10	781	7.7	374	n.d.	n.d.	n.d.	n.d.	13
12	808	7.8	391	6.1	18	1.6	2.3	14
16	862	7.8	415	5.4	15	1.6	2.0	14
22	935	7.9	391	6.3	15	1.8	3.0	14
28	1046	8.1	381	7.6	10	1.6	2.3	15
34	1064	8.0	371	6.9	21	2.1	1.8	15
40	1065	8.0	369	6.4	15	1.7	1.7	14
48	1082	8.0	93	6.8	18	1.9	2.8	14
50	1168	7.4	439	11.65	13	3.1	n.d.	n.d.

A 1.4.3.2. Main elements

1. Flow-path sampling after 90 days

flow-path [cm]	Al [µg/L]	Ca [mg/L]	Fe [µg/L]	K [µg/L]	Mg [µg/L]	Na [mg/L]	S [mg/L]	Si [mg/L]
1	12	9.0	<2	395	671	1.1	<1	0.93
2	25	11	<2	<100	741	1.1	3.9	1.1
3	36	12	5.2	355	856	1.1	<1	1.3
4	36	14	8.3	393	895	1.0	4.0	1.6
5	37	13	41	389	815	1.0	3.9	1.6
6	31	14	24	529	796	1.0	4.1	1.8
7	32	15	23	405	880	1.0	4.1	2.0
8	35	15	24	426	846	1.0	4.2	2.0
10	34	16	47	449	801	1.0	4.3	2.5
16	35	20	42	618	871	1.2	4.7	3.5
22	34	22	35	543	845	1.1	4.8	4.4
28	29	23	19	645	818	1.2	5.0	5.1
34	76	23	197	647	844	1.3	4.9	5.5
40	44	24	47	580	900	1.2	4.8	5.8
48	13	26	76	693	1058	1.2	4.5	6.9

2. Flow-path sampling after 282 days

flow-path [cm]	Al [µg/L]	Ca [mg/L]	Fe [µg/L]	K [µg/L]	Mg [µg/L]	Na [mg/L]	S [mg/L]	Si [mg/L]
1	71	2.9	13	354	0.36	1.4	3.2	0.43
2	63	3.0	8.7	799	0.34	1.1	3.1	0.47
3	17	3.7	42	376	0.40	1.2	3.2	0.56
4	7.7	3.8	<2	367	0.41	1.1	3.3	0.65
5	3.1	4.1	<2	352	0.45	1.1	3.4	0.76
6	3.0	5.0	<2	343	0.51	1.1	3.6	0.89
7	2.7	6.8	14	359	0.78	1.1	3.6	1.0
8	2.7	7.8	3.6	479	0.85	1.3	3.7	1.0
9	28	11	<2	392	0.95	1.1	3.9	1.2
10	30	11	<2	372	0.90	1.1	3.9	1.2
12	29	11	<2	387	0.89	1.1	3.9	1.3
16	24	12	33	358	0.78	1.1	4.0	1.4
22	20	14	32	324	0.91	1.1	4.3	1.8
28	29	16	87	323	0.92	1.1	4.4	2.3
34	19	16	27	387	0.98	1.1	4.5	2.7
40	27	16	66	374	0.98	1.1	4.6	2.9
48	47	17	65	491	0.88	1.1	4.4	3.9
50	48	17	94	1698	0.93	1.3	4.3	3.9

A 1.4.3.3. Trace elements

1. Flow-path sampling after 90 days

flow-path [cm]	Ba [µg/L]	Cd [µg/L]	Co [µg/L]	Cr [µg/L]	Cu [µg/L]	Mn [µg/L]	Mo [µg/L]	Ni [µg/L]	Sr [µg/L]	V [µg/L]	Zn [µg/L]
1	14	<2	<2	<2	<2	32	<2	<2	27	<2	20.00
2	9.2	<2	<2	<2	<2	<2	<2	<2	31	<2	<5
3	11	<2	<2	<2	<2	<2	<2	<2	35	<2	<5
4	11	<2	<2	<2	<2	<2	<2	<2	40	<2	<5
5	9.6	<2	<2	<2	<2	<2	<2	<2	37	<2	<5
6	11	<2	<2	<2	<2	<2	<2	<2	41	<2	<5
7	10	<2	<2	<2	<2	<2	<2	<2	46	<2	<5
8	10	<2	<2	<2	<2	<2	<2	<2	46	<2	<5
10	13	<2	<2	<2	<2	<2	<2	<2	52	<2	<5
16	14	<2	<2	<2	<2	6.5	<2	<2	68	<2	8.1
22	17	<2	<2	<2	<2	56	<2	<2	83	<2	<5
28	16	<2	<2	<2	<2	48	<2	<2	92	<2	6.7
34	18	<2	<2	<2	2.5	72	<2	<2	89	<2	10
40	17	<2	<2	<2	<2	143	<2	<2	96	<2	<5
48	33	<2	<2	<2	<2	916	<2	<2	110	<2	<5

2. Flow-path sampling after 282 days

flow-path [cm]	Ba [µg/L]	Cd [µg/L]	Co [µg/L]	Cr [µg/L]	Cu [µg/L]	Mn [µg/L]	Mo [µg/L]	Ni [µg/L]	Sr [µg/L]	V [µg/L]	Zn [µg/L]
1	12	<2	<2	<2	23	48	<2	<2	9.9	<2	22.34
2	15	<2	<2	<2	104	62	<2	<2	9.9	<2	22
3	89	<2	<2	<2	<2	275	<2	<2	13	<2	83
4	96	<2	<2	<2	219	341	<2	<2	12	<2	147
5	89	<2	<2	<2	<2	446	<2	<2	14	<2	252
6	63	<2	<2	<2	<2	285	<2	<2	17	<2	352
7	37	<2	<2	<2	<2	155	<2	10	17	<2	244
8	25	<2	<2	<2	2.7	31	<2	3	18	<2	45
9	5.7	<2	<2	<2	<2	<2	<2	<2	25	<2	<2
10	5.9	<2	<2	<2	<2	<2	<2	<2	25	<2	<2
12	6.6	<2	<2	<2	<2	<2	<2	<2	27	2.1	<2
16	7.2	<2	<2	<2	<2	<2	<2	<2	30	2.2	<2
22	7.6	<2	<2	<2	<2	<2	<2	<2	34	3.1	<2
28	8.9	<2	<2	<2	<2	2.7	<2	<2	41	3.2	5.4
34	8.8	<2	<2	<2	<2	145	<2	<2	46	2.9	4.6
40	8.9	2.7	<2	<2	<2	115	<2	<2	48	3.2	38
48	15	3.5	<2	<2	<2	249	<2	<2	58	3.1	701
50	19	<2	<2	<2	<2	249	<2	<2	60	2.7	3.8

A 1.5. Data of the unsaturated column experiments with the sandy fraction of harbour sediments

A 1.5.1. Characteristic data of the column

Length	[cm]	48
Diameter	[cm]	5
Volume	[cm ³]	942
Amount material original	[kg]	5.88
Amount material dry	[kg]	5.26
Amount water at start	[L]	1.22
Volume outlet	[L/d]	0.05
Volume inlet	[L/d]	0.03
Specific weight	[g/cm ³]	2.61
Absolut porosity		0.4
Watersaturation	[% abs. porosity]	60
kf-value	[m/s]	8.3 * 10 ⁻⁵
Seepage velocity	[cm/d]	3.4

A 1.5.2. Outlet

A.1.5.2.1. Liquid/solid ratio, electric conductivity, pH and E_H value, inorganic carbon, dissolved organic carbon and anions

Time	L/S	el. cond.	pH	E _H	IC	DOC	Cl	NO ₃	SO ₄
[d]	[L/kg]	[mS/cm]		[mV]	[mg/L]	[mg/L]	[mg/L]	[mg/L]	[mg/L]
1.81	0.01	43	3.8	573	2.3	22	74	410	397
4.87	0.03	39	6.5	465	6.2	16	57	368	400
6.02	0.04	39	7.0	438	14	27	38	266	447
6.90	0.04	35	6.9	484	15	19	29	200	445
7.83	0.05	33	6.5	416	3.1	20	22	170	469
8.83	0.05	32	6.8	448	16	20	18	131	460
11.95	0.07	31	7.2	535	4.1	22	10	79	456
12.80	0.07	28	7.4	532	21	25	5.9	54	475
13.86	0.08	27	7.2	527	20	25	4.5	42	455
14.84	0.08	27	6.6	521	16	21	3.4	43	456
15.85	0.09	23	8.0	476	3.5	19	3.1	29	457
18.93	0.11	27	7.3	530	23	34	1.8	24	463
19.85	0.11	27	7.4	n.d.	23	19	0.2	19	458
21.84	0.12	26	7.2	n.d.	23	9.3	<0.2	17	456
22.86	0.12	26	7.5	n.d.	23	18	<0.2	13	446
26.85	0.13	26	7.6	n.d.	25	23	<0.2	13	459
27.84	0.14	26	6.5	n.d.	17	16	<0.2	21	460
28.87	0.14	26	7.3	n.d.	26	14	<0.2	13	448
29.85	0.15	25	7.0	n.d.	18	17	<0.2	19	459
32.87	0.16	25	7.4	n.d.	23	18	<0.2	12	456
34.86	0.17	25	7.5	511	22	17	<0.2	11	432
35.84	0.17	25	7.5	475	22	23	<0.2	11	446
36.85	0.17	24	7.6	433	22	70	<0.2	11	437
39.84	0.19	25	7.6	489	22	19	<0.2	11	439
41.81	0.20	24	7.6	458	22	16	<0.2	11	430
42.95	0.21	24	7.6	497	21	16	<0.2	11	425
46.88	0.23	25	7.2	550	19	16	<0.2	14	421

Time [d]	L/S [L/kg]	el. cond. [mS/cm]	pH	E _H [mV]	IC [mg/L]	DOC [mg/L]	Cl [mg/L]	NO ₃ [mg/L]	SO ₄ [mg/L]
47.78	0.23	24	7.5	529	21	26	<0.2	10	410
48.79	0.24	23	7.5	477	21	15	<0.2	10	406
49.80	0.24	22	7.6	511	20	31	<0.2	9.2	400
50.78	0.25	23	7.5	452	21	22	<0.2	9.9	406
53.83	0.26	23	7.4	402	21	17	<0.2	9.4	393
54.84	0.27	23	7.5	415	22	18	<0.2	8.2	359
55.83	0.27	23	7.5	407	22	17	0.89	6.8	310
56.86	0.28	22	7.5	454	23	24	0.54	7.2	310
57.84	0.28	21	7.6	443	24	21	0.68	6.2	268
60.92	0.30	n.d.	n.d.	210	26	28	0.69	5.9	155
61.88	0.31	9	7.4	480	23	20	0.92	5.3	67
64.84	0.32	n.d.	n.d.	210	25	19	0.77	5.8	48
67.84	0.34	6.6	7.5	449	27	12	0.76	4.7	38
69.87	0.35	6.3	7.6	458	28	23	0.87	4.7	35
71.84	0.36	n.d.	n.d.	n.d.	27	14	0.69	5.1	34
74.84	0.37	6.2	7.5	454	28	18	0.64	4.2	33
77.89	0.39	6.3	7.1	542	27	28	0.54	4.5	32
78.82	0.40	6.0	7.6	486	27	24	0.64	4.2	31
81.82	0.41	6.0	7.6	497	27	13	0.85	3.8	31
83.82	0.42	5.8	7.6	471	27	30	1.08	4.3	31
85.82	0.43	5.8	7.7	480	27	19	0.67	4.1	31
88.87	0.45	5.8	7.6	468	27	16	0.88	4.1	30
90.83	0.46	5.7	7.5	547	45	20	0.77	3.7	29
92.82	0.46	5.8	7.7	349	26	21	0.76	4.0	30
98.84	0.49	5.7	7.7	392	26	11	1.0	4.0	30
104.84	0.52	5.8	7.7	406	27	14	0.65	3.8	30
111.83	0.55	5.7	7.7	431	26	15	1.3	4.2	30
118.81	0.57	5.8	7.6	453	26	20	n.d.	n.d.	n.d.
125.81	0.60	5.9	7.7	433	26	14	1.3	4.4	31
132.82	0.63	5.9	7.6	432	25	9.9	0.86	4.4	31
139.83	0.66	5.8	7.7	408	26	8.2	0.78	4.9	32
146.83	0.69	5.6	7.7	400	25	13	0.64	4.2	30
153.82	0.73	5.5	7.6	419	25	12	0.79	4.2	27
160.83	0.76	5.4	7.7	413	25	12	1.1	4.0	26
167.84	0.78	5.4	7.7	411	25	5.8	0.73	4.4	26
174.85	0.81	5.2	7.7	414	5.5	13	1.0	4.5	26
201.74	0.82	5.2	7.8	450	5.0	13	1.0	4.9	26
202.84	0.83	5.3	7.8	431	5.1	18	0.98	4.9	27
203.60	0.83	5.9	7.7	435	5.4	5.5	1.4	6.4	27
209.83	0.86	6.1	7.7	419	5.8	9.4	0.70	5.4	30
216.83	0.88	6.6	7.8	413	5.5	11	0.80	6.9	34
223.23	0.91	7.1	7.7	458	n.d.	n.d.	0.69	8.8	39
237.23	0.93	8.1	7.5	439	n.d.	n.d.	1.0	11	47

A.1.5.2.2. Main elements

Time [d]	Al [µg/L]	Ca [mg/L]	Fe [µg/L]	K [mg/L]	Mg [mg/L]	Na [mg/L]	Si [mg/L]
1.81	9155	943	360	35	139	170	12
4.87	90	943	11	35	138	164	14
6.02	55	851	4.0	33	125	150	17
6.90	50	791	3.6	31	116	141	16
7.83	66	752	4.1	31	110	135	16
8.83	54	726	5.3	30	106	129	16
11.95	47	675	3.7	29	98	111	15

Time [d]	Al [µg/L]	Ca [mg/L]	Fe [µg/L]	K [mg/L]	Mg [mg/L]	Na [mg/L]	Si [mg/L]
12.80	39	644	3.9	28	93	92	14
13.86	44	627	3.2	29	93	86	14
14.84	98	617	3.5	28	91	77	13
15.85	34	632	<2	29	93	71	14
18.93	51	627	<2	28	92	57	13
19.85	35	621	2.9	27	91	45	13
21.84	28	630	<2	27	91	39	13
22.86	39	625	<2	26	90	33	13
26.85	38	624	<2	26	90	29	13
27.84	40	625	3.7	26	89	26	13
28.87	33	629	<2	26	90	23	13
29.85	28	627	<2	25	89	21	13
32.87	52	624	<2	25	88	18	12
34.86	23	636	<2	25	90	14	12
35.84	32	628	<2	24	88	13	12
36.85	11	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
39.84	22	632	<2	24	86	10	12
41.81	22	627	<2	23	82	8.2	12
42.95	18	631	2.2	23	79	7.3	12
46.88	26	628	<2	22	73	6.1	12
47.78	25	625	<2	21	65	5.2	12
48.79	21	631	<2	21	62	4.8	12
49.80	18	626	<2	20	57	4.5	12
50.78	18	639	<2	20	55	4.3	12
53.83	22	636	<2	19	47	3.8	12
54.84	19	642	<2	17	37	3.3	12
55.83	17	643	<2	15	32	3.1	12
56.86	20	634	<2	14	26	2.9	12
57.84	4.2	612	<2	13	21	3.1	13
60.92	5.2	379	5.6	9.8	10	3.1	13
61.88	4.8	202	2.8	8.5	4.8	3.1	13
64.84	6.3	153	2.4	7.9	3.7	3.1	12
67.84	5.9	131	5.5	7.7	3.3	3.1	12
69.87	18	n.d.	3.5	n.d.	n.d.	n.d.	n.d.
71.84	6.0	n.d.	<2	n.d.	n.d.	n.d.	n.d.
74.84	7.7	127	<2	8.1	3.4	3.4	13
77.89	6.1	121	<2	7.9	3.3	3.4	12
78.82	5.7	121	<2	8.0	3.4	3.3	12
81.82	5.1	113	<2	7.6	3.2	3.1	12
83.82	5.9	116	<2	7.7	3.3	3.3	12
85.82	5.1	115	<2	7.7	3.3	3.2	12
88.87	4.4	111	<2	7.4	3.3	3.1	12
90.83	4.3	110	<2	7.4	3.3	3.0	12
92.82	4.8	111	3.0	7.5	3.3	3.0	12
98.84	5.2	n.d.	<2	n.d.	n.d.	n.d.	n.d.
104.84	5.2	113	<2	7.4	3.6	3.0	13
111.83	5.1	110	<2	7.1	3.5	2.9	12
118.81	6.5	109	<2	6.9	3.6	2.8	12
125.81	12	112	15	6.8	3.7	2.8	13
132.82	4.4	110	3.4	6.6	3.7	2.7	13
139.83	6.4	110	<2	6.6	3.8	2.6	13
146.83	7.0	107	2.8	6.4	3.7	2.6	13
153.82	4.1	103	2.8	6.2	3.6	2.6	13
160.83	4.0	100	<2	6.0	3.6	2.5	13
167.84	4.3	100	<2	6.0	3.5	2.5	13
174.85	2.9	100	<2	5.9	3.5	2.5	13
201.74	11	98	<2	5.8	3.5	2.5	13

Time [d]	Al [µg/L]	Ca [mg/L]	Fe [µg/L]	K [mg/L]	Mg [mg/L]	Na [mg/L]	Si [mg/L]
202.84	7.3	101	<2	5.7	3.6	2.6	14
203.60	7.6	109	3.4	6.1	3.9	2.6	14
209.83	2.1	115	<2	6.1	4.2	2.7	14
216.83	2.3	125	<2	6.4	4.6	2.8	14
223.23	1.7	134	<2	6.5	5.0	2.9	14
237.23	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.

A 1.5.2.3. Trace elements

Time [d]	As [µg/L]	Ba [µg/L]	Cd [µg/L]	Co [µg/L]	Cr [µg/L]	Cu [µg/L]	Mn [µg/L]	Mo [µg/L]	Ni [µg/L]	Sr [µg/L]	V [µg/L]	Zn [µg/L]
1.81	26	72	<2	7.3	45	472	474	<2	88	3591	<2	166
4.87	10	44	<2	<2	<2	57	64	<2	19	3554	<2	27
6.02	7.9	36	<2	<2	<2	27	15	<2	13	3214	<2	11
6.90	<2	34	<2	<2	<2	21	14	<2	12	2972	<2	11
7.83	<2	33	<2	<2	<2	21	12	<2	7.6	2833	<2	12
8.83	<2	32	<2	<2	<2	20	8.0	<2	6.7	2740	<2	13
11.95	<2	30	<2	<2	<2	25	3.8	<2	6.7	2553	<2	5.7
12.80	<2	29	<2	<2	<2	18	3.6	<2	<2	2435	<2	11
13.86	<2	29	<2	<2	<2	12	4.2	<2	5.0	2525	<2	13
14.84	<2	28	<2	<2	<2	13	4.4	<2	6.9	2477	<2	8.8
15.85	<2	29	<2	<2	<2	13	4.9	<2	5.7	2538	<2	13
18.93	<2	29	<2	<2	<2	14	5.5	<2	<2	2508	<2	5.3
19.85	<2	28	<2	<2	<2	14	6.3	<2	6.6	2475	<2	6.1
21.84	<2	27	<2	<2	<2	13	7.0	<2	5.7	2497	<2	7.2
22.86	<2	27	<2	<2	<2	12	8.1	<2	5.1	2466	<2	6.5
26.85	<2	27	<2	<2	<2	12	5.3	<2	5.0	2459	<2	5.4
27.84	<2	26	<2	<2	<2	13	8.9	<2	4.9	2458	<2	5.7
28.87	<2	n.d.	n.d.	n.d.	n.d.	12	19	<2	5.5	2475	<2	<2
29.85	<2	35	<2	<2	<2	12	18	<2	5.1	2453	<2	9.6
32.87	15	32	<2	<2	<2	14	26	<2	5.1	2436	<2	9.2
34.86	<2	32	<2	<2	5.6	11	40	<2	5.5	2449	<2	12
35.84	11	31	<2	<2	<2	10	49	<2	6.4	2410	<2	15
39.84	<2	31	<2	<2	<2	13	79	<2	6.8	2412	<2	12
41.81	<2	32	<2	<2	<2	11	111	<2	8.2	2375	<2	14
42.95	<2	33	<2	<2	<2	11	133	<2	6.5	2380	<2	20
46.88	<2	30	<2	<2	<2	17	151	<2	8.2	2359	<2	14
47.78	<2	30	<2	<2	3.4	12	171	<2	7.2	2321	<2	22
48.79	<2	30	<2	<2	<2	11	176	<2	7.0	2326	<2	17
49.80	<2	31	5.7	<2	<2	11	192	<2	8.3	2295	<2	20
50.78	<2	30	7.1	<2	2.9	12	194	<2	8.7	2331	<2	21
53.83	<2	30	<2	<2	<2	13	200	<2	7.4	2291	<2	23
54.84	<2	29	<2	<2	<2	11	211	<2	8.0	2266	<2	25
55.83	<2	29	<2	<2	<2	12	221	<2	8.0	2216	<2	24
56.86	<2	28	<2	<2	3.1	10	218	<2	7.0	2094	<2	24
57.84	<2	26	<2	<2	<2	7.8	175	<2	4.7	1750	<2	19
60.92	<2	26	<2	<2	<2	9.4	87	3.31	3.6	1046	<2	16
61.88	<2	27	3.2	<2	<2	8.6	41	<2	2.9	594	<2	14
64.84	18	32	<2	<2	3.4	9.6	25	5.0	2.7	514	<2	13
67.84	9.8	38	3.5	<2	4.2	10	15	4.1	2.8	468	<2	13
69.87	<2	43	<2	<2	<2	13	11	3.4	2.7	434	<2	9.6
71.84	<2	43	2.0	<2	<2	9.6	9.0	2.9	2.6	409	<2	9.2
74.84	<2	47	<2	<2	<2	9.8	7.0	<2	2.6	418	<2	<2
77.89	2.4	47	<2	<2	<2	9.7	6.8	3.1	2.4	388	<2	5.8
78.82	<2	n.d.	<2	<2	<2	8.8	<2	<2	2.3	n.d.	<2	<2

Time [d]	As [µg/L]	Ba [µg/L]	Cd [µg/L]	Co [µg/L]	Cr [µg/L]	Cu [µg/L]	Mn [µg/L]	Mo [µg/L]	Ni [µg/L]	Sr [µg/L]	V [µg/L]	Zn [µg/L]
81.82	<2	46	<2	<2	<2	9.3	5.8	3.1	2.2	371	<2	7.0
83.82	<2	50	<2	<2	<2	9.3	7.1	3.4	2.4	395	<2	8.3
85.82	<2	51	<2	<2	<2	8.6	5.0	3.4	2.3	402	<2	20
88.87	<2	49	2.1	<2	<2	9.3	5.6	3.7	2.1	383	<2	12
90.83	<2	49	<2	<2	<2	9.1	4.6	3.7	2.2	386	<2	14
92.82	<2	48	2.8	<2	<2	9.4	2.2	3.7	2.1	377	<2	14
98.84	<2	49	<2	<2	<2	14	<2	3.7	2.0	390	<2	4.8
104.84	<2	50	<2	<2	<2	12	<2	3.8	2.1	394	<2	5.0
111.83	<2	46	<2	<2	<2	12	<2	4.0	2.3	373	<2	9.8
118.81	<2	49	<2	<2	<2	11	<2	4.2	2.0	401	<2	22
125.81	5.5	49	<2	<2	2.4	14	<2	4.3	2.7	413	<2	9.5
132.82	<2	48	4.6	<2	<2	12	<2	4.4	2.2	411	<2	6.2
139.83	<2	48	<2	<2	<2	11	<2	4.7	2.2	414	<2	7.2
146.83	2.1	48	<2	<2	<2	11	<2	4.8	2.3	419	<2	8.6
153.82	2.1	45	<2	<2	<2	12	<2	5.0	2.2	396	<2	8.4
160.83	<2	44	<2	<2	<2	11	<2	4.5	2.1	388	<2	8.1
167.84	<2	42	<2	<2	<2	10	<2	4.3	2.0	373	<2	21
174.85	<2	42	<2	<2	<2	10	<2	4.2	<2	377	<2	10
201.74	<2	43	<2	<2	<2	9.8	<2	4.6	<2	390	<2	45
202.84	<2	42	<2	<2	<2	9.1	<2	4.3	<2	397	<2	60
203.60	<2	47	<2	<2	<2	9.7	<2	4.1	<2	425	<2	23
209.83	<2	48	<2	<2	<2	9.8	<2	3.9	<2	449	<2	5.4
216.83	<2	53	<2	<2	<2	9.6	<2	3.7	<2	494	<2	5.7
223.23	<2	55	<2	<2	<2	9.6	<2	3.6	<2	523	<2	5.7
237.23	n.d.	65	<2	<2	<2	9.2	<2	<2	<2	491	<2	7.5

A 1.5.3. Flow-path sampling after 24 days

A 1.5.3.1. Liquid/solid ratio, electric conductivity, pH and E_H value, inorganic carbon, dissolved organic carbon, anions and oxygen saturation

Seepage path [cm]	el. cond. [mS/cm]	pH	E_H [mV]	IC [mg/L]	DOC [mg/L]	Cl [mg/L]	NO ₃ [mg/L]	SO ₄ [mg/L]	Seepage path [cm]	O ₂ -saturation %
21	n.d.	n.d.	n.d.	8.5	5.0	n.d.	n.d.	n.d.	9	24
26	3.5	7.7	238	14	n.d.	2.4	n.d.	88	25	23
29	3.9	7.7	247	n.d.	n.d.	2.3	n.d.	97	37	23
32	4.0	7.8	250	n.d.	n.d.	2.4	n.d.	111	49	19
35	4.3	7.7	240	n.d.	n.d.	2.4	25	125		
38	5.6	7.6	241	32	5.6	3.1	39	178		
41	6.8	7.4	241	11	5.3	4.1	63	232		
44	7.7	7.5	238	13	5.8	4.7	68	250		
47	7.7	7.3	256	20	4.6	4.1	71	239		

A 1.5.3.2. Main elements

Seepage path [cm]	Al [µg/L]	Ca [mg/L]	Fe [µg/L]	K [mg/L]	Mg [mg/L]	Na [mg/L]	S [mg/L]	Si [mg/L]
21	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
26	<2	30	1.7	1.8	1.3	1.3	14	5.9
29	6.4	32	1.5	2.0	1.3	1.2	16	6.0
32	8.8	13	4.2	0.87	0.57	0.55	7.0	2.3
35	4.6	38	2.1	2.3	1.6	1.3	20	6.2
38	6.5	50	1.4	2.7	2.0	1.4	29	6.1
41	<2	63	4.9	3.0	2.5	1.6	38	6.2
44	8.0	73	1.7	3.3	2.8	1.6	42	6.3
47	<2	75	1.0	3.3	2.7	1.6	40	6.7

A 1.5.3.2. Trace elements

Seepage path [cm]	Ba [µg/L]	Cd [µg/L]	Co [µg/L]	Cr [µg/L]	Cu [µg/L]	Mn [µg/L]	Mo [µg/L]	Ni [µg/L]	Sr [µg/L]	V [µg/L]	Zn [µg/L]
21	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
26	18	<2	<2	<2	15	<2	3.4	<2	187	2.0	182
29	20	<2	<2	<2	13	<2	4.7	<2	206	2.2	69
32	22	<2	<2	<2	15	<2	5.3	2.6	228	2.2	27
35	26	<2	<2	<2	15	<2	5.6	<2	253	2.2	24
38	36	<2	<2	<2	12	<2	5.3	2.2	335	<2	32
41	50	<2	<2	<2	11	<2	4.7	3.11	434	<2	48
44	55	<2	<2	<2	10	<2	4.8	2.3	499	<2	26
47	58	<2	<2	<2	8.9	292	4.7	4.7	509	<2	64

A 2. Data for Chapter 3.3.

A 2.1 Demolition waste: Material description

A 2.1.1. Material parameter

Water content	[% weight]	2,2
Specific weight	[g/cm ³]	2,59
Specific surface	[m ² /g]	2,3
pH		11,9
E _H	[mV]	243
El. conductivity	[mS/cm]	3,4
Total carbon	[%]	1.4
Inorganic carbon	[%]	0.73
Organic carbon	[%]	0.67

A 2.1.2. Bulk concentration

[mg/kg]	Al	Ca	Fe	K	Mg	Na	P	S	Si	Cl
Microwave digestion	20679	61215	7529	9318	2954	4423	n.m.	5796	n.m.	n.m.
RFA	18413	105900	8668	8071	5860	n.m.	502	17953	170567	135

[mg/kg]	As	Ba	Cd	Co	Cr	Cu	Mn	Mo	Ni	Pb	Sr	V	Zn
Microwave digestion	n.m.	248	0.89	4.9	20	18	189	1.3	12	21	232	21	71
RFA	3.5	262	n.m.	n.m.	34	18	265	n.m.	8.5	26	238	52	78

A 2.2. Data of the batch experiments with construction waste

A 2.2.1. Electric conductivity, pH and E_H value and acid neutralisation capacity

Sample_ID	el. cond. [mS/cm]	pH	E _H [mV]	ANC/BNC [meq/kg]
pH4-stat	14	4	563	1635
pH5-stat	11	5	450	1372
pH6-stat	11	6	405	1142
pH7-stat	7.4	7	404	717
pH11-stat	3.5	11	294	218
NH ₄ NO ₃	76	9.1	219	n.d.
SSE	39	13	128	n.d.
DEV S4	3.4	12	208	n.d.

A 2.2.2. Main elements

Sample ID	Al	St.Dev.	Ca	St.Dev.	Fe	St.Dev.	K	St.Dev.
	[mg/L]	[µg/L]	[mg/L]	[mg/L]	[mg/L]	[mg/L]	[mg/L]	[mg/L]
pH4-stat	16	1.2	3723	22	2.0	0.45	55	0.89
pH5-stat	0.86	0.13	2766	160	0.15	0.07	50	2.2
pH6-stat	<0.05		2601	133	<0.05		51	1.8
pH7-stat	<0.05		1829	79	<0.05		50	0.78
pH11-stat	<0.05		814	143	<0.05		50	1.9
NH ₄ NO ₃	<0.05		6428	97	<0.05		170	4.8
SSE	0.04		942	19	45		792	10
DEV S4	<0.05		573	11	<0.05		38	0.27

Sample ID	Mg	St.Dev.	Na	St.Dev.	S	St.Dev.	Si	St.Dev.
	[mg/L]	[mg/L]	[mg/L]	[mg/L]	[mg/L]	[mg/L]	[mg/L]	[mg/L]
pH4-stat	68	3.6	13	0.14	322	19	63	2.1
pH5-stat	43	7.1	12	0.05	316	7.3	40	4.6
pH6-stat	24	1.8	12	0.21	323	22	33	1.1
pH7-stat	10	1.4	12	0.22	332	30	23	1.4
pH11-stat	<0.05		11	0.29	289	59	5.9	0.94
NH ₄ NO ₃	8.9	0.12	43	0.62	647	11	5.2	0.15
SSE	<0.05		336	6.6	514	10	1.50	0.05
DEV S4	<0.05		11	0.08	310	12	1.4	0.05

A 2.2.3. Trace elements

Sample_ID	As	St.Dev.	Ba	St.Dev.	Cd	St.Dev.	Co	St.Dev.	Cr	St.Dev.
	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]
pH4-stat	<10		395	7.6	10	0.40	36	1.9	28	0.68
pH5-stat	<10		503	64	<10		20	4.0	28	9.3
pH6-stat	<10		665	56	<10		14	0.30	45	4.8
pH7-stat	<10		413	14	<10		10	0.27	56	2.3
pH11-stat	<10		160	17	<10		9.3	0.41	51	2.6
NH ₄ NO ₃	<10		1353	187	12	0.36	16	0.85	158	1.4
SSE	<10		233	11	<10		19	0.97	266	7.5
DEV S4	<10		93	1.4	<10		8.8	0.16	39	0.57

Sample_ID	Cu	St.Dev.	Mn	St.Dev.	Mo	St.Dev.	Ni	St.Dev.	Pb	St.Dev.
	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]
pH4-stat	107	8.9	2769	72	<10		111	18	<10	
pH5-stat	24	6.3	1169	391	<10		48	12	<10	
pH6-stat	21	2.0	560	51	13	0.78	28	0.97	<10	
pH7-stat	22	0.47	115	11	15	0.59	<10		<10	
pH11-stat	33	2.2	6.5	0.09	14	0.32	<10		<10	
NH ₄ NO ₃	1195	85	19	1.3	27	1.6	70	2.9	<10	
SSE	224	20	<5		63	4.3	52	1.4	<10	
DEV S4	31	1.65	<5		13	0.79	<10		<10	

Sample_ID	Sr	St.Dev.	V	St.Dev.	Zn	St.Dev.
	[$\mu\text{g/L}$]	[$\mu\text{g/L}$]	[$\mu\text{g/L}$]	[$\mu\text{g/L}$]	[$\mu\text{g/L}$]	[$\mu\text{g/L}$]
pH4-stat	12951	116	<8		1826	264
pH5-stat	10558	416	10	0.37	367	164
pH6-stat	10148	202	12	0.38	127	4.6
pH7-stat	7804	316	13	0.24	67	4.5
pH11-stat	5184	488	13	0.88	26	3.0
NH ₄ NO ₃	28090	430	14	0.06	2204	247
SSE	n.d.		<8		12	3.8
DEV S4	4778	43	9.0	0.10	19	2.1

A 2.3. Data of the saturated column experiment with demolition waste

A 2.3.1. Characteristic data of the column

Length	[cm]	48
Diameter	[cm]	5
Volume	[cm ³]	942
Amount material original	[kg]	1.964
Amount material dry	[kg]	1.924
Amount water in column	[L]	n.d.
Vol. inlet/outlet	[L/d]	0.16
Specific weight	[g/cm ³]	2.61
Absolut porosity		0.21
Specific porosity		n.d.
Watersaturation	[% abs. porosity]	n.d.
kf-value	[m/s]	$1.01 \cdot 10^{-4}$
Vf (flow rate 0.1 mL/min)	[cm/d]	8
Va (flow rate 0.1 mL/min)	[m/d]	n.d.

A 2.3.2. Outlet

A 2.3.2.1. Liquid/solid ratio, electric conductivity, pH and E_H value, inorganic carbon, dissolved organic carbon and anions

Time	L/S	el. cond.	pH	E _H	IC	DOC	Cl	NO ₃	SO ₄
[d]	[L/kg]	[mS/cm]		[mV]	[mg/L]	[mg/L]	[mg/L]	[mg/L]	[mg/L]
0.24	0.02	39	12.5	138	14	378	172	<0.6	2301
0.42	0.03	n.d.	n.d.	n.d.	18	297	145	<0.6	2109
0.56	0.04	n.d.	n.d.	n.d.	33	290	142	<0.6	2055
0.71	0.05	n.d.	n.d.	n.d.	21	276	141	<0.6	2078
0.85	0.06	n.d.	n.d.	n.d.	14	270	139	<0.6	2048
1.00	0.07	n.d.	n.d.	n.d.	10	267	143	<0.6	2086
1.15	0.08	42	12.5	138	19	260	145	<0.6	2120
1.42	0.10	n.d.	n.d.	n.d.	46	256	142	<0.6	2006
1.56	0.11	n.d.	n.d.	n.d.	42	236	144	<0.6	2124
1.71	0.13	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
2.00	0.15	n.d.	n.d.	n.d.	22	138	99	<0.6	1972
2.15	0.16	n.d.	n.d.	n.d.	30	106	71	<0.6	2067
2.56	0.19	n.d.	n.d.	n.d.	14	70	35	<0.6	1651
3.15	0.23	n.d.	n.d.	n.d.	24	62	26	<0.6	1494
3.56	0.26	n.d.	n.d.	n.d.	23	65	18	<0.6	1343

Time [d]	L/S [L/kg]	el. cond. [mS/cm]	pH	E _H [mV]	IC [mg/L]	DOC [mg/L]	Cl [mg/L]	NO ₃ [mg/L]	SO ₄ [mg/L]
4.15	0.30	n.d.	n.d.	n.d.	17	52	14	<0.6	1313
4.56	0.33	42	12.3	182	30	46	11	<0.6	1266
5.15	0.38	n.d.	n.d.	n.d.	16	38	9.4	<0.6	1213
5.56	0.41	n.d.	n.d.	n.d.	17	45	7.8	<0.6	1229
6.15	0.45	41	12.3	188	23	40	6.5	<0.6	1238
6.56	0.48	n.d.	n.d.	n.d.	50	47	5.5	<0.6	1224
7.15	0.52	41	12.3	205	19	44	3.3	<0.6	1232
7.56	0.55	n.d.	n.d.	n.d.	29	59	3.3	<0.6	1208
8.15	0.60	40	12.2	209	14	30	5.6	<0.6	1179
9.20	0.67	40	12.3	211	17	38	3.5	<0.6	1240
9.56	0.70	n.d.	n.d.	n.d.	16	36	3.2	<0.6	1324
11.54	0.75	n.d.	n.d.	n.d.	18	41	3.1	<0.6	1248
11.69	0.76	n.d.	n.d.	n.d.	12	38	3.8	<0.6	1220
11.83	0.77	n.d.	n.d.	n.d.	14	37	3.2	<0.6	1253
11.98	0.78	n.d.	n.d.	n.d.	24	45	3.5	<0.6	1310
12.13	0.79	39	12.2	219	24	30	2.4	<0.6	1261
12.56	0.82	n.d.	n.d.	n.d.	12	34	3.5	<0.6	1322
13.15	0.86	40	12.2	224	20	34	1.3	<0.6	1298
13.56	0.89	n.d.	n.d.	n.d.	9.2	31	0.69	<0.6	1311
14.15	0.94	n.d.	n.d.	n.d.	7.3	26	0.34	<0.6	1267
14.56	0.97	n.d.	n.d.	n.d.	7.7	24	<0.3	<0.6	1264
15.15	1.01	38	12.3	227	23	23	<0.3	<0.6	1269
15.56	1.04	n.d.	n.d.	n.d.	14	21	0.57	<0.6	1270
16.15	1.08	37	12.3	262	13	24	<0.3	<0.6	1271
16.56	1.11	n.d.	n.d.	n.d.	23	19	<0.3	<0.6	1351
17.15	1.16	n.d.	n.d.	n.d.	16	22	<0.3	<0.6	1498
17.56	1.19	n.d.	n.d.	n.d.	25	25	<0.3	<0.6	1272
18.15	1.23	n.d.	n.d.	n.d.	17	28	<0.3	<0.6	1214
18.56	1.26	n.d.	n.d.	n.d.	15	18	<0.3	<0.6	1242
19.15	1.30	37	12.2	174	10	27	<0.3	<0.6	1203
19.56	1.33	n.d.	n.d.	n.d.	17	23	<0.3	<0.6	1208
20.15	1.38	37	12.2	203	14	18	<0.3	<0.6	1195
20.56	1.41	n.d.	n.d.	n.d.	11	25	<0.3	<0.6	1204
21.15	1.45	37	12.2	217	15	23	<0.3	<0.6	1242
21.56	1.48	n.d.	n.d.	n.d.	12	21	<0.3	<0.6	1148
22.15	1.52	36	12.2	219	11	19	<0.3	<0.6	1196
22.56	1.55	n.d.	n.d.	n.d.	26	22	<0.3	<0.6	1221
23.15	1.60	36	12.1	213	13	17	<0.3	<0.6	1206
23.56	1.63	n.d.	n.d.	n.d.	5.2	18	<0.3	<0.6	1266
24.15	1.67	n.d.	n.d.	n.d.	6.6	21	<0.3	<0.6	1268
24.56	1.70	n.d.	n.d.	n.d.	17	20	<0.3	<0.6	1269
25.15	1.74	n.d.	n.d.	n.d.	8.2	23	<0.3	<0.6	1280
25.56	1.77	n.d.	n.d.	n.d.	25	19	5.7	<0.6	1362
26.15	1.82	36	12.2	221	14	20	4.8	<0.6	1293
26.56	1.85	n.d.	n.d.	n.d.	36	25	4.5	<0.6	1513
27.15	1.89	36	12.2	n.d.	35	19	7.0	<0.6	1492
28.15	1.96	n.d.	n.d.	n.d.	23	19	5.4	<0.6	1475
28.56	1.99	n.d.	n.d.	n.d.	32	18	7.3	<0.6	1452
29.15	2.04	35	12.1	n.d.	38	20	n.d.	n.d.	n.d.
29.56	2.07	n.d.	n.d.	n.d.	27	15	4.7	<0.6	1395
30.17	2.11	35	12.1	n.d.	21	16	3.2	<0.6	1394
30.56	2.14	n.d.	n.d.	n.d.	17	18	3.1	<0.6	1469
31.15	2.18	n.d.	n.d.	n.d.	26	16	6.9	<0.6	1436
31.56	2.21	n.d.	n.d.	n.d.	30	13	3.7	<0.6	1423
32.15	2.26	n.d.	n.d.	n.d.	27	16	3.9	<0.6	1385
32.56	2.29	n.d.	n.d.	n.d.	6.3	16	1.7	<0.6	1340
33.15	2.33	34	12.1	n.d.	14	17	1.7	<0.6	1304
33.56	2.36	n.d.	n.d.	n.d.	23	10	4.9	<0.6	1284

Time [d]	L/S [L/kg]	el. cond. [mS/cm]	pH	E _H [mV]	IC [mg/L]	DOC [mg/L]	Cl [mg/L]	NO ₃ [mg/L]	SO ₄ [mg/L]
34.15	2.40	34	12.1	n.d.	12	12	2.7	<0.6	1301
34.56	2.43	n.d.	n.d.	n.d.	10	12	2.3	<0.6	1302
35.21	2.48	n.d.	n.d.	n.d.	7.1	17	3.4	<0.6	1268
35.56	2.51	n.d.	n.d.	n.d.	42	19	2.3	<0.6	1267
36.15	2.55	35	12.1	n.d.	11	20	4.5	<0.6	1233
37.17	2.62	34	12.1	n.d.	11	16	8.0	<0.6	1166
38.15	2.70	n.d.	n.d.	n.d.	46	17	3.2	<0.6	1202
39.15	2.77	n.d.	n.d.	n.d.	6	14	4.6	<0.6	1117
40.15	2.84	33	12.1	n.d.	14	14	4.5	<0.6	947
41.15	2.92	n.d.	n.d.	n.d.	13	15	4.6	<0.6	937
42.15	2.99	31	12.2	211	14	12	6.4	<0.6	798
43.15	3.06	32	12.2	203	27	14	7.2	<0.6	749
44.17	3.14	32	12.0	200	25	12	5.7	<0.6	664
45.15	3.21	n.d.	n.d.	n.d.	22	13	12.72	<0.6	598
46.15	3.28	n.d.	n.d.	n.d.	9.3	11	2.9	<0.6	531
47.15	3.36	31	12.2	203	8.9	14	3.1	<0.6	451
48.15	3.43	31	12.1	197	33	15	2.9	<0.6	411
49.15	3.50	30	12.2	209	14	14	<0.3	<0.3	313
51.15	3.65	n.d.	n.d.	n.d.	12	23	3.7	<0.3	334
52.15	3.72	n.d.	n.d.	n.d.	4.0	16	2.8	<0.3	281
53.15	3.80	n.d.	n.d.	n.d.	31	25	3.3	<0.3	277
54.15	3.87	30	12.2	232	22	36	3.4	<0.3	315
55.15	3.94	30	12.2	198	n.d.	n.d.	n.d.	n.d.	n.d.
56.15	4.02	29	12.1	207	19	18	3.9	1.8	260
57.15	4.09	28	12.2	206	11	19	4.6	<0.3	309
58.17	4.16	30	12.1	187	18	25	5.0	<0.3	298
59.15	4.24	n.d.	n.d.	n.d.	18	19	6.7	<0.3	382
60.15	4.31	n.d.	n.d.	n.d.	7.0	14	4.0	<0.3	267
61.15	4.38	29	12.1	172	10	15	6.5	<0.3	347
62.15	4.46	28	12.2	189	13	12	5.1	<0.3	304
63.15	4.53	28	12.2	207	17	13	3.9	<0.3	232
64.15	4.60	28	12.1	139	17	14	3.4	<0.3	178
65.15	4.68	28	12.1	153	18	15	4.6	<0.3	217
66.15	4.75	n.d.	n.d.	n.d.	12	22	4.0	<0.3	218
67.15	4.82	n.d.	n.d.	n.d.	7	19	3.6	<0.3	177
68.15	4.90	n.d.	n.d.	n.d.	21	12	3.4	0.75	162
69.15	4.97	27	12.0	220	12	17	4.9	0.31	245
70.15	5.04	n.d.	n.d.	n.d.	11	8	14.69	3.7	282
72.15	5.19	n.d.	n.d.	n.d.	65	19	3.9	0.51	168
75.15	5.41	27	12.1	212	63	13	3.8	2.1	167
77.15	5.56	26	12.1	224	26	15	3.1	0.58	143
79.15	5.70	n.d.	n.d.	n.d.	13	17	4.0	3.3	144
82.15	5.92	26	12.0	238	n.d.	n.d.	5.2	1.9	230
85.15	6.14	25	12.1	242	10	17	5.3	2.2	188
86.15	6.22	25	12.1	243	61	24	4.7	0.72	139
89.15	6.44	25	12.1	279	124	21	3.8	1.9	169
91.15	6.58	24	12.0	240	102	24	5.0	2.9	195
93.15	6.73	23	12.1	241	102	22	7.0	4.2	257
96.15	6.95	24	12.1	236	108	16	4.4	3.6	201
98.15	7.10	24	12.1	249	118	17	n.d.	n.d.	n.d.
100.15	7.24	24	12.1	316	83	17	4.8	2.9	202
106.15	7.68	22	12.0	178	30	13	6.6	3.6	237
112.15	8.12	22	11.9	184	19	15	5.5	4.7	258
119.15	8.64	22	12.0	187	23	13	6.3	5.9	197
126.15	9.15	21	12.0	203	103	21	5.2	4.5	166
133.15	9.66	21	12.1	203	14	18	7.2	7.4	317
140.15	10.17	20	12.0		12	16	n.d.	n.d.	n.d.
147.15	10.69	20	11.9	224	86	27	2.6	1.7	149

Time [d]	L/S [L/kg]	el. cond. [mS/cm]	pH	E _H [mV]	IC [mg/L]	DOC [mg/L]	Cl [mg/L]	NO ₃ [mg/L]	SO ₄ [mg/L]
154.15	11.20	19	11.9	182	73	22	n.d.	n.d.	n.d.
161.15	11.71	19	12.0	190	71	15	n.d.	n.d.	n.d.
168.15	12.23	19	12.0	201	90	13	n.d.	n.d.	n.d.
175.15	12.74	19	12.0	207	68	16	n.d.	n.d.	n.d.
182.15	13.25	18	12.0	193	n.d.	n.d.	n.d.	n.d.	n.d.

A 2.3.2.2. Main elements

Time [d]	Al [µg/L]	Ca [mg/L]	Fe [µg/L]	K [mg/L]	Mg [µg/L]	Na [mg/L]	Si [mg/L]
0.24	70	782	58	1522	44	636	3.4
0.42	441	698	447	1246	223	493	3.2
0.56	27	789	<10	1215	15	485	1.9
0.71	34	780	14	1204	17	475	1.8
0.85	31	788	9.4	1184	13	463	1.9
1.00	26	788	136	1165	15	455	1.9
1.15	35	779	14	1165	21	455	1.8
1.42	29	724	<10	1187	12	459	1.7
1.56	34	720	13	1164	124	449	1.7
1.71	48	668	17	1148	100	428	1.6
2.00	20	730	<10	1141	22	355	1.1
2.15	45	822	30	1083	28	233	1.0
2.56	33	899	16	671	23	88	0.87
3.15	18	849	<10	487	20	56	0.67
3.56	19	853	<10	327	16	36	0.63
4.15	16	859	<10	258	17	29	0.58
4.56	24	904	<10	208	15	23	0.65
5.15	15	871	<10	187	15	21	0.55
5.56	14	852	<10	164	<10	19	0.54
6.15	29	870	<10	138	13	17	0.53
6.56	24	830	<10	125	17	16	0.49
7.15	13	886	<10	110	15	15	0.53
7.56	14	901	<10	105	17	15	0.63
8.15	12	887	<10	95	18	14	0.58
9.20	33	839	<10	104	17	17	0.69
9.56	16	736	<10	101	18	17	0.55
11.54	35	810	<10	94	17	17	0.70
11.69	22	778	<10	96	19	18	0.67
11.83	25	828	<10	93	17	18	0.63
11.98	14	830	<10	90	15	18	0.63
12.13	20	824	<10	87	16	17	0.61
12.56	13	873	<10	84	17	17	0.62
13.15	17	886	<10	74	19	15	0.62
13.56	9.1	880	<10	70	20	14	0.65
14.15	6.5	884	<10	62	16	12	0.63
14.56	18	884	<10	62	16	11	0.68
15.15	13	904	<10	56	17	9.9	0.65
15.56	8.6	908	<10	53	<10	9.6	0.00
16.15	16	900	<10	48	19	9.0	0.66
16.56	24	749	<10	49	17	9.2	0.58
17.15	<10	733	<10	46	19	8.8	0.59
17.56	30	748	<10	39	22	7.6	0.67
18.15	41	809	<10	37	19	7.5	0.60
18.56	31	821	<10	36	19	7.5	0.62
19.15	41	808	<10	34	20	7.3	0.61

Time [d]	Al [µg/L]	Ca [mg/L]	Fe [µg/L]	K [mg/L]	Mg [µg/L]	Na [mg/L]	Si [mg/L]
19.56	32	820	<10	33	19	7.3	0.63
20.15	39	807	<10	31	26	7.1	0.62
20.56	27	754	<10	30	21	7.1	0.62
21.15	51	807	<10	29	22	6.8	0.63
21.56	33	805	<10	29	18	6.9	0.63
22.15	45	808	<10	28	20	6.7	0.64
22.56	38	771	<10	28	22	6.8	0.65
23.15	30	802	<10	27	21	6.7	0.66
23.56	27	791	<10	28	23	7.1	0.70
24.15	35	731	<10	27	21	7.0	0.66
24.56	38	741	<10	26	24	6.8	0.67
25.15	24	720	<10	25	23	6.5	0.67
25.56	30	783	<10	24	24	6.4	0.68
26.15	27	800	<10	23	21	6.1	0.65
26.56	23	772	18	23	20	6.2	0.69
27.15	33	795	12	22	19	6.0	0.69
28.15	28	660	103	23	17	6.4	0.65
28.56	34	739	12	22	17	6.4	0.70
29.15	31	780	<10	21	20	6.0	0.66
29.56	33	785	10	21	19	6.2	0.68
30.17	30	779	<10	20	19	6.0	0.68
30.56	31	698	<10	21	21	6.2	0.62
31.15	29	716	18	20	20	6.0	0.65
31.56	28	773	<10	20	21	5.8	0.68
32.15	28	772	<10	19	20	5.6	0.67
32.56	28	770	<10	18	19	5.5	0.68
33.15	26	766	<10	17	21	5.2	0.66
33.56	22	765	<10	17	20	5.4	0.66
34.15	24	754	<10	16	21	5.1	0.67
34.56	38	765	<10	16	19	5.2	0.69
35.21	30	761	13	16	23	5.2	0.70
35.56	32	736	11	16	19	5.3	0.69
36.15	35	734	33	15	20	5.0	0.68
37.17	24	721	<10	15	22	5.1	0.69
38.15	39	631	<10	15	17	5.3	0.64
39.15	33	651	<10	14	20	5.0	0.68
40.15	29	642	<10	14	18	4.7	0.69
41.15	36	603	<10	14	18	5.0	0.72
42.15	32	582	<10	13	<10	4.7	0.67
43.15	37	557	11	13	14	4.7	0.69
44.17	30	530	10	12	13	4.6	0.67
45.15	34	478	<10	13	15	4.9	0.69
46.15	28	449	<10	12	12	4.6	0.68
47.15	46	458	<10	12	13	4.2	0.68
48.15	36	427	<10	12	12	4.5	0.71
49.15	77	392	11	11	16	4.4	0.79
51.15	63	357	<10	12	12	4.9	0.76
52.15	60	297	<10	11	11	4.5	0.70
53.15	52	469	<10	11	12	4.7	0.86
54.15	52	353	10	11	10	4.9	0.70
55.15	48	351	11	12	<10	5.7	0.75
56.15	48	331	<10	12	<10	5.6	0.73
57.15	54	334	106	12	<10	5.1	0.74
58.17	58	329	14	11	<10	4.6	0.73
59.15	67	292	11	12	<10	4.8	0.86
60.15	62	335	10	11	<10	4.4	0.90
61.15	72	330	12	10	<10	4.2	0.87
62.15	68	322	<10	9.86	<10	4.1	0.87

Time [d]	Al [µg/L]	Ca [mg/L]	Fe [µg/L]	K [mg/L]	Mg [µg/L]	Na [mg/L]	Si [mg/L]
63.15	68	324	21	9.69	<10	4.0	0.90
64.15	78	315	<10	9.32	<10	3.9	0.89
65.15	77	311	<10	9.08	<10	3.9	0.89
66.15	84	288	<10	9.64	<10	4.3	0.96
67.15	93	307	<10	9.24	<10	4.1	0.98
68.15	90	305	<10	8.69	<10	3.9	0.92
69.15	94	299	19	8.50	<10	3.9	0.95
70.15	144	300	15	8.41	<5	3.8	0.95
71.15	77	304	<1	8.63	6.6	3.8	0.99
72.15	74	306	<1	8.43	<5	3.9	1.0
73.15	77	219	1.3	8.52	6.4	4.0	1.0
74.15	83	293	3.5	8.63	7.2	4.0	1.1
75.15	79	296	<1	7.8	<5	3.7	1.1
76.15	83	291	1.2	7.8	6.6	3.7	1.1
77.15	83	291	<1	7.6	<5	3.6	1.1
78.15	103	293	<1	7.8	6.8	3.8	1.1
79.15	89	295	<1	7.7	<5	3.9	1.1
80.15	98	236	<1	8.3	<5	4.2	1.2
81.15	105	270	<1	8.3	<5	4.4	1.2
82.15	101	288	<1	8.3	<5	4.6	1.2
83.15	117	290	1.0	8.8	7.1	4.6	1.2
84.15	105	286	<1	7.7	6.6	3.9	1.2
85.15	112	281	<1	7.4	<5	3.7	1.2
86.15	110	270	<1	7.1	<5	3.4	1.2
87.15	123	224	<1	7.6	6.6	3.7	1.2
88.15	127	260	<1	7.3	6.5	3.5	1.2
90.15	132	257	<1	6.8	6.3	3.4	1.2
91.15	125	267	<1	6.4	<5	3.2	1.2
92.15	143	258	<1	7.0	6.7	3.5	1.3
93.15	138	270	<1	6.4	<5	3.3	1.3
94.15	152	259	13	7.0	6.7	3.6	1.4
95.15	160	273	<1	7.2	8.8	4.4	1.5
96.15	143	266	<1	6.1	<5	3.2	1.3
97.15	152	244	<1	6.5	6.3	3.4	1.4
98.15	144	260	<1	5.8	<5	3.1	1.3
99.15	156	256	<1	6.2	6.8	3.2	1.4
100.15	144	257	<1	5.6	<5	2.9	1.3
102.15	187	264	<1	7.0	<5	3.7	1.6
103.15	189	266	<1	6.6	<5	3.5	1.6
104.15	179	270	<1	6.3	7.0	3.2	1.6
105.15	172	258	1.3	6.0	6.9	3.1	1.5
105.15	154	n.d.	<1	n.d.	7.0	n.d.	n.d.
106.15	165	257	<1	5.5	<5	3.1	1.5
107.15	170	n.d.	<1	n.d.	7.6	n.d.	n.d.
108.15	168	n.d.	<1	n.d.	7.5	n.d.	n.d.
109.15	161	n.d.	<1	n.d.	7.3	n.d.	n.d.
119.15	170	253	<1	5.5	8.2	3.1	1.6
111.15	182	251	<1	5.5	6.6	3.0	1.6
112.15	176	247	<1	5.3	<5	2.9	1.6
114.15	171	n.d.	1.0	n.d.	7.2	n.d.	n.d.
117.15	177	253	<1	5.5	6.6	3.0	1.6
119.15	184	239	<1	5.0	<5	2.8	1.6
121.15	n.d.	n.d.	<1	n.d.	5.9	n.d.	n.d.
126.15	196	235	<1	4.9	<5	2.8	1.7
128.15	257	605	<1	6.1	<5	3.5	2.6
133.15	214	245	1.2	4.5	7.3	2.7	1.8
135.15	264	279	<1	5.7	7.3	3.4	2.2
140.15	221	233	<1	4.6	6.5	2.8	1.9

Time [d]	Al [µg/L]	Ca [mg/L]	Fe [µg/L]	K [mg/L]	Mg [µg/L]	Na [mg/L]	Si [mg/L]
142.15	282	268	3.0	5.6	14.5	3.6	2.3
147.15	241	224	1.7	4.5	6.6	2.8	1.9
149.15	284	258	1.0	5.4	7.3	3.3	2.4
154.15	254	218	<1	4.2	6.6	2.6	2.0
161.15	269	219	<1	4.0	6.6	2.5	2.1
168.15	279	208	<1	3.6	6.5	2.3	2.2
175.15	293	206	<1	3.3	6.2	2.2	2.3
182.15	311	199	<1	3.1	7.7	2.2	2.4

A 2.3.2.3. Trace elements

Time [d]	Ba [µg/L]	Cd [µg/L]	Co [µg/L]	Cr [µg/L]	Cu [µg/L]	Mn [µg/L]	Mo [µg/L]	Ni [µg/L]	Sr [mg/L]	V [µg/L]	Zn [µg/L]
0.24	321	<10	30	497	515	<10	131	99	20	<10	54
0.42	274	<10	24	441	411	<10	112	83	19	<10	51
0.56	302	<10	24	420	408	<10	108	82	21	<10	43
0.71	298	<10	24	417	403	<10	108	84	21	<10	43
0.85	294	<10	23	408	390	<10	104	81	21	<10	43
1.00	292	<10	24	405	393	80	102	84	20	<10	40
1.15	289	<10	23	405	387	<10	102	81	21	<10	44
1.42	273	<10	23	416	388	<10	105	83	21	<10	38
1.56	270	<10	22	412	369	<10	105	80	21	<10	36
1.71	245	<10	19	412	347	<10	104	78	19	<10	40
2.00	258	<10	14	401	298	<10	103	68	22	<10	34
2.15	272	<10	11	411	256	<10	104	63	25	<10	45
2.56	259	<10	7.0	313	176	<10	76	45	25	<10	45
3.15	233	<10	5.9	283	154	<10	69	40	25	<10	39
3.56	219	<10	<5	236	127	<10	59	34	25	<10	39
4.15	215	<10	<5	222	119	<10	57	32	25	<10	41
4.56	217	<10	<5	209	107	<10	53	29	24	<10	39
5.15	210	<10	<5	196	101	<10	51	27	24	<10	38
5.56	203	<10	<5	196	98	<10	52	26	24	<10	38
6.15	205	<10	<5	195	94	<10	47	25	24	<10	40
6.56	198	<10	<5	192	92	<10	47	25	25	<10	35
7.15	206	<10	<5	188	86	<10	46	24	25	<10	37
7.56	207	<10	<5	189	87	<10	47	24	26	<10	40
8.15	207	<10	<5	184	83	<10	44	24	25	<10	38
9.20	211	<10	<5	172	89	<10	42	27	25	<10	38
9.56	181	<10	<5	170	91	<10	45	24	25	<10	<10
11.54	196	<10	<5	149	88	<10	41	25	25	<10	53
11.69	197	<10	<5	128	90	<10	40	25	24	<10	<10
11.83	206	<10	<5	126	89	<10	39	24	25	<10	37
11.98	206	<10	<5	121	88	<10	39	24	25	<10	36
12.13	203	<10	<5	122	86	<10	39	23	25	<10	35
12.56	200	<10	<5	119	84	<10	36	24	26	<10	37
13.15	199	<10	<5	115	76	<10	33	21	25	<10	37
13.56	195	<10	<5	107	71	<10	30	20	25	<10	39
14.15	194	<10	<5	104	64	<10	28	20	24	<10	37
14.56	193	<10	<5	106	63	<10	28	20	25	<10	38
15.15	195	<10	<5	104	59	<10	25	19	24	<10	37
15.56	195	<10	<5	103	58	<10	25	18	24	<10	39
16.15	189	<10	<5	103	54	<10	22	19	23	<10	38
16.56	149	<10	<5	113	57	<10	24	18	22	<10	32
17.15	144	<10	<5	107	53	<10	23	17	21	<10	29
17.56	154	<10	<5	119	51	<10	27	17	19	<10	30

Time [d]	Ba [µg/L]	Cd [µg/L]	Co [µg/L]	Cr [µg/L]	Cu [µg/L]	Mn [µg/L]	Mo [µg/L]	Ni [µg/L]	Sr [mg/L]	V [µg/L]	Zn [µg/L]
18.15	164	<10	<5	118	50	<10	25	19	19	<10	28
18.56	166	<10	<5	114	48	<10	23	18	19	<10	32
19.15	168	<10	<5	112	47	<10	22	17	18	<10	32
19.56	170	<10	<5	110	47	<10	21	17	18	<10	31
20.15	167	<10	<5	108	46	<10	19	17	17	<10	35
20.56	151	<10	<5	103	45	<10	22	16	16	<10	34
21.15	163	<10	<5	103	44	<10	19	18	16	<10	29
21.56	157	<10	<5	98	44	<10	18	16	16	<10	33
22.15	161	<10	<5	94	42	<10	17	17	15	<10	32
22.56	150	<10	<5	94	43	<10	18	15	15	<10	29
23.15	162	<10	<5	87	41	<10	16	14	14	<10	32
23.56	150	<10	<5	91	44	<10	17	16	15	<10	32
24.15	132	<10	<5	88	42	<10	19	13	14	<10	28
24.56	138	<10	<5	85	41	<10	18	14	13	<10	28
25.15	135	<10	<5	76	39	<10	17	13	12	<10	34
25.56	149	<10	<5	77	38	<10	17	14	12	<10	31
26.15	152	<10	<5	71	38	<10	15	15	12	<10	31
26.56	143	<10	<5	70	37	<10	17	14	12	<10	29
27.15	149	<10	<5	68	36	<10	15	14	11	<10	34
28.15	116	<10	<5	67	38	<10	16	13	11	<10	<10
28.56	139	<10	<5	67	36	<10	15	13	11	<10	<10
29.15	151	<10	<5	59	35	<10	13	14	11	<10	<10
29.56	145	<10	<5	60	35	<10	13	13	11	<10	<10
30.17	145	<10	<5	57	32	<10	13	12	10	<10	<10
30.56	127	<10	<5	56	34	<10	14	14	10	<10	14
31.15	131	<10	<5	54	33	<10	15	12	10	<10	<10
31.56	141	<10	<5	51	31	<10	13	12	9.9	<10	<10
32.15	141	<10	<5	49	30	<10	12	10	9.5	<10	<10
32.56	140	<10	<5	43	30	<10	11	8.2	9.2	<10	<10
33.15	139	<10	<5	43	47	<10	10	10	8.9	<10	22
33.56	137	<10	<5	39	28	<10	10	9.8	8.8	<10	<10
34.15	139	<10	<5	38	27	<10	<10	11	8.4	<10	<10
34.56	137	<10	<5	43	28	<10	11	11	8.5	<10	<10
35.21	139	<10	<5	37	26	<10	<10	9.1	8.2	<10	<10
35.56	131	<10	<5	36	27	<10	<10	11	8.1	<10	<10
36.15	135	<10	<5	36	27	<10	<10	10	7.8	<10	14
37.17	134	<10	<5	31	24	<10	<10	8.6	7.6	<10	<10
38.15	121	<10	<5	35	25	<10	<10	9.0	7.5	<10	<10
39.15	128	<10	<5	28	22	<10	<10	8.3	7.3	<10	<10
40.15	135	<10	<5	27	23	<10	<10	7.1	7.0	<10	12
41.15	135	<10	<5	27	23	<10	<10	6.9	7.1	<10	<10
42.15	140	<10	<5	23	21	<10	<10	7.1	6.9	<10	<10
43.15	147	<10	<5	23	21	<10	<10	6.5	6.9	<10	<10
44.17	152	<10	<5	21	20	<10	<10	4.5	6.9	<10	<10
45.15	156	<10	<5	22	21	<10	<10	7.1	7.1	<10	14
46.15	165	<10	<5	20	20	<10	<10	4.2	6.8	<10	12
47.15	179	<10	<5	23	19	<10	<10	5.8	6.6	<10	<10
48.15	192	<10	<5	20	19	<10	<10	4.8	6.6	<10	<10
49.15	215	<10	<5	22	17	<10	<10	5.3	5.9	<10	<10
51.15	221	<10	<5	30	19	<10	<10	5.2	6.1	<10	<10
52.15	191	<10	<5	28	18	<10	<10	<4	5.1	<10	<10
53.15	243	<10	<5	25	19	<10	<10	6.1	5.3	<10	13
54.15	229	<10	<5	25	32	<10	<10	4.3	5.1	<10	12
55.15	233	<10	<5	23	19	<10	<10	4.0	5.2	<10	<10
56.15	243	<10	<5	22	19	<10	<10	<4	4.8	<10	<10
57.15	259	<10	<5	24	18	<10	<10	4.8	4.7	<10	<10
58.17	262	<10	<5	24	19	<10	<10	<4	4.4	<10	<10
59.15	258	<10	<5	28	19	<10	<10	4.5	n.d.	<10	<10

Time [d]	Ba [µg/L]	Cd [µg/L]	Co [µg/L]	Cr [µg/L]	Cu [µg/L]	Mn [µg/L]	Mo [µg/L]	Ni [µg/L]	Sr [mg/L]	V [µg/L]	Zn [µg/L]
60.15	273	<10	<5	23	19	<10	<10	<4	n.d.	<10	<10
61.15	285	<10	<5	27	18	<10	<10	6.2	n.d.	<10	<10
62.15	280	<10	<5	28	18	<10	<10	<4	n.d.	<10	<10
63.15	282	<10	<5	25	17	<10	<10	<4	n.d.	<10	<10
64.15	283	<10	<5	25	17	<10	<10	<4	n.d.	<10	16
65.15	282	<10	<5	26	17	<10	<10	<4	n.d.	<10	<10
66.15	260	<10	<5	29	17	<10	<10	<4	n.d.	<10	<10
67.15	263	<10	<5	32	18	<10	<10	<4	n.d.	<10	<10
68.15	272	<10	<5	29	20	<10	<10	<4	n.d.	<10	<10
69.15	275	17	<5	28	16	<10	<10	<4	n.d.	<10	<10
70.15	272	9.7	<2	31	16	<2	<2	<2	n.d.	<2	<2
71.15	215	<2	<2	19	12	<2	4.6	2.5	n.d.	<2	3.47
73.15	174	2.4	<2	20	13	<2	3.8	2.3	n.d.	<2	4.29
74.15	210	6.2	<2	20	12	<2	3.5	2.5	n.d.	<2	3.91
76.15	205	<2	<2	20	12	<2	3.2	2.5	n.d.	<2	3.7
78.15	203	<2	<2	20	12	<2	3.2	2.2	n.d.	<2	3.5
83.15	198	<2	<2	23	14	<2	3.4	2.7	n.d.	<2	3.7
84.15	194	<2	<2	21	12	<2	3.1	2.4	n.d.	<2	3.5
87.15	178	<2	<2	23	13	<2	3.2	2.3	n.d.	<2	3.2
88.15	182	<2	<2	22	12	<2	2.9	2.3	n.d.	<2	3.4
90.15	179	<2	<2	21	12	<2	2.7	2.3	n.d.	<2	3.3
92.15	176	<2	<2	22	12	<2	2.8	2.3	n.d.	<2	3.4
94.15	177	<2	<2	22	12	<2	2.8	2.2	n.d.	<2	3.4
95.15	180	<2	<2	23	13	<2	2.7	2.6	n.d.	<2	436
97.15	174	<2	<2	21	11	<2	2.5	2.2	n.d.	<2	3.1
99.15	169	<2	<2	20	11	<2	2.4	2.3	n.d.	<2	3.3
104.15	167	<2	<2	20	12	<2	2.3	2.2	n.d.	<2	3.8
105.15	168	<2	<2	20	12	<2	3.1	2.3	1.20	<2	3.7
105.15	153	<2	<2	19	11	<2	2.3	2.1	1.12	<2	2.7
107.15	168	<2	<2	20	12	<2	2.3	2.3	1.19	<2	3.0
108.15	163	<2	<2	20	12	<2	2.2	2.2	1.11	<2	3.1
109.15	160	<2	<2	19	11	<2	2.1	2.0	1.10	<2	2.7
119.15	161	<2	<2	18	11	<2	<2	2.0	1.08	<2	2.8
111.15	158	<2	<2	18	11	<2	2.0	2.0	1.06	<2	2.8
114.15	158	<2	<2	19	12	<2	2.1	2.2	1.07	<2	3.0
117.15	160	<2	<2	17	11	<2	<2	2.1	1.03	<2	2.6
121.15	123	<2	<2	20	9.3	<2	2.3	2.1	0.87	<2	<2
133.15	149	<2	<2	15	10	<2	<2	2.0	0.86	<2	2.9
135.15	171	<2	<2	17	12	<2	<2	2.2	1.0	<2	2.7
140.15	146	<2	<2	14	11	<2	<2	<2	0.81	<2	2.6
142.15	162	<2	<2	16	13	<2	<2	2.3	0.93	<2	3.7
147.15	138	<2	<2	14	11	<2	<2	<2	0.76	<2	2.5
149.15	162	<2	<2	16	12	<2	<2	2.0	0.89	<2	2.6
154.15	135	<2	<2	13	10	<2	<2	<2	0.72	<2	2.4
161.15	132	<2	<2	13	10	<2	<2	<2	0.69	<2	3.2
168.15	126	<2	<2	11	9.3	<2	<2	<2	0.65	<2	2.3
175.15	126	<2	<2	11	9.4	<2	<2	<2	0.61	<2	2.5
182.15	120	<2	<2	10	8.7	<2	<2	2.8	0.54	<2	3.1

A 3. Data for Chapter 3.4.

A 3.1 Municipal waste incineration ash: Material description

A 3.1.1. Material parameter

Water content	[% weight]	3,7
Specific weight	[g/cm ³]	2,62
Specific surface	[m ² /g]	5,4
pH		11,4
E _H	[mV]	304
El. conductivity	[mS/cm]	1,2
Total carbon	[%]	0,9
Inorganic carbon	[%]	0,46
Organic carbon	[%]	0,44

A 3.1.2. Bulk concentration

[mg/kg]	Al	Ca	Fe	K	Mg	Na	P	S	Si	Cl
Microwave digestion	35771	70155	55110	9245	9620	22225	n.d.	4158	n.d.	n.d.
RFA	41497	86263	44713	9339	12127	n.d.	3873	10373	190200	3704

[mg/kg]	As	Ba	Cd	Co	Cr	Cu	Mn	Mo	Ni	Pb	Sr	V	Zn
Microwave digestion	n.d.	1113	4	18	231	1219	585	6	153	810	269	13	2154
RFA	5,7	1050	5,9	26	312	1109	684	n.d.	126	814	194	23	2179

A 3.2. Data of the batch experiments with municipal waste incineration ash

A 3.2.1. Electric conductivity, pH and E_H value and acid neutralisation capacity

Sample ID	el. cond. [mS/cm]	pH	E _H [mV]	ANC/BNC [meq/kg]
pH4-stat	10	4	538	1168
pH5-stat	8.1	5	487	824
pH6-stat	7.0	6	463	668
pH7-stat	6.3	7	425	555
pH11-stat	1.2	11	310	10
NH ₄ NO ₃	81	8.5	315	n.d.
SSE	36	11	202	n.d.
DEV S4	1.2	11.3	264	n.d.

A 3.2.2. Main elements

Sample ID	Al	St.Dev.	Ca	St.Dev.	Fe	St.Dev.	K	St.Dev.
	[mg/L]	[µg/L]	[mg/L]	[mg/L]	[mg/L]	[mg/L]	[mg/L]	[mg/L]
pH4-stat	59	5.0	2287	74	11	3.1	79	2.7
pH5-stat	1.1	0.19	1782	300	0.2	0.08	72	1.7
pH6-stat	<0.05		1529	141	<0.05		67	0.92
pH7-stat	<0.05		1335	160	<0.05		65	0.93
pH11-stat	18	1.2	80	5	<0.05		54	1.4
NH ₄ NO ₃	5	0.14	2465	14	<0.05		236	2.6
SSE	1.7	132	171	1.1	96		594	7.3
DEV S4	20	0.55	84	1.1	<0.05		39	0.91

Sample ID	Mg	St.Dev.	Na	St.Dev.	S	St.Dev.	Si	St.Dev.
	[mg/L]	[mg/L]	[mg/L]	[mg/L]	[mg/L]	[mg/L]	[mg/L]	[mg/L]
pH4-stat	73	3.7	145	4.2	233	4.7	84	1.39
pH5-stat	40	8.9	132	3.3	218	34	32	3.8
pH6-stat	20	1.7	127	0.71	220	22	14	1.83
pH7-stat	15	2.8	124	1.9	229	4.5	8.4	1.94
pH11-stat	<0.05		114	1.8	57	2.6	1.6	0.14
NH ₄ NO ₃	8.2	0.09	406	4.0	635	3.5	0.2	0.02
SSE	<0.05		2970	52	137	5.6	7.8	0.09
DEV S4	<0.05		99	2.1	42	0.82	2.4	0.03

A 3.2.3. Trace elements

Sample ID	As	St.Dev.	Ba	St.Dev.	Cd	St.Dev.	Co	St.Dev.	Cr	St.Dev.
	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]
pH4-stat	<10		173	7.6	98	12	51	3.7	70	2.9
pH5-stat	<10		157	3.9	40	11	23	6.7	23	3.2
pH6-stat	<10		175	20	14	1.2	12	0.63	38	2.9
pH7-stat	<10		189	12	10	1.3	10	0.74	58	4.0
pH11-stat	<10		23	3.4	<10		<5		35	4.4
NH ₄ NO ₃	<10		1333	29	189	48	9.1	0.01	152	2.5
SSE	<10		84	1.3	<10		<5		122	7.6
DEV S4	<10		26		<10		7.6	0.08	24	5.3

Sample ID	Cu	St.Dev.	Mn	St.Dev.	Mo	St.Dev.	Ni	St.Dev.	Pb	St.Dev.
	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]
pH4-stat	5746	698	6278	367	<5		355	54	1595	117
pH5-stat	657	122	3451	1023	8.2	0.58	103	50	103	17
pH6-stat	161	22	1401	64	41	3.7	28	3.2	<10	
pH7-stat	126	2.9	770	277	64	5.9	20	2.6	<10	
pH11-stat	143	1.5	6.9	0.06	56	1.4	<5		<10	
NH ₄ NO ₃	64927	127	120	13	153	1.8	130	101	<10	
SSE	1570	20	6.5		839	13	15	0.34	<10	
DEV S4	132	1.8	7.0	0.15	52	0.87	<5		<10	

Sample ID	Sr [µg/L]	St.Dev. [µg/L]	V [µg/L]	St.Dev. [µg/L]	Zn [µg/L]	St.Dev. [µg/L]
pH4-stat	3948	92	<5		49989	2388
pH5-stat	3094	405	<5		14429	4427
pH6-stat	2471	160	8.6	0.15	2445	669
pH7-stat	2170	195	10	0.29	949	402
pH11-stat	115	14	20	0.36	5.6	0.74
NH ₄ NO ₃	3435	45	17	0.80	70550	3076
SSE	452	12	15	1.3	18	3.0
DEV S4	104	3.0	17	0.18	29	

A 3.3. Data of the saturated column experiment with municipal waste incineration ash

A 3.3.1. Characteristic data of the column

Length	[cm]	48.3
Diameter	[cm]	5
Volume	[cm ³]	947.89
Amount material original	[kg]	1.783
Amount material dry	[kg]	1.719
Amount water in column	[L]	0.29
Vol. inlet/outlet	[L/d]	0.16
Specific weight	[g/cm ³]	2.61
Absolut porosity		0.31
Specific porosity		0.3
Watersaturation	[% abs. porosity]	99.5
kf-value	[m/s]	6.45 * 10 ⁻⁵
Vf (flow rate 0.1 mL/min)	[cm/d]	8
Va (flow rate 0.1 mL/min)	[cm/d]	26

A 3.3.2. Outlet

A 3.3.2.1. Liquid/solid ratio, electric conductivity, pH and E_H value, inorganic carbon, dissolved organic carbon and anions

Time [d]	L/S [L/kg]	el. cond. [mS/cm]	pH	E _H [mV]	IC [mg/L]	DOC [mg/L]	Cl [mg/L]	NO ₃ [mg/L]	SO ₄ [mg/L]
0.27	0.02	26	11.0	258	n.d.	472	11345	n.d.	929
0.43	0.04	n.d.	n.d.	n.d.	n.d.	374	8711	n.d.	920
0.58	0.05	n.d.	n.d.	n.d.	n.d.	370	8597	n.d.	855
0.77	0.06	26	11.2	236	n.d.	369	8889	n.d.	857
0.89	0.07	20	11.1	241	n.d.	380	9559	n.d.	864
0.93	0.08	n.d.	n.d.	n.d.	n.d.	n.d.	11800	n.d.	712
1.08	0.09	n.d.	n.d.	n.d.	n.d.	n.d.	10733	n.d.	723
1.23	0.10	n.d.	n.d.	n.d.	n.d.	472	9559	n.d.	765
1.37	0.11	n.d.	n.d.	n.d.	n.d.	378	10472	n.d.	892
1.52	0.12	n.d.	n.d.	n.d.	n.d.	361	n.d.	n.d.	n.d.
1.66	0.13	n.d.	n.d.	n.d.	n.d.	333	3821	n.d.	1185
1.94	0.16	48	11.6	211	n.d.	296	1273	n.d.	1402
2.20	0.18	n.d.	n.d.	n.d.	n.d.	263	622	7.3	1397

Time [d]	L/S [L/kg]	el. cond. [mS/cm]	pH	E _H [mV]	IC [mg/L]	DOC [mg/L]	Cl [mg/L]	NO ₃ [mg/L]	SO ₄ [mg/L]
2.35	0.19	n.d.	n.d.	n.d.	n.d.	220	492	6.2	1323
2.50	0.20	n.d.	n.d.	n.d.	n.d.	211	399	4.9	1189
2.64	0.21	n.d.	n.d.	n.d.	n.d.	203	336	10.2	1102
2.79	0.23	n.d.	n.d.	n.d.	n.d.	159	292	6.1	976
2.93	0.24	38	11.8	204	n.d.	n.d.	259	5.3	921
5.95	0.48	32	11.9	219	n.d.	n.d.	n.d.	n.d.	n.d.
6.35	0.51	n.d.	n.d.	n.d.	n.d.	40	n.d.	n.d.	n.d.
6.93	0.56	32	12.0	222	n.d.	32	46	3.1	266
7.35	0.59	n.d.	n.d.	n.d.	n.d.	33	43	2.4	241
7.93	0.64	30	11.9	239	n.d.	28	40	2.8	203
9.93	0.80	25	11.8	254	n.d.	20	33	2.3	147
12.93	1.04	21	11.8	240	n.d.	18	32	3.0	106
13.93	1.13	19	11.9		n.d.	14	24	3.1	94
14.93	1.21	18	11.9	263	n.d.	13	18	3.4	83
15.93	1.29	17	11.8	235	n.d.	12	16	2.9	76
18.93	1.53	14	11.7	251	n.d.	8.8	13	3.3	66
19.93	1.61	13	11.7	251	n.d.	7.0	12	2.6	61
20.93	1.69	13	11.6	280	n.d.	8.3	10	3.3	63
23.35	1.72	n.d.	n.d.	n.d.	n.d.	9.8	21	3.1	73
23.93	1.77	n.d.	n.d.	n.d.	n.d.	10.1	20	2.7	77
24.35	1.81	n.d.	n.d.	n.d.	n.d.	9.3	19	2.4	72
24.93	1.85	n.d.	n.d.	n.d.	n.d.	10.6	11	2.6	71
25.35	1.89	n.d.	n.d.	n.d.	n.d.	8.5	11	2.5	66
25.93	1.93	12	11.6	275	n.d.	9.2	8.8	3.3	63
26.93	2.01	11	11.6	255	n.d.	7.8	8.2	3.1	61
27.93	2.09	11	11.6	260	n.d.	7.2	7.9	2.8	58
28.93	2.18	11	11.6	261	n.d.	8.0	7.6	3.0	61
29.93	2.26	10	11.6	281	n.d.	7.9	6.6	2.2	55
32.93	2.50	9.7	11.5	288	n.d.	8.6	6.9	3.0	59
33.93	2.58	12	11.4	299	28	9.7	6.6	3.0	58
34.93	2.66	9.1	11.4	260	24	9.3	6.5	3.2	59
35.93	2.74	9.2	11.5	280	26	9.5	5.7	2.6	58
36.93	2.82	9.3	11.5	259	26	6.8	6.5	2.8	62
39.93	3.06	8.9	11.4	223	9.5	6.8	5.4	3.0	57
40.93	3.14	8.8	11.4	311	16	7.5	4.9	3.2	57
41.93	3.23	8.6	11.4	301	10	8.2	5.0	2.7	57
42.93	3.31	8.4	11.4	296	103	5.9	4.8	2.3	56
43.93	3.39	8.3	11.4	163	171	9.3	4.8	3.1	56
46.93	3.63	8.2	11.4	304	113	6.5	4.6	3.1	66
47.93	3.71	8.1	11.4	281	2.1	6.6	4.8	2.8	58
48.93	3.79	7.9	11.4	256	33	32	4.6	3.3	56
49.79	3.86	8.0	11.5	240	34	15	4.0	2.8	54
53.93	4.19	7.7	11.4	273	29	12	3.5	3.0	52
54.93	4.28	7.7	11.4	288	34	22	3.2	2.4	51
55.93	4.36	7.6	11.4	298	30	11	3.4	3.0	54
56.93	4.44	7.6	11.4	293	34	38	3.7	2.7	52
57.93	4.52	7.6	11.4	347	31	28	3.8	2.5	51
61.93	4.84	7.3	11.4	319	28	11	3.5	3.2	55
62.93	4.92	7.5	11.4	311	32	10	3.5	3.0	56
63.93	5.00	7.4	11.4	311	34	9.0	3.2	3.7	54
64.93	5.08	7.4	11.4	317	32	16	3.4	3.1	53
67.93	5.33	7.4	11.3	305	31	11	3.0	3.1	51
68.93	5.41	7.2	11.4	232	31	16	3.1	3.5	53
70.00	5.49	7.2	11.4	256	31	24	3.0	3.0	50
70.93	5.57	7.2	11.4	231	32	11	1.3	1.5	26
71.79	5.64	7.2	11.4	282	33	12	3.5	3.4	53
74.35	5.84	7.1	11.4	273	29	11	3.3	3.6	57

Time [d]	L/S [L/kg]	el. cond. [mS/cm]	pH	E _H [mV]	IC [mg/L]	DOC [mg/L]	Cl [mg/L]	NO ₃ [mg/L]	SO ₄ [mg/L]
75.35	5.92	n.d.	n.d.	n.d.	29	12	3.0	3.5	55
75.93	5.97	7.1	11.4	281	31	13	3.0	3.3	57
76.93	6.05	7.1	11.4	274	29	13	2.8	3.2	54
77.93	6.13	6.9	11.1	257	30	8.4	3.0	3.5	54
78.93	6.21	7.0	11.4	289	16	25	3.1	3.4	53
81.33	6.30	n.d.	n.d.	n.d.	10	17	4.3	3.6	61
81.48	6.31	n.d.	n.d.	n.d.	15	15	4.6	3.5	59
81.62	6.32	n.d.	n.d.	n.d.	18	16	4.0	3.5	54
81.77	6.33	n.d.	n.d.	n.d.	15	12	4.1	3.0	55
81.91	6.34	7.0	11.4	274	16	10	4.4	3.2	53
83.64	6.48	n.d.	n.d.	n.d.	11	11	3.9	3.7	55
84.64	6.56	n.d.	n.d.	n.d.	19	10	3.9	3.0	55
85.64	6.64	n.d.	n.d.	n.d.	29	13	4.2	3.2	53
88.93	6.91	6.9	11.4	240	11	15	3.0	2.9	50
89.93	6.99	6.9	11.4	268	31	14	2.8	3.1	50
91.00	7.08	6.9	11.4	281	32	13	2.5	3.6	51
91.93	7.15	6.8	11.4	259	30	22	3.3	4.2	52
92.95	7.24	6.8	11.4	260	32	14	2.7	4.1	49
95.93	7.48	6.8	11.4	281	32	14	2.9	3.6	50
96.93	7.56	6.7	11.4	n.d.	32	12	2.9	3.7	51
99.95	7.80	6.6	11.3	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
102.35	7.83	n.d.	n.d.	n.d.	23	15	5.7	4.0	65
102.93	7.88	n.d.	n.d.	n.d.	25	16	5.9	3.8	57
103.93	7.96	6.9	11.3	n.d.	32	n.d.	5.7	3.0	56
105.00	8.05	6.8	11.4	n.d.	32	15	4.0	2.8	55
105.93	8.12	6.8	11.4	n.d.	32	29	2.9	2.9	56
106.95	8.20	6.7	11.4	n.d.	32	13	2.3	2.9	52
109.93	8.45	6.5	11.3	n.d.	32	15	2.0	3.5	47
111.93	8.61	6.6	11.4	273	31	15	2.2	2.9	45
112.93	8.69	6.5	11.4	282	31	15	2.9	2.9	46
113.95	8.77	6.5	11.2	264	30	12	2.1	2.8	46
116.93	9.01	6.6	11.3	284	32	15	2.1	2.9	44
117.93	9.09	6.5	11.3	264	18	14	2.8	2.7	48
119.93	9.25	6.5	11.4	268	31	16	3.2	3.7	44
123.93	9.58	6.5	11.3	220	28	20	2.3	3.3	48
124.93	9.66	6.4	11.3	266	12	15	2.2	3.1	45
125.93	9.74	6.3	11.3	269	11	12	2.2	2.9	46
126.93	9.82	6.2	11.3	254	28	26	2.1	2.6	46
127.93	9.90	6.4	11.4	249	29	21	2.0	2.7	44
130.93	10.14	6.3	11.3	243	11	20	n.d.	n.d.	n.d.
131.93	10.22	6.3	11.3	271	10	17	2.2	3.2	47
132.93	10.30	6.3	11.4	270	27	16	2.5	3.0	49
133.93	10.38	6.3	11.2	270	20	17	2.6	3.5	49
134.93	10.46	6.2	11.2	274	28	13	2.4	3.2	46
137.93	10.71	n.d.	n.d.	n.d.	11	20	2.8	3.5	47
138.93	10.79	6.2	11.2	284	13	17	3.1	3.4	48
139.93	10.87	n.d.	n.d.	n.d.	10	13	2.1	3.6	49
144.93	11.27	6.2	11.3	281	32	21	2.5	3.2	48
146.93	11.43	6.0	11.2	275	33	14	2.5	3.5	49
151.93	11.84	6.1	11.3	277	19	16	2.3	2.8	48
154.93	12.08	6.0	11.3	295	8.9	15	7.9	3.5	47
155.93	12.16	6.0	11.3	280	31	14	3.4	3.3	51
158.93	12.40	6.1	11.3	301	31	26	2.6	3.7	49
160.93	12.56	5.9	11.2	282	n.d.	26	4.7	3.5	48
162.93	12.73	5.9	11.3	271	n.d.	25	2.3	3.6	49
165.93	12.97	5.9	11.3	292	n.d.	20	2.4	3.3	49
167.93	13.13	5.9	11.3	284	n.d.	20	3.0	4.0	50

Time [d]	L/S [L/kg]	el. cond. [mS/cm]	pH	E _H [mV]	IC [mg/L]	DOC [mg/L]	Cl [mg/L]	NO ₃ [mg/L]	SO ₄ [mg/L]
169.93	13.29	5.9	11.3	337	n.d.	n.d.	n.d.	n.d.	n.d.
175.93	13.78	5.9	11.3	227	n.d.	n.d.	n.d.	n.d.	n.d.
181.93	14.26	5.7	11.3	230	32	14	n.d.	n.d.	n.d.
188.93	14.83	5.9	11.1	228	32	24	3.2	2.2	40
195.93	15.39	5.8	11.1	243	32	19	3.1	2.0	40
202.93	15.96	5.6	11.0	239	n.d.	n.d.	3.0	1.7	41
209.93	16.52	5.6	11.1	239	19	11	2.8	2.1	38
216.93	17.09	5.4	11.2	272	32	24	3.0	2.8	40
223.93	17.65	5.4	11.3	234	31	16	2.7	1.9	39
230.93	18.22	5.6	11.3	n.d.	32	17	2.9	1.7	39
237.93	18.78	5.4	11.3	235	30	12	3.0	1.1	39
244.93	19.35	5.4	11.3	247	8.2	11	3.2	2.4	39
251.93	19.91	5.3	11.3	233	6.3	19	2.9	1.4	40
279.93	20.08	6.0	10.9	242	6.5	19	7.3	1.7	56
280.10	20.09	n.d.	n.d.	n.d.	6.7	13	8.0	1.9	57
280.25	20.10	n.d.	n.d.	n.d.	5.9	11	8.3	1.9	55
280.39	20.11	n.d.	n.d.	n.d.	6.1	11	8.6	1.7	55
280.54	20.12	n.d.	n.d.	n.d.	6.1	11	9.1	2.0	54
280.68	20.14	5.9	10.9	221	6.1	9.5	10	2.2	53
281.93	20.24	5.7	10.8	229	6.1	9.7	3.9	1.6	52
286.93	20.64	5.4	11.1	252	4.5	11	2.6	1.3	34
293.93	21.21	5.4	11.2	264	1.5	11	3.5	1.3	42
300.93	21.77	5.2	11.2	270	n.d.	n.d.	2.9	1.5	42

A 3.3.2.2. Main elements

Time [d]	Al [mg/L]	Ca [mg/L]	Fe [µg/L]	K [mg/L]	Mg [µg/L]	Na [mg/L]	Si [mg/L]
0.27	4.7	358	15	1370	<8	6456	7.4
0.43	5.8	299	18	1106	<8	5139	7.6
0.58	5.7	308	14	1083	<8	5054	7.5
0.77	5.7	317	13	1071	<8	5030	7.5
0.89	5.7	341	19	1111	<8	5166	7.7
0.93	4.4	416	14	1279	<8	5971	5.4
1.08	4.5	417	13	1284	<8	6016	5.7
1.23	6.1	407	14	1206	<8	5640	7.8
1.37	6.8	315	15	1028	<8	4772	8.5
1.52	7.9	222	15	792	<8	3667	9.5
1.66	9.9	129	13	621	<8	2845	11
1.94	15	54	<10	355	<8	1666	13
2.20	19	54	<10	286	<8	1291	15
2.35	21	42	<10	260	<8	1144	15
2.50	22	33	<10	233	<8	1011	15
2.64	23	26	<10	217	<8	927	15
2.79	24	23	<10	197	<8	850	15
2.93	26	21	20	188	<8	803	15
5.95	36	13	68	127	31	466	16
6.35	37	12	24	127	<8	457	16
6.93	35	10	<10	119	<8	417	16
7.35	35	n.d.	<10	120	<8	409	17
7.93	31	10	<10	112	<8	367	16
9.93	25	11	<10	103	<8	281	15
12.93	19	13	33	102	<8	213	15
13.93	17	13	<10	99	<8	178	14
14.93	15	15	<10	99	<8	148	14

Time [d]	Al [mg/L]	Ca [mg/L]	Fe [µg/L]	K [mg/L]	Mg [µg/L]	Na [mg/L]	Si [mg/L]
15.93	13	16	13	101	<8	126	13
18.93	9.8	21	<10	108	<8	79	12
19.93	9.1	23	<10	110	<8	69	12
20.93	8.5	24	<10	110	<8	60	11
23.35	9.1	49	15	130	<8	73	13
23.93	8.5	50	24	125	<8	65	13
24.35	8.2	51	<10	121	<8	61	12
24.93	7.9	48	26	116	<8	55	12
25.35	7.6	42	<10	112	<8	52	12
25.93	7.1	30	<10	107	<8	48	11
26.93	6.7	31	<10	102	<8	43	11
27.93	6.5	33	<10	98	<8	40	11
28.93	5.7	34	<10	91	<8	35	11
29.93	5.5	35	<10	87	<8	33	11
32.93	5.0	39	<10	77	<8	29	11
33.93	4.8	42	<10	74	<8	28	11
34.93	4.7	43	<10	70	<8	27	11
35.93	4.5	43	<10	67	<8	26	11
36.93	4.5	45	12	65	45	26	11
39.93	4.5	46	68	59	36	23	11
40.93	4.1	46	<10	57	9.5	23	11
41.93	3.9	46	<10	54	9.1	22	11
42.93	3.9	47	<10	52	9.4	22	11
43.93	3.6	46	<10	49	9.2	21	11
46.93	3.6	54	<10	52	13	23	12
47.93	3.5	50	<10	43	<8	19	12
48.93	3.4	50	<10	42	11	19	12
49.79	3.3	49	<10	40	12	18	11
53.93	3.2	55	109	37	21	18	12
54.93	3.1	52	<10	35	<8	17	12
55.93	3.0	91	<10	34	12	17	12
56.93	3.0	56	<10	33	12	17	12
57.93	3.0	55	<10	33	12	17	12
61.93	2.9	64	<10	31	12	16	13
62.93	2.7	55	<10	29	15	16	13
63.93	2.7	55	<10	28	12	16	13
64.93	2.7	54	<10	28	13	16	13
67.93	2.6	54	<10	26	14	15	13
68.93	2.5	56	11	25	18	15	13
70.00	2.4	55	<10	24	<8	14	13
70.93	2.5	56	<10	24	15	15	13
71.79	2.5	57	<10	24	14	14	13
74.35	2.4	56	<10	23	20	14	14
75.35	2.3	59	<10	22	17	14	13
75.93	2.3	57	<10	22	15	14	14
76.93	2.3	58	12	21	23	14	14
77.93	2.2	57	<10	21	16	14	14
78.93	2.1	55	<10	20	17	13	13
81.33	2.2	73	33	22	35	15	14
81.48	2.2	63	<10	21	15	15	14
81.62	2.2	58	<10	21	16	15	14
81.77	2.2	58	<10	21	16	15	14
81.91	2.2	58	<10	21	17	15	14
83.64	2.1	58	<10	20	16	15	14
84.64	2.1	58	<10	21	16	15	14
85.64	2.1	57	<10	21	14	16	14
88.93	2.0	55	<10	19	15	14	14

Time [d]	Al [mg/L]	Ca [mg/L]	Fe [µg/L]	K [mg/L]	Mg [µg/L]	Na [mg/L]	Si [mg/L]
89.93	2.0	57	3.5	19	11	15	18
91.00	2.0	56	1.2	19	9.6	14	18
91.93	2.0	56	1.6	18	9.5	14	18
92.95	2.0	56	1.8	18	9.9	15	18
95.93	1.9	57	1.0	17	9.3	14	18
96.93	1.9	56	1.4	17	9.6	14	18
99.95	1.9	58	<1	18	<8	15	19
102.35	2.4	64	46	22	48	19	20
102.93	1.9	63	2.3	19	11	18	19
103.93	1.8	57	1.5	19	9.6	18	19
105.00	1.8	57	1.6	18	9.7	17	20
105.93	1.8	56	1.5	18	9.8	17	20
106.95	1.8	56	1.4	18	10	16	20
109.93	1.7	55	4.9	17	11	15	21
111.93	1.7	55	1.2	16	10	15	21
112.93	1.7	56	1.1	16	9.8	15	21
113.95	1.7	56	1.7	16	11	14	21
116.93	1.6	57	9.1	15	11	14	22
117.93	1.6	70	1.4	15	10	14	22
119.93	1.6	57	1.2	15	11	13	22
123.93	1.5	57	<1	14	10	12	22
124.93	1.5	57	<1	14	11	12	22
125.93	1.5	58	10	14	10	12	23
126.93	1.5	58	2.1	14	11	12	23
127.93	1.4	58	1.5	14	10	12	23
130.93	1.4	58	1.5	13	11	12	23
131.93	1.4	58	<1	13	<8	11	23
132.93	1.4	59	1.7	13	11	12	23
133.93	1.4	58	1.2	13	11	11	23
134.93	1.3	58	1.1	13	11	11	23
137.93	1.3	59	2.2	12	11	11	24
138.93	1.3	59	27	12	11	11	24
139.93	1.3	60	1.6	13	11	11	23
140.93	1.3	62	1.1	13	8.6	11	21
144.93	1.3	63	12	12	9.1	11	22
145.93	1.2	62	1.3	12	9.1	11	22
146.93	1.2	62	2.7	12	9.3	11	22
147.93	1.2	63	1.4	12	9.3	11	22
151.93	1.2	63	1.3	12	9.3	11	22
153.93	1.2	63	1.3	11	9.5	11	22
154.93	1.2	62	1.7	11	9.4	11	22
155.93	1.1	62	1.7	11	9.4	10	22
157.93	1.2	65	1.5	11	9.5	11	23
158.93	1.2	66	1.7	12	9.8	11	23
159.93	1.2	65	1.1	11	9.7	10	23
160.93	1.2	64	1.2	11	9.6	11	23
162.93	1.2	64	1.1	11	9.6	10	23
165.93	1.1	64	<1	11	9.6	10	23
167.93	1.1	65	<1	11	9.5	10	23
169.93	1.4	64	146	11	74	10	25
181.93	1.0	61	<1	9.9	10	9.7	23
188.93	1.0	n.d.	<1	n.d.	11	0.1	n.d.
195.93	1.0	64	1.0	9.2	10	8.7	24
202.93	1.0	n.d.	<1	n.d.	11	n.d.	n.d.
209.93	1.0	63	1.6	8.2	10	8.1	23
216.93	1.0	63	1.2	7.9	11	8.1	23
223.93	1.0	62	1.0	7.3	11	8.0	23

Time [d]	Al [mg/L]	Ca [mg/L]	Fe [µg/L]	K [mg/L]	Mg [µg/L]	Na [mg/L]	Si [mg/L]
230.93	1.0	63	<1	7.0	11	7.8	23
237.93	1.0	63	<1	6.6	11	7.4	23
244.93	1.0	63	1.1	6.3	12	7.1	22
251.93	1.0	63	<1	6.3	12	6.9	22
279.93	0.8	66	1.2	9.2	11	15	24
280.10	0.9	71	1.3	9.9	12	16	26
280.25	0.9	69	1.0	9.7	12	16	26
280.39	0.9	69	<1	9.5	10	16	25
280.54	0.8	68	<1	9.4	11	15	25
280.68	0.8	67	1.0	9.1	10	15	24
281.93	0.9	63	<1	8.8	11	14	25
286.93	0.9	57	<1	7.5	9.8	11	25
293.93	0.9	60	4.0	7.1	11	9	24
300.93	0.8	n.d.	n.d.	n.d.	<8	n.d.	n.d.

A 3.3.2.3. Trace elements

Time [d]	Ba [µg/L]	Cd [µg/L]	Co [µg/L]	Cr [µg/L]	Cu [µg/L]	Mn [µg/L]	Mo [µg/L]	Ni [µg/L]	Sr [µg/L]	V [µg/L]	Zn [µg/L]
0.27	184	<10	<10	221	3066	<10	1931	25	1289	16	572
0.43	158	<10	<10	207	2729	<10	1654	24	1065	18	228
0.58	164	<10	<10	195	2682	<10	1619	24	1145	17	436
0.77	167	<10	<10	189	2676	<10	1607	21	1190	16	33
0.89	184	<10	<10	185	2680	<10	1648	22	1288	16	38
0.93	211	<10	<10	189	2768	<10	1667	23	1584	17	51
1.08	211	<10	<10	183	2794	<10	1629	23	1585	16	32
1.23	213	<10	<10	177	2960	<10	1656	22	1473	16	32
1.37	166	<10	<10	183	3298	<10	1910	28	1160	19	27
1.52	117	<10	<10	207	3905	<10	2450	26	790	22	28
1.66	70	<10	<10	294	4990	<10	3027	34	469	32	46
1.94	31	<10	<10	421	5361	<10	2315	34	202	59	34
2.20	31	<10	<10	432	3988	<10	1166	28	180	80	100
2.35	28	<10	<10	421	3340	<10	863	24	152	83	70
2.50	25	<10	<10	403	2675	<10	711	24	116	86	257
2.64	24	<10	<10	389	2332	<10	629	20	103	88	43
2.79	23	<10	<10	374	2038	<10	559	18	93	88	45
2.93	23	<10	<10	366	1762	<10	521	18	89	89	52
5.95	32	<10	<10	153	555	<10	194	10	64	89	86
6.35	29	<10	<10	147	514	<10	182	17	65	90	225
6.93	25	<10	<10	118	418	<10	148	<10	59	86	69
7.35	13	<10	<10	110	392	<10	142	<10	28	88	<5
7.93	25	<10	<10	93	332	<10	121	<10	59	81	50
9.93	29	<10	<10	65	224	<10	85	<10	63	66	36
12.93	32	<10	<10	43	196	<10	82	<10	73	54	41
13.93	29	<10	<10	36	151	<10	60	<10	74	46	30
14.93	35	<10	<10	31	126	<10	46	<10	83	40	22
15.93	39	<10	<10	30	114	<10	39	<10	92	36	23
18.93	47	<10	<10	27	92	<10	32	<10	115	28	21
19.93	50	<10	<10	27	89	<10	32	<10	125	27	51
20.93	50	<10	<10	25	85	<10	29	<10	133	26	48
23.35	88	<10	<10	30	134	<10	55	<10	228	30	34
23.93	91	<10	<10	30	124	<10	48	<10	231	28	43
24.35	90	<10	<10	28	115	<10	48	<10	239	27	33
24.93	82	<10	<10	29	104	<10	41	<10	210	26	29
25.35	74	<10	<10	30	95	<10	34	<10	194	25	25

Time [d]	Ba [µg/L]	Cd [µg/L]	Co [µg/L]	Cr [µg/L]	Cu [µg/L]	Mn [µg/L]	Mo [µg/L]	Ni [µg/L]	Sr [µg/L]	V [µg/L]	Zn [µg/L]
25.93	56	<10	<10	28	89	<10	23	<10	157	24	157
26.93	56	<10	<10	23	82	<10	20	<10	158	24	24
27.93	58	<10	<10	22	79	<10	18	<10	167	23	16
28.93	57	<10	<10	18	81	<10	17	<10	167	26	12
29.93	58	<10	<10	18	77	<10	16	<10	176	26	14
32.93	59	<10	<10	17	77	<10	15	<10	186	26	49
33.93	61	<10	<10	17	77	<10	15	<10	194	26	19
34.93	61	<10	<10	17	75	<10	15	<10	191	26	17
35.93	60	<10	<10	18	73	<10	14	<10	198	26	13
36.93	63	<10	<10	17	78	<10	14	<10	206	26	39
39.93	72	<10	<10	18	80	<10	12	<10	200	26	28
40.93	58	<10	<10	16	71	<10	12	<10	209	27	11
41.93	58	<10	<10	16	70	<10	12	<10	204	26	12
42.93	59	<10	<10	17	70	<10	12	<10	208	27	9
43.93	56	<10	<10	14	65	<10	11	<10	204	26	10
46.93	62	<10	<10	16	72	<10	12	<10	228	31	11
47.93	57	<10	<10	14	64	<10	<8	<10	216	28	<5
48.93	57	<10	<10	15	63	<10	9.9	<10	216	29	<5
49.79	56	<10	<10	13	61	<10	<8	<10	212	28	<5
53.93	60	<10	<10	14	63	<10	8.9	<10	225	31	13
54.93	56	<10	<10	15	60	<10	8.6	<10	224	31	<5
55.93	78	<10	<10	14	65	<10	8.8	<10	277	33	22
56.93	57	<10	<10	14	61	<10	9.2	<10	227	32	6.1
57.93	57	<10	<10	15	63	<10	9.5	<10	228	33	5.7
61.93	61	<10	<10	15	60	<10	9.2	<10	244	36	32
62.93	55	<10	<10	13	57	<10	8.7	<10	231	34	<5
63.93	54	<10	<10	14	57	<10	8.5	<10	226	35	<5
64.93	54	<10	<10	13	57	<10	8.5	<10	231	36	<5
67.93	52	<10	<10	15	56	<10	7.8	<10	231	38	<5
68.93	54	<10	<10	14	55	<10	7.8	<10	229	38	6.0
70.00	52	<10	<10	15	53	<10	7.3	<10	220	38	5.2
70.93	53	<10	<10	13	54	<10	8.4	<10	231	42	6.3
71.79	53	<10	<10	13	54	<10	7.9	<10	235	42	<5
74.35	52	<10	<10	13	55	<10	8.0	<10	230	45	6.9
75.35	53	<10	<10	14	53	<10	8.1	<10	236	43	<5
75.93	51	<10	<10	13	56	<10	7.6	<10	232	44	<5
76.93	53	<10	<10	12	56	<10	7.4	<10	237	44	8.2
77.93	52	<10	<10	13	55	<10	7.9	<10	239	45	<5
78.93	50	<10	<10	12	54	<10	7.5	<10	228	44	<5
81.33	62	<10	<10	14	64	<10	9.7	<10	262	28	31
81.48	55	<10	<10	15	62	<10	10	<10	251	47	<5
81.62	53	<10	<10	14	62	<10	11	<10	241	47	<5
81.77	52	<10	<10	14	61	<10	10	<10	243	47	<5
81.91	52	<10	<10	15	61	<10	10	<10	238	47	<5
83.64	52	<10	<10	16	60	<10	11	<10	241	48	<5
84.64	53	<10	<10	15	64	<10	11	<10	243	28	<5
85.64	52	<10	<10	15	64	<10	11	<10	236	28	<5
88.93	49	<10	<10	13	55	<10	7.1	<10	228	28	<5
89.93	49	<2	<2	14	54	<2	9.7	<2	228	28	6.8
91.00	49	<2	<2	14	53	<2	9.3	<2	227	28	7.1
91.93	49	<2	<2	13	49	<2	8.8	<2	230	28	4.9
92.95	49	<2	<2	14	53	<2	9.4	<2	221	28	4.3
95.93	48	<2	<2	14	53	<2	10	<2	223	28	4.0
96.93	48	<2	<2	15	53	<2	10	<2	223	28	4.8
102.35	76	<2	<2	17	90	2.6	16	<2	227	30	17
102.93	60	<2	<2	17	85	<2	17	<2	240	30	4.5
103.93	50	<2	<2	17	77	<2	19	<2	224	30	3.7

Time [d]	Ba [µg/L]	Cd [µg/L]	Co [µg/L]	Cr [µg/L]	Cu [µg/L]	Mn [µg/L]	Mo [µg/L]	Ni [µg/L]	Sr [µg/L]	V [µg/L]	Zn [µg/L]
105.00	50	<2	<2	17	66	<2	16	<2	225	29	3.1
105.93	48	<2	<2	15	59	<2	9.9	<2	223	30	3.9
106.95	48	<2	<2	13	56	<2	8.4	<2	221	31	3.0
109.93	47	<1	<1	14	53	<1	8.5	<1	224	30	3.2
111.93	47	<1	<1	13	51	<1	8.4	<1	221	30	3.1
112.93	47	<1	<1	13	50	<1	8.2	<1	223	30	2.6
113.95	47	<1	<1	13	50	<1	8.3	1.4	221	30	4.0
116.93	47	<1	<1	13	47	<1	8.0	1.5	224	29	2.9
117.93	54	<1	<1	14	49	<1	8.3	<1	248	31	3.6
119.93	47	<1	<1	14	46	<1	8.0	<1	227	30	3.0
123.93	53	<1	<1	13	44	<1	7.9	<1	225	28	2.4
124.93	46	<1	<1	13	43	<1	7.7	<1	224	29	3.0
125.93	47	25	<1	13	42	<1	7.5	<1	225	28	3.8
126.93	48	3.7	<1	13	43	<1	7.7	<1	223	28	2.6
127.93	47	<1	<1	13	42	<1	7.7	<1	224	28	2.3
130.93	46	<1	<1	14	43	<1	8.1	<1	237	29	2.6
131.93	46	<1	<1	n.d.	n.d.	<1	n.d.	<1	235	29	<1
132.93	46	<1	<1	13	43	<1	8.1	<1	229	28	2.4
133.93	46	<1	<1	14	42	<1	7.7	<1	232	28	2.6
134.93	46	<1	<1	13	40	<1	7.4	<1	228	28	2.5
137.93	46	<1	<1	13	40	<1	7.3	<1	230	27	2.4
138.93	45	2.5	<1	13	39	<1	7.2	<1	231	27	2.8
139.93	46	<1	<1	13	40	<1	7.1	<1	227	26	7.3
140.93	41	<1	<1	11	34	<1	5.7	<1	210	24	<1
144.93	40	23	<1	12	32	<1	5.4	<1	208	23	1.6
145.93	40	1.1	<1	12	32	<1	5.5	<1	207	22	1.4
146.93	41	4.5	<1	12	32	<1	5.4	<1	209	22	1.1
147.93	41	<1	<1	11	32	<1	5.3	<1	208	22	1.6
151.93	40	1.0	<1	11	31	<1	5.5	<1	209	22	1.1
153.93	41	<1	<1	11	31	<1	5.5	<1	209	22	1.3
154.93	40	1.6	<1	11	30	<1	5.3	<1	208	21	1.2
155.93	40	1.6	<1	11	30	<1	5.4	<1	208	21	1.4
157.93	43	1.2	<1	12	30	<1	5.8	<1	218	23	1.0
158.93	41	1.6	<1	12	30	<1	5.6	<1	211	22	1.4
159.93	42	<1	<1	11	31	<1	5.6	<1	216	22	2.0
160.93	42	<1	<1	12	29	<1	5.5	<1	213	22	1.1
162.93	41	<1	<1	12	29	<1	5.4	<1	211	21	1.0
165.93	40	<1	<1	11	28	<1	5.5	<1	208	21	2.3
167.93	40	<1	<1	12	28	<1	5.5	<1	209	21	1.2
169.93	52	5.0	<1	13	41	6.8	5.6	<1	207	21	33
181.93	39	<1	<1	11	24	<1	4.8	<1	204	20	1.5
188.93	39	<1	<1	11	22	<1	4.8	<1	205	20	1.5
195.93	39	<1	<1	11	21	<1	5.0	<1	203	19	1.7
202.93	39	<1	<1	11	20	4.9	0.0	<1	207	18	1.7
209.93	37	<1	<1	12	20	<1	6.3	<1	206	18	3.0
216.93	37	<1	<1	11	19	<1	6.0	1.1	206	17	3.0
223.93	36	<1	<1	11	18	<1	6.0	<1	203	16	2.9
230.93	37	<1	<1	11	17	<1	5.5	<1	205	16	3.1
237.93	36	<1	<1	11	16	<1	5.6	1.5	200	15	3.2
244.93	37	1.8	<1	11	16	<1	5.7	<1	201	15	2.6
251.93	38	<1	<1	10	15	<1	6.0	<1	204	15	2.6
279.93	44	<1	<1	6.3	56	<1	22	1.1	218	19	78
280.10	47	<1	<1	3.4	60	<1	26	<1	228	20	42
280.25	45	<1	<1	2.3	57	<1	27	1.2	224	20	32
280.39	45	<1	<1	1.5	57	<1	28	<1	223	19	26
280.54	44	<1	<1	1.1	58	<1	31	<1	218	19	23
280.68	43	<1	<1	1.0	56	<1	37	<1	214	18	22

Time [d]	Ba [µg/L]	Cd [µg/L]	Co [µg/L]	Cr [µg/L]	Cu [µg/L]	Mn [µg/L]	Mo [µg/L]	Ni [µg/L]	Sr [µg/L]	V [µg/L]	Zn [µg/L]
281.93	41	<1	<1	1.1	30	<1	10	<1	203	18	16
286.93	36	<1	<1	8.8	18	<1	4.5	<1	183	18	4.7
293.93	37	<1	<1	7.2	15	<1	6.0	<1	193	17	4.1
300.93	43	<1	<1	3.6	15	<1	8.0	<1	185	17	3.7

A 3.3.3. Flow-path sampling

A 3.3.3.1. Liquid/solid ratio, electric conductivity, pH and E_H value, inorganic carbon, dissolved organic carbon and anions

1. Flow-path sampling after 83 days

Flow-path [cm]	el. cond. [mS/cm]	pH	E_H [mV]	IC [mg/L]	DOC [mg/L]	Cl [mg/L]	NO ₃ [mg/L]	SO ₄ [mg/L]
1	2.5	10.6	293	13	8.7	n.d.	n.d.	n.d.
2	3.1	10.9	209	17	5.0	n.d.	n.d.	n.d.
3	3.3	10.9	313	18	12	n.d.	n.d.	n.d.
4	4.2	10.9	320	20	12	n.d.	n.d.	n.d.
5	4.2	11.1	300	20	4.0	n.d.	n.d.	n.d.
6	4.3	11.1	288	6.6	6.8	n.d.	n.d.	n.d.
7	4.4	11.1	272	18	11	n.d.	n.d.	n.d.
8	7.8	11.2	273	26	8.2	n.d.	n.d.	n.d.
10	5.3	11.5	173	7.9	10	n.d.	n.d.	n.d.
16	5.9	11.3	220	2.4	12	n.d.	n.d.	n.d.
22	6.4	11.3	216	26	11	n.d.	n.d.	n.d.
28	6.7	11.3	221	28	18	n.d.	n.d.	n.d.
34	6.6	11.3	237	25	16	n.d.	n.d.	n.d.
40	6.6	11.3	224	28	17	n.d.	n.d.	n.d.
48	7.0	11.3	140	5.6	14	n.d.	n.d.	n.d.
50	6.9	11.4	240	n.d.	n.d.	n.d.	n.d.	n.d.

2. Flow-path sampling after 276 days

Flow-path [cm]	el. cond. [mS/cm]	pH	E _H [mV]	IC [mg/L]	DOC [mg/L]	Cl [mg/L]	NO ₃ [mg/L]	SO ₄ [mg/L]
1	1.0	9.8	379	7.8	35	1.8	2.2	14
2	1.4	10.2	352	9.6	18	1.8	2.0	19
3	1.5	10.4	328	11	15	1.9	2.0	19
4	1.8	10.4	357	10	67	2.9	2.1	18
5	1.9	10.5	334	12	17	1.8	1.9	24
6	2.2	10.5	300	13	16	1.9	2.4	27
7	2.3	10.5	337	15	23	2.2	1.9	28
8	2.5	10.7	295	16	19	1.9	2.1	31
9	2.6	10.7	294	15	22	1.9	2.0	30
10	2.8	10.8	276	23	22	n.d.	n.d.	n.d.
12	3.1	10.8	276	17	19	2.0	1.7	31
16	3.7	10.9	300	20	20	2.5	2.3	38
22	4.2	11.0	273	24	19	2.7	1.0	38
28	4.6	11.3	278	25	20	2.5	1.3	38
34	4.9	11.1	226	27	18	3.2	1.8	38
40	5.1	11.0	201	26	17	2.9	1.8	38
48	5.2	11.2	200	26	20	2.9	2.0	37
50	5.4	11.2	264	n.d.	n.d.	n.d.	n.d.	n.d.

A 3.3.3.2. Main elements

1. Flow-path sampling after 83 days

Flow-path [cm]	Al [mg/L]	Ca [mg/L]	Fe [mg/L]	K [mg/L]	Mg [mg/L]	Na [mg/L]	Si [mg/L]
1	3.0	32	<0.1	<0.1	<0.1	<0.1	4.6
2	2.7	41	<0.1	<0.1	<0.1	<0.1	7.0
3	2.8	43	<0.1	<0.1	<0.1	<0.1	7.2
4	2.5	48	<0.1	<0.1	<0.1	0.22	9.0
5	2.4	51	<0.1	0.82	<0.1	0.28	9.5
6	2.2	53	<0.1	1.3	<0.1	0.89	11
7	2.2	55	<0.1	1.4	<0.1	1.0	11
8	2.0	58	<0.1	2.2	<0.1	1.7	13
10	1.7	61	<0.1	3.2	<0.1	2.4	14
16	1.6	63	<0.1	7.4	<0.1	5.3	16
22	1.8	64	<0.1	13	<0.1	9.5	16
28	1.8	62	<0.1	15	<0.1	12	16
34	1.8	62	<0.1	15	<0.1	12	16
40	1.9	61	<0.1	17	<0.1	14	16
48	1.4	62	<0.1	22	<0.1	16	16
50	2.0	55	<0.1	19	15.4	14	14

2. Flow-path sampling after 276 days

Flow-path [cm]	Al [mg/L]	Ca [mg/L]	Fe [mg/L]	K [mg/L]	Mg [mg/L]	Na [mg/L]	Si [mg/L]
1	1.3	14	<0.1	0.38	585	1.4	3.3
2	2.1	21	2.5	0.52	318	1.4	3.3
3	2.5	23	3.2	0.52	252	1.5	3.3
4	2.9	26	59	0.71	220	1.7	3.9
5	3.2	29	2.6	0.69	109	1.7	3.9
6	3.3	31	3.8	0.86	87	1.8	4.3
7	3.2	33	3.5	1.0	78	1.9	4.5
8	3.2	35	4.6	1.1	68	2.0	5.0
9	3.2	36	<0.1	1.1	<0.1	2.1	5.4
10	3.1	38	2.6	1.2	57	2.2	6.0
12	2.8	41	3.8	1.4	45	2.4	7.2
16	2.1	48	3.9	2.0	35	3.0	10
22	1.5	55	3.6	2.8	21	3.7	14
28	1.3	59	13.3	3.6	22	4.4	16
34	1.2	60	3.2	4.1	15	5.0	18
40	1.0	62	13.4	5.3	19	6.0	22
48	0.90	61	5.4	6.1	15	6.8	23
50	0.87	60	4.0	7.1	11	9.2	24

A 3.3.3.2. Trace elements

1. Flow-path sampling after 83 days

Flow-path [cm]	As [µg/L]	Ba [µg/L]	Cd [µg/L]	Co [µg/L]	Cr [µg/L]	Cu [µg/L]	Mn [µg/L]	Mo [µg/L]	Ni [µg/L]	Sr [µg/L]	V [µg/L]	Zn [µg/L]
1	11	31	<2	<2	3.0	2.8	<2	<2	<2	71	2.7	4.0
2	<5	35	<2	<2	5.5	4.9	<2	<2	<2	102	5.6	3.4
3	<5	32	<2	<2	7.1	4.0	<2	<2	<2	111	6.1	<2
4	<5	31	<2	<2	10	8.4	<2	<2	<2	131	8.8	2.2
5	<5	32	<2	<2	10	8.7	<2	<2	<2	145	10	<2
6	<5	32	<2	<2	12	8.4	3.8	<2	<2	159	13	<2
7	<5	40	<2	<2	13	16	<2	<2	<2	166	14	<2
8	<5	38	<2	<2	16	18	5.5	<2	<2	177	18	<2
10	<5	39	<2	<2	20	28	<2	<2	<2	197	25	<2
16	<5	48	<2	<2	25	29	11	<2	<2	226	45	<2
22	<5	53	<2	<2	33	48	21	<2	<2	244	64	<2
28	<5	54	<2	<2	39	55	26	<2	<2	242	29	<2
34	<5	50	<2	<2	41	53	25	<2	<2	237	29	<2
40	<5	51	<2	<2	50	58	35	<2	<2	236	30	7.1
48	<5	37	<2	<2	38	42	33	<2	<2	245	73	<2
50	<5	49	<2	<2	25	55	<2	14	<2	228	28	<2

2. Flow-path sampling after 276 days

Flow-path [cm]	As [µg/L]	Ba [µg/L]	Cd [µg/L]	Co [µg/L]	Cr [µg/L]	Cu [µg/L]	Mn [µg/L]	Mo [µg/L]	Ni [µg/L]	Sr [µg/L]	V [µg/L]	Zn [µg/L]
1	<5	15	<2	<2	<2	<2	<2	<2	<2	29	<2	<2
2	<5	19	<2	<2	<2	<2	<2	<2	<2	40	1.3	3.4
3	<5	23	<2	<2	<2	<2	<2	<2	<2	42	1.6	3.5
4	<5	31	<2	<2	<2	12	4.0	<2	<2	54	2.0	34
5	<5	20	<2	<2	<2	2.6	<2	<2	<2	61	2.4	5.1
6	<5	19	<2	<2	<2	3.1	<2	<2	<2	73	2.9	4.3
7	<5	20	<2	<2	2.6	3.8	<2	<2	<2	77	2.9	4.5
8	<5	22	<2	<2	2.7	3.6	<2	<2	<2	82	3.4	4.5
10	<5	21	<2	<2	3.6	4.2	<2	2.5	2.6	95	4.2	4.3
12	<5	23	3.5	<2	4.9	5.0	<2	4.0	<2	106	5.2	4.6
16	<5	25	<2	<2	8.1	6.7	<2	5.0	<2	123	7.4	5.1
22	<5	29	<2	<2	10	8.7	<2	5.8	<2	159	9.6	4.3
28	<5	34	<2	<2	10	12	<2	6.5	<2	176	11	9.2
34	<5	32	<2	<2	8.6	12	<2	6.7	<2	186	12	4.8
40	<5	34	3.4	<2	6.5	16	<2	7.4	<2	196	13	8.2
48	<5	36	<2	<2	5.1	16	<2	9.3	<2	200	15	6.1
50	<5	37	<2	<2	7.2	15	<2	6.0	<2	193	17	4.1