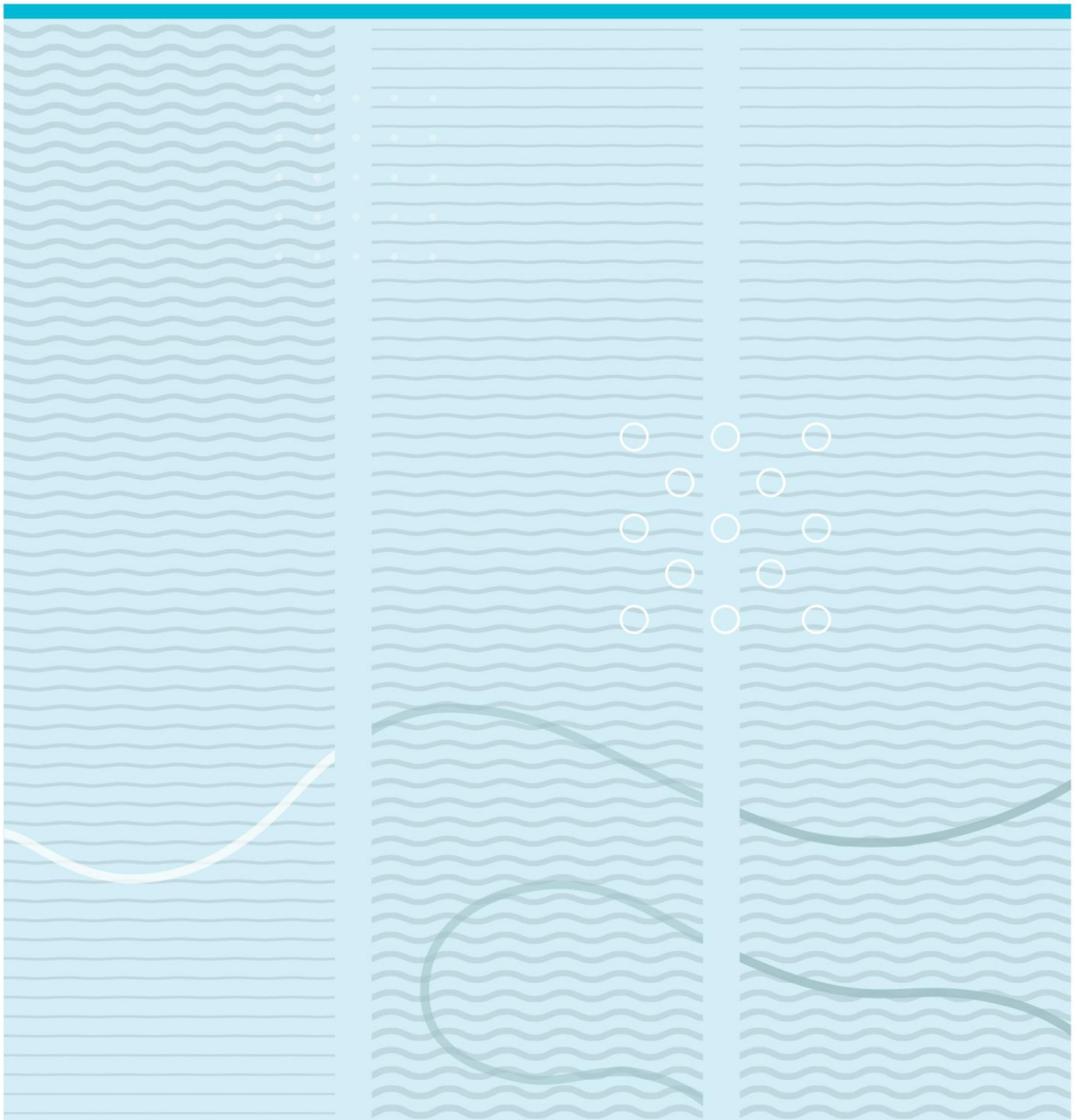# University of South-Eastern Norway

Reidar Dahl Rasmussen

The effect of soil organic matter on the leaching of hexavalent chromium (Cr-VI) from recycled concrete aggregates

Abstract

Norway has a goal of 70 % material recycling of construction and demolition waste by 2020. This portion was only 44 % in 2020. To meet the target, increased reuse of concrete and brick waste is crucial. Recycled concrete aggregates (RCA) are suitable as building materials in ground constructions such as roads or used in landscaping. One challenge with such reuse is possible leaching of hexavalent chromium (Cr-VI) from the RCA with subsequent possible negative effects on recipients.

In the environment, chromium exists in the trivalent (Cr-III) and the hexavalent (Cr-VI) states. Cr(III) is an essential trace metal in humans and is generally less toxic than Cr(VI). Cr(VI) can cause cancer and is highly allergenic. Cr(VI) is also very toxic to aquatic organisms and may have negative long-term effects.

Cr(VI) is found in the cement in concrete because Cr(III) is oxidised to Cr(VI) when the cement is produced. Due to the alkaline nature of concrete, Cr(VI) can leach out from RCA in relatively high concentrations. Soil organic matter (SOM) is found to reduce Cr(VI) to Cr(III) with subsequent retention in soil. In this study the interplay between Cr(VI) and SOM has been investigated by using a standard up-flow column test. The effect of SOM was determined with soil either placed over or under RCA in the columns with different amounts of water added.

Results shown that chromium was found mainly as Cr(VI) in the leachate from RCA and that Cr(VI) in RCA presents a long-term source of Cr(VI) leaching. When RCA leachate percolated SOM, close to 100 % of Cr(VI) was reduced to Cr(III) at first, and then this decreased to approximately 85 %. The decrease was caused by elevated leaching of DOC from the soil trigged by the alkaline RCA leachate. Cr(III) was soluble at low pH ($4-6$) and was adsorbed to SOM or precipitated when the pH increased. No reduction of Cr(VI) was found when SOM leachate percolated RCA. It was found elevated Cr(VI) leaching from RCA due to a first flush effect. It was a positive correlation between increased Cr(VI) leaching from RCA and increased amounts of water added during the column test. SOM was found to buffer against the increased Cr(VI) leaching, through reduction of Cr(VI) to Cr(III), when the RCA leachate percolated SOM.

Sammendrag

Norge har et mål om 70 % materialgjenvinning av bygge- og riveavfall innen 2020. Denne andelen var kun 44 % i 2020. For å nå målet er økt gjenbruk av betong- og teglavfall avgjørende. Resirkulerte betongaggregater (RCA) egner seg som byggematerialer i grunnkonstruksjoner ved f.eks. bygging av veier og ved landskapsutforming. En utfordring med slik gjenbruk er imidlertid mulig utlekking av seksverdig krom (Cr-VI) fra betongen med påfølgende mulige negative effekter på resipienten.

I naturen finnes krom i treverdig (Cr-III) og seksverdig form (Cr-VI). Cr(III) er et essensielt sporstoff hos mennesker og er generelt mindre giftig enn Cr(VI). Cr(VI) kan forårsake kreft og er svært allergifremkallende. Cr(VI) er svært giftig for vannlevende organismer og kan ha negative langtidseffekter.

Cr(VI) finnes i sementen i betong på grunn av oksidering av Cr(III) til Cr(VI) når sementen blir produsert. På grunn av betongens alkaliske egenskaper kan Cr(VI) lekke ut i relativt høye konsentrasjoner fra RCA. Organisk materiale i jord (SOM) er funnet å redusere Cr(VI) til Cr(III) med påfølgende retensjon i jord. I denne studien har samspillet mellom Cr(VI) og SOM blitt undersøkt ved å bruke en standard oppstrøms kolonnetest. I forsøket ble jord enten ble plassert over eller under RCA i kolonnene med tilførsel av forskjellige mengde vann.

Resultatene viste at krom ble funnet hovedsakelig som Cr(VI) i eluatet fra RCA og at Cr(VI) i RCA representerer en kilde til Cr(VI)-utlekking over lang tid. Når RCA-eluatet perkolerte SOM, ble først ca. 100 % av Cr(VI) redusert til Cr(III), og deretter ca. 85 %. Nedgangen var forårsaket av økt utlekking av DOC fra jorda utløst av det alkaliske RCA-eluatet. Cr(III) var løselig ved lav pH (4 – 6) og ble adsorbert til SOM eller felt ut når pH økte. Ingen reduksjon av Cr(VI) ble funnet når eluat med SOM fra jord perkolerte RCA. Det ble funnet forhøyet Cr(VI) utlekking fra RCA forårsaket av en «first flush effect». Det ble funnet en positiv korrelasjon mellom økt Cr(VI) utlekking fra RCA og økte mengder vann tilsatt. SOM ble funnet å bufre mot den økte Cr(VI)-utlekkingen, gjennom reduksjon av Cr(VI) til Cr(III), når RCA-eluatet perkolerte SOM.

Contents

Abbreviations and Acronyms

ALS - ALS Laboratory Group Norway AS

CDW - Construction and demolition waste

Cr(III) – Trivalent chromium

Cr(VI) – Hexavalent chromium

DOC – Dissolved organic carbon

DW – Dry weight

EEA - European Economic Area

EU - European Union

LOR – Limit of reporting

LOQ – Limit of quantification

L/S ratio – Liquid/solid ratio

RCA – Recycled concrete aggregates

REACH - Registration, Evaluation, Authorization and Restriction of Chemicals

NGI – Norwegian Geotechnical Institute

SOM – Soil organic matter

TOC – Total organic carbon

Total Cr – Total chromium

WW – Wet weight

Foreword

This master's thesis forms the final part of the program Master of Environmental Science at University of South-Eastern Norway (USN), Campus Bø. The work was supported by the EarthresQue centre for research driven innovation, Rescue of Earth Materials and Wastes in the Circular Economy, partly funded by the Research Council of Norway, project number 310042/F40.

In accordance with the plan, the laboratory experiments should be conducted at Norwegian Geotechnical Institute (NGI). But due to difficulties in accessing the NGI lab caused by the covid-19 pandemic, the experiments were conducted in the basement of my home.

I would like to thank my supervisor Espen Lydersen at USN and my external supervisor Sarah Hale at NGI for valuable feedback, guidance, and encouragement. I would also like to thank my employer Asplan Viak AS who offered me a reduced work schedule so that I could complete my thesis.

Sandefjord, 07.11.22

Reidar Dahl Rasmussen

1 Introduction

Recycling goals for construction and demolition waste

In Norway, 2.1 million tonnes of construction and demolition waste (CDW) was generated in 2020 (Statistisk sentralbyrå, 2021a). As member of the European Economic Area (EEA), Norway has to follow the European Union's (EU) Waste Framework Directive (Waste Framework Directive, 2018). The directive states that by 2020, 70 % of CDW must be material recycled (Stortinget, 2018). In 2020, this portion was only 44 % in Norway (Statistisk sentralbyrå, 2021b). Most EU countries reached the 70 % goal in 2016 (European Environment Agency, 2020) showing Norway has some progress to make in order to meet the target set. Since concrete and brick waste makes up a large proportion of CDW, at about 60 %, (Statistisk sentralbyrå, 2021b), it is crucial to increase the recycling rate of this waste fraction to reach the 70 % goal. Recycled concrete aggregates (RCA[1]) from demolition is suitable as building materials for constructions such as roads, car parks or used in landscaping. However, one challenge with such reuse is possible leaching of hexavalent chromium (Cr-VI) from the concrete with subsequent possible negative effects on the recipient.

Chromium in the environment

Chromium (Cr) is amongst the 25 most widespread elements in the Earth´s crust (Roskovic et al., 2011). Chromium exist in the environment in the trivalent (Cr-III) and the hexavalent (Cr-VI) states (Bjerregaard, 2010); (Rai et al., 1989). Cr(III) is an essential trace metal in humans contributing in the metabolism of cholesterol, fat and glucose (Barceloux, 1999), while Cr(VI) has several negative damaging effects on the human body, even in small amounts. It can cause cancer, is highly allergenic, and some compounds can also damage genetic material and reproduction. Cr(VI) is very toxic to aquatic organisms and may have negative long-term effects. Cr(III) is generally less toxic than Cr(VI), but some animal species may also be sensitive for Cr(III) compounds (Miljøstatus, 2021).

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[1] RCA is crushed concrete gained from demolition waste which either partially or completely replaces natural aggregates in constructions (Guo et al., 2018).

In water without organic ligands Cr(III) is found as Cr^{3+} at pH < 5.5, $Cr(OH)^{2+}$ at pH < 6.5 or $Cr(OH)_2^+$ at pH > 4.5. At pH ≈ 5.5 – 6.5 Cr(III) hydroxides starts to precipitate (Lydersen et al., 2002). Cr(III) forms stable complexes with negatively charged organic and inorganic compounds in water and soil (Lydersen et al., 2002) (Steven et al., 1976). In pure water Cr(VI) is found as oxyanions in the form chromate (CrO_4^{-2}) at pH > 6.5, hydrochromate $(HCrO_4^-)$ at pH < 6.5 or dichromate $(Cr_2O_7^{-2})$ at very low pH (Lydersen et al., 2002). These oxyanions do not make complexes with anionic particulate matter and are therefore quite mobile in the environment (Saleh et al., 1989) (Steven et al., 1976). Cr(VI) compounds are powerful oxidizing agents especially in acidic solutions and will react with oxidizable substances, usually organic matter, to form Cr(III) (Steven et al., 1976). For this reason, Cr(VI) can only exist in water that contains little organic matter. Fe(II) and sulfides have same effect on reducing Cr(VI) to Cr(III) as organic matter (Lydersen et al., 2002). Due to the high redox potential of the Cr(VI)/Cr(III) couple there are few oxidants present in natural systems which can oxidize Cr(III) to Cr(VI). Oxygen and manganese oxides are two oxidants that are most likely to be able to achieve this. The oxidation of Cr(III) by oxygen is a very slow process but it is faster by manganese oxides (Rai et al., 1989).

Leaching of Cr(VI) from recycled concrete aggregates

Concrete contains chromium which mainly originates from iron agents, clay, limestone, and fuels used in the production of cement. In the production of cement clinker, which occurs at high temperatures, Cr(III) oxidizes to form Cr(VI) (Roskovic et al., 2011; Wærner et al., 2019). Concrete is alkaline because calcium hydroxide $Ca(OH)_2$ in equilibrium with water gives a pH of 12.35. Small amounts of sodium and potassium hydroxide (NaOH and KOH) in the concrete can increase the water pH to 13.5 when leaching out (Engelsen et al., 2012a). In alkaline leachates both Cr(III) and Cr(VI) will be in the oxyanionic form as $Cr(OH)_4^-$ and CrO_4^{2-}, respectively. CrO_4^{2-} can leach out in relatively high concentrations but not $Cr(OH)_4^-$ because the equilibrium with insoluble Ca-Cr[III] minerals in alkaline systems causes the $Cr(OH)_4^-$ concentrations to be very low (Cornelis et al., 2008)[2].

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[2] Cornelis et al. (2008) referred to (Kuehn & Mudersbach, 2004) but the information was not found there. Cornelis et al. (2008) have probably referred to wrong article.

The REACH[3] Directive (REACH, 2006) has set a limit value of 2 mg kg^{-1} Cr(VI) in cement and cement containing mixtures which is implemented in Norwegian regulations (Miljøstatus, 2021). To meet the limit value, the European cement industry is using reducing agents, mostly Iron(II) sulphate, during the production of cement. This practise have been followed for more than 20 years (Engelsen et al., 2020). Previous leaching test showed little or no leaching of Cr(VI) from a concrete produced in 2018 (Eckbo, 2019) showing compliance with this limit value. However, leaching of Cr(VI) from older concrete structures which are being demolished may become an environmental problem.

Indeed, previous studies have shown that the levels of chromium leached from RCA can exceed safe threshold concentrations. Miljøstyrelsen (2018) found in leaching tests on RCA that the leaching of total chromium (total Cr) exceeded Danish limit values with limited reuse possibilities consequently. Butera et al. (2015a) found leaching of Cr(VI) from RCA in some cases were even above the EU acceptance criteria for inert waste landfills. In 2020 a limit solid concentration of 8 mg kg^{-1} Cr(VI) came into force when reusing RCA and bricks in Norway (FOR-2020-02-03-510, Kap. 14A). In previous tests Eckbo et al. (2022) found that the limit value was exceeded in some, but not all, concrete samples from demolition projects. When exceeding the limit value disposal at landfill sites rather than reuse often is the consequence. They also found no correlation between the solid concentration of Cr(VI) in concrete and the Cr(VI) leached. As a solution to these challenges, previous works has shown retention of Cr(VI) when leachate from RCA is in contact with soil organic matter (SOM) (Eckbo et al., 2022) (Butera et al., 2015b). When RCA is reused, it will most often be in direct contact with soil rather than in direct contact with a recipient such as groundwater. Therefore it is relevant to investigate the leaching of Cr(VI) when RCA and soil are used in one system in order to get a more realistic picture of environmental risk (Eckbo, 2020b).

The effect of soil organic matter on the leaching of Cr(VI)

Soil organic matter (SOM) can be divided into three main parts; small fresh plant residues, organic matter that undergoes decomposition and more stable organic matter, often termed humus (United States Department of Agriculture, 2013). Humus is organic matter

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[3] REACH (Registration, Evaluation, Authorization and Restriction of Chemicals).

that due to extensive decomposition is resistant to further alteration (Foth, 1984). During humification of organic matter, lignin is altered in such a way that there is an increase in the carboxyl and phenolic OH groups. The hydrogen of both groups can subsequently be replaced by cations which gives rise to cation exchange capacity (Foth, 1984). Humus contains a significant amount of organic acids. These acids play an important role when it comes to water pH, pH buffering and by metal-complexing properties (Lydersen et al., 2002).

Organic soils (Histosols as peat) have their origin almost entirely from organic matter in different levels of decomposition. The organic matter content is ≥ 20 %. Mineral soils are soils primarily composed of inorganic mineral matter, and their properties are more determined by mineral matter, as the organic matter content usually is < 20 % (Foth, 1984).

In soils Cr(VI) can be reduced to Cr(III) by soil humic substances, but also by aqueous inorganic species, electron transfer at mineral surfaces or by non humic organic elements such as carbohydrates and proteins. For Cr(VI) reduction, soil humic substances provide a large amount of electron donors (Kožuh et al., 2000). Soil humic acid can reduce Cr(VI) to Cr(III) in aqueous solution in the pH range 2 − 7. The reduction rate increases with decreasing pH, decreasing amounts of Cr(VI) and increasing amounts of soil humic acid (Wittbrodt & Palmer, 1997). Bartlett and Kimble (1976) found about spontaneous reduction of Cr(VI) to Cr(III) with presence of organic matter in soil, also at pH above neutrality. They found no reduction in soil with very low content of organic matter except if an energy source was added.

The effect of precipitation on the leaching of Cr(VI) from recycled concrete aggregates

As a result of climate change, forecasts show that annual precipitation will increase by about 18 % towards the end of this century i Norway. Heavy rainfall events will be more intense and occur more frequently (Miljødirektoratet, 2016). Since precipitation can affect the leaching of Cr(VI) from RCA (Eckbo et al., 2022) (Kayhanian et al., 2009) (Takahashi et al., 2007), climate change makes this issue even more relevant.

In previous leaching tests, Eckbo et al. (2022) reported an elevated initial leaching of Cr(VI) from RCA followed by a rapid decrease and explained this as a first flush effect. The first flush effect is related to an assumption that the first part of runoff from surfaces in a storm event is the most polluted (Deletic, 1998). Kayhanian et al. (2009) found elevated leaching of Cr(VI) from concrete surfacing materials on highways, followed by a significant decrease, explained as a first flush effect. In tank leaching experiments on concrete and mortar,[4] Takahashi et al. (2007) reported similar findings with elevated leaching of Cr(VI) from dried concrete in contact with water, followed by a rapid decrease. Takahashi et al. (2007) also found an increase in cumulative leaching of chromium from mortar with increased L/S ratio (liquid/solid ratio). As explanation they suggest that increased concentrations of chromium in the water eluate restricted further diffusion of chromium from the mortar.

Aim of this study

The overall aim of this study was to investigate the effect of soil organic matter (SOM) on the leaching of Cr(VI) from recycled concrete aggregates (RCA). Experiments were carried out to identify how to minimize leaching of Cr(VI) from concrete and thus provide a viable reuse solution that is protective for the environment. The investigations were carried out using a column set up. The following hypotheses were tested:

a. Most of the chromium that leaches from the RCA will be in the hexavalent form (Cr-VI).
b. The concentration of Cr(VI) in the leachate water will decrease when the RCA is in contact with SOM.
c. The leaching of Cr(VI) from the RCA will increase with increasing simulated precipitation volumes.

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[4] Mortar contain about 30 % cement and concrete contain 10 − 15 % cement (Portland Cement Association, 2022), which means that mortar have a higher amount of Cr(VI) than concrete.

2 Materials and method

2.1 Experimental column standard

The experiments were conducted using up-flow percolation column tests mainly following standard NS-EN 14405:2017 (Standard Norge, 2017). Deviations from the standard set up are discussed in chapter 2.9. The standard describes an up-flow percolation column test which can be used to investigate leaching behaviour of inorganic and non-volatile organic substances from granular waste materials under standardized percolation conditions.

2.2 Material sampling and preparation

The materials used in the column test were concrete (RCA), peat and sand. Since the focus of the experiment was to investigate the effect of soil organic matter (SOM) on the leaching of Cr(VI), a peat soil was chosen. Sand was chosen to compare with peat, as sand is an organic poor soil type, about neutral pH in suspension and with low cation exchange capacity (Foth, 1984).

Crushed concrete material (RCA) from the demolition of Lambertseter nursing home, built in 1969 was used in the column test. At NGI (Norwegian Geotechnical Institute) the sample was crushed to approximately 40 mm first using a sledgehammer and then a jaw breaker with an adjustable aperture (Retsch Germany jaw breaker type BB100 wolfram carbide), before the material was sieved to ≤ 4 mm grain size (C. Eckbo (NGI), pers.com.). The sand was from Sigma Aldrich and was quartz (SiO_2) with a grain size from 0.2 to 0.3 mm. The peat (hereafter called soil) was from Sundland Torv og Jord AS and was sieved to a grain size ≤ 4 mm before use. The materials used in in the experiments are shown in Figure 1.

Figure 1: The prepared materials used in in the leakage experiments. From the left; concrete (RCA), soil (peat) and sand.

2.3 Column setup

The experiments were carried out using the column set up as described below (Table 1 and Figure 2) with concrete (RCA), soil and sand in different combinations/order, all combinations in triplicates, and one blank for each material included in the combination tests., i.e. totally 15 columns.

Table 1: Column setup (n =15) in triplicates and blanks with concrete (RCA), sand and soil. The use of "under" and "over" in the names of the triplicates was given in accordance with the up-flow direction of the water (i.e. that the columns are fed from the bottom with water), which is reversed in relation to reality where precipitation moves from the top of the ground and downwards.

	Column No.	Name
Triplicates	1	A. Concrete + sand under
	2	
	3	
	4	B. Concrete + soil under
	5	
	6	
	7	C. Concrete + sand over
	8	
	9	
	10	D. Concrete + soil over
	11	
	12	
Blanks	13	Concrete
	14	Sand
	15	Soil
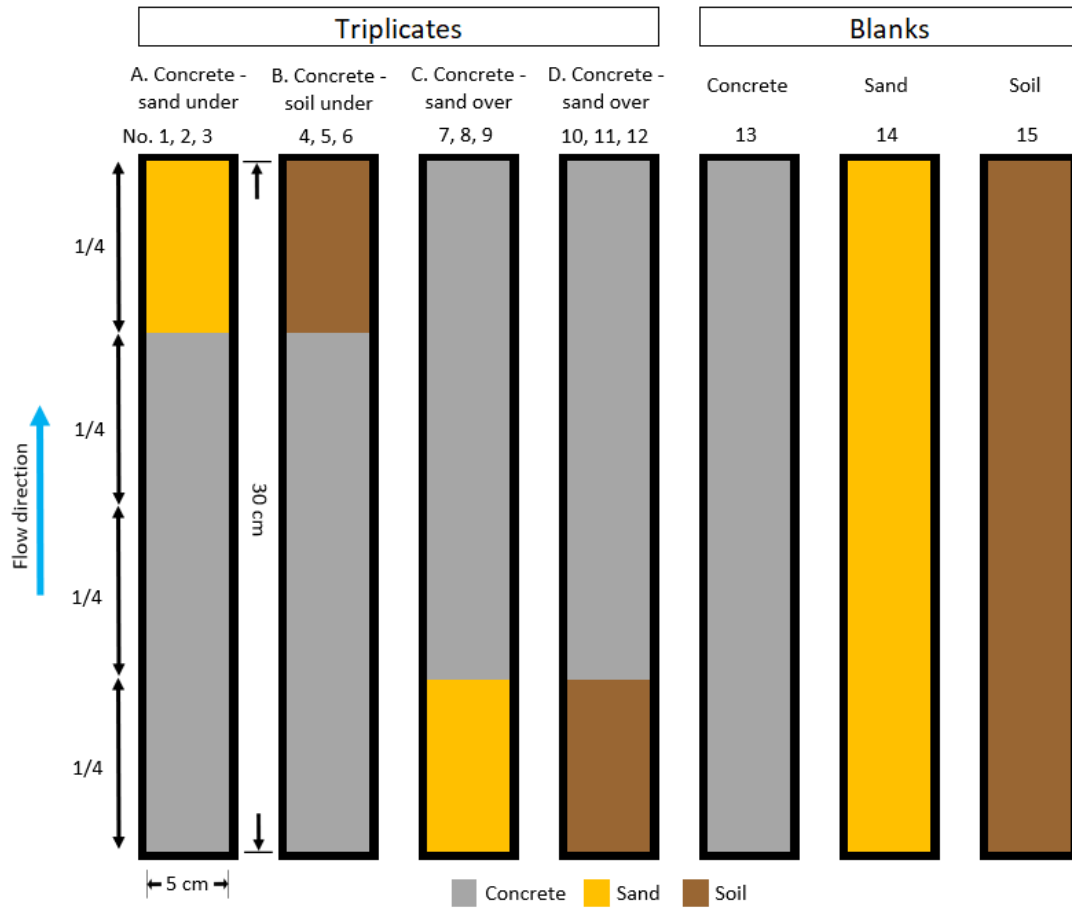
Figure 2: Column setup (n = 15) in triplicate and blanks with concrete (RCA), sand and soil. Flow direction (blue arrow) of the water is shown. The use of "under" and "over" in the names of the triplicates was given in accordance with the up-flow direction of the water which is reversed in relation to reality where precipitation moves down into the ground. The triplicates were filled with on quarter sand or soil and three quarters concrete (RCA). The inner diameter of the columns was 5 cm and filling height 30 cm +/- 5 cm.

2.4 Column equipment and packing

The equipment used in the column test is shown in Figure 3 with pictures of equipment items numbered. Before packing, a bottom plug (1) with O-ring (2) and teflon tube (6) was mounted in the column (1) and a membrane filter (3) was put on the top.
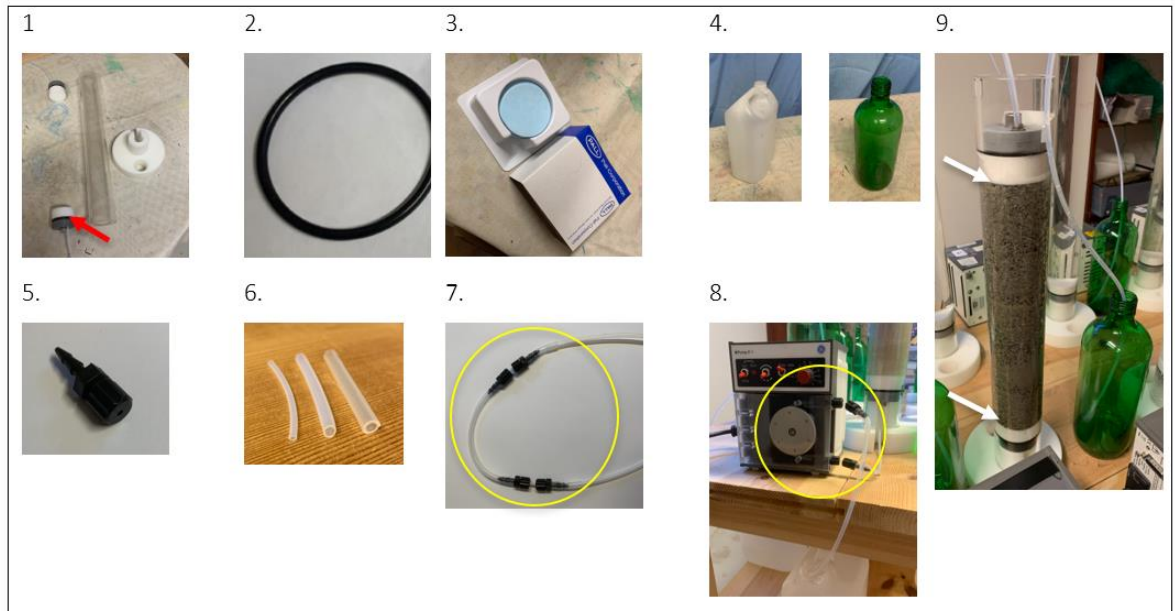
Figure 3: The equipment:

- *1 – Column (both glass and plastics) (inner diameter 5 cm and height approx. 45 cm), plugs (top/bottom, with hole for tube) and foot.*
- *2 – O-ring, 3 mm (red arrow in 1. indicate location on plugs).*
- *3 – Membrane filter (PES, polyethersulfone), 0.45 µm cut-off, 47 mm diameter (white arrows in picture 9 indicate location of the filter in the column).*
- *4 – Plastic jug (5 L) for milli-Q water and 1 L glass bottle for water eluate sampling.*
- *5 – Tube fitting.*
- *6 – Tubes. From left; tube to connect fittings (hard plastic, diameter internal/external 1 mm/2 mm), tube to connect to the plugs (teflon, 3 mm/4 mm) and pump tube (silicone, 3 mm/5 mm).*
- *7 – Tubes and fittings finished (yellow rings in 7 and 8 indicate location in the pump).*
- *8 – Peristaltic pump (type P1, GE Healthcare Bio-Sciences AB).*
- *9 – One column finished set up and filled with material.*

The columns were packed with materials as shown in Figure 4. To obtain an evenly distributed mass the materials were packed in twelve 2.5 cm layers to give a total height of 30 cm (+/- 5 cm). Before packing the density was found by weighing 1 litre of each material 3 times on a kitchen scale without compressing. Based on the density, the weight needed to fill 2.5 cm of the column was found. Each layer was compressed by a rammer of 125 g which was dropped 3 times on the top of the added material (Figure 4).

Figure 4: Packing of the 2.5 cm layers in the column using a rammer of 125 g. White arrow shows up/down movement of the rammer.

After packing, a similar plug as at the bottom, with O-ring, teflon tube and membrane filter, were put on top. Then the rest of the tubes and fittings (picture 5, 6, 7 in Figure 3) were connected to the column and the pump (picture 8 in Figure 3). Finally, the plastic jug for milli-Q water and 1 L glass bottle for water eluate sampling were added to the set-up (picture 4 in Figure 3). The total column setup is shown in Figure 5.

Figure 5: The total column set up.

2.5 L/S ratio calculations

The results of the column test (NS-EN 14405:2017) are expressed as a function of Liquid/Solid ratio (L/S ratio). When presenting the release of a substance from a material in a column, the L/S ratio describes how much water (L) which has percolated through a given amount of material (S) at the defined sampling point. L/S is expressed in L kg^{-1} dry matter. Before starting the test, the amount of water needed to achieve L/S 0.1 and L/S 10 was calculated (equation 1., next page). These ratios correspond to short and long term leakage (Sørmo et al., 2019) and are most commonly used in leaching tests. Based on available equipment, the water was weighed (reported in mL).

Equation 1:

$$L = S_w \times {}^{L}/_{S}$$ (1)

Where

L *Volume of water eluate in mL*

S_w *Wet weight of solid materials in g (later adjusted to dry weight)*

${}^{L}/_{S}$ *Unitless L/S ratio, e.g. 0.1*

The column test was run for 4 months to simulate as close as possible natural precipitation. To reach L/S 10 within that time frame, it was necessary to add 500 mL of water per week to each column. L/S 0.1 would then be reached after 1 - 2 days. 500 mL/week is like a linear water velocity of 3.7 cm/day in the columns and corresponds to about 15 times the average weekly precipitation[5] in Oslo. Over the experimental period, 11 samples were taken, weekly at first and then about every second week. At each sampling the L/S ratios were calculated by dividing the accumulated volume of water eluate (L) by the wet weight of the solid material (S_W).

While running the test, calculations were based on wet weight of the materials. After the test was finished, the L/S ratios were calculated based on dry weights by dividing the accumulated volume of the water eluate (L) by the dry weights of the materials (S_d). The dry weight was found using equation 2:

$$S_d = \frac{S_w \times S_{d\%}}{100}$$ (2)

Where

S_d *Dry weight of the materials in g*

S_w *Wet weight of the materials in g*

$S_{d\%}$ *Dry weight in % of wet weight*

All presented results are based on L/S ratios where S is given in dry weight.

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[5] The calculation is based on yearly average precipitation of 889 mm for the period 2016 to 2020 (Blindern, SN 18700) (Norsk Klimaservicesenter, 2022).

2.6 Running the column test

Before the pumps were started, 5 L plastic jugs had been filled up with milli-Q water. The speed switches on the pumps were set to deliver 500 mL per week. Then the pumps were started. After about 12 hours, the columns were saturated with water. The pumps were then switched off for 3 days to achieve equilibrium conditions while saturated. After the equilibration period, the pumps were started again to run the column test. After 12 weeks the speed was increased to 1000 mL/week (30 times the average weekly precipitation in Oslo (Norsk Klimaservicesenter, 2022).

The total duration of the test was about 4 months. Samples were taken out for analysis from each of the 15 columns. The first eluate samples were taken out after 1 to 2 days, when L/S 0.1 were reached. Thereafter, samples were taken out weekly during the first month and about every second week for the remaining three months until L/S 10 was reached. One last sample were taken out after L/S 10 was reached.

2.7 Analysis

Solid materials

Samples of 200 g of each solid material were put into Rilsan bags and sent to ALS Laboratory Group Norway AS (ALS) and analysed for dry weight, pH, conductivity, TOC, total Cr, Cr(III) and Cr(VI). Analytical methods at ALS are shown in Table 2. ALS states that LOR is equal to or higher than the limit of quantification (LOQ). In most cases LOR is equal to LOQ, but in some cases LOR is higher than LOQ for instance according to legislation in a country (R. Telstad (ALS), pers.com.). LOQ is the lowest concentration where an analyte can be reliably detected and predefined goals for bias and imprecision are met (Armbruster & Pry, 2008).

The dry weight of the concrete (RCA) was taken from the NGI report "Leaching and transport of hexavalent chromium (Cr-VI) from reused concrete" (Eckbo, 2020a).

Table 2: Overview of analytical methods for parameters analysed in solid materials (concrete (RCA), sand, and soil) at ALS. Limit of reporting (LOR) and measurement uncertainty (MU) is described.

Parameter	Method
Dry weight	DS 204 – A representative sample quantity is weighed up and dried at 105 °C for about 20 hours. After cooling, the sample is weighed again, and the dry matter percentage is calculated. MU: 20%. LOR: 0.1 %.
pH	DS/EN ISO 10523:2012 - Electrochemical measurement of water and soil suspension.
TOC	EN 13137:2001 - Determination of total organic carbon by IR method. MU: 15 %. LOR: 0.1 %.
Conductivity	DS 288 - The soil is dried at 60 degrees, before a certain amount of water is added. Then the sample is shaken for 30 minutes and filtered before the conductivity is measured in the sample. MU: 3 %. LOR: 0.1 mS m^{-1}.
Total Cr, Cr(VI) and Cr(III)	Total Cr: DS 259:2003 + DS/EN 16170:2016: The sample (3 g) is dissolved with HNO_3 and autoclaved (boiled) at 120 degrees for 30 min. The sample is then analysed with an ICP-MS device (Inductively coupled plasma mass spectrometry). MU: 30 %. LOR: 0.2 mg kg^{-1}. Cr(VI): ISO 15192:2010: The sample is dried in an oven at 40 °C overnight. A smaller subsample is taken out and added manganese chloride ($MnCl_2$) to prevent oxidation from Cr(III) to Cr(VI). The sample is then dissolved in a basic solution consisting of NaOH and Na_2CO_3, and then thermally broken down at 90 – 95 °C in minimum one hour. The sample is then diluted with HNO_3 and analysed with an ICP-MS device. MU: 40 %. LOR: 0.2 mg kg^{-1}. Cr(III): Is calculated based on the analysis result for total Cr and Cr(VI). Total Cr - Cr(VI) = Cr(III).

Water eluate

Water eluates were filtrated through a 0.45 µm membrane filter mounted at the top (and bottom) of the columns (Figure 3). When water eluate samples were taken, weight, colour, and other visual features were noted before collected in polyethylene (PE) plastic bottles from ALS and stored at about 15 °C prior to sending to ALS. At ALS the water eluates were analysed for content of DOC, total Cr, Cr(III) and Cr(VI) without dissolving. pH and conductivity were analysed in situ. Experiments were carried out at room temperature (20 °C). The equipment and bottles used during the water eluate analyses is shown in Figure 6. Analytical methods in situ and at ALS are shown Table 3.
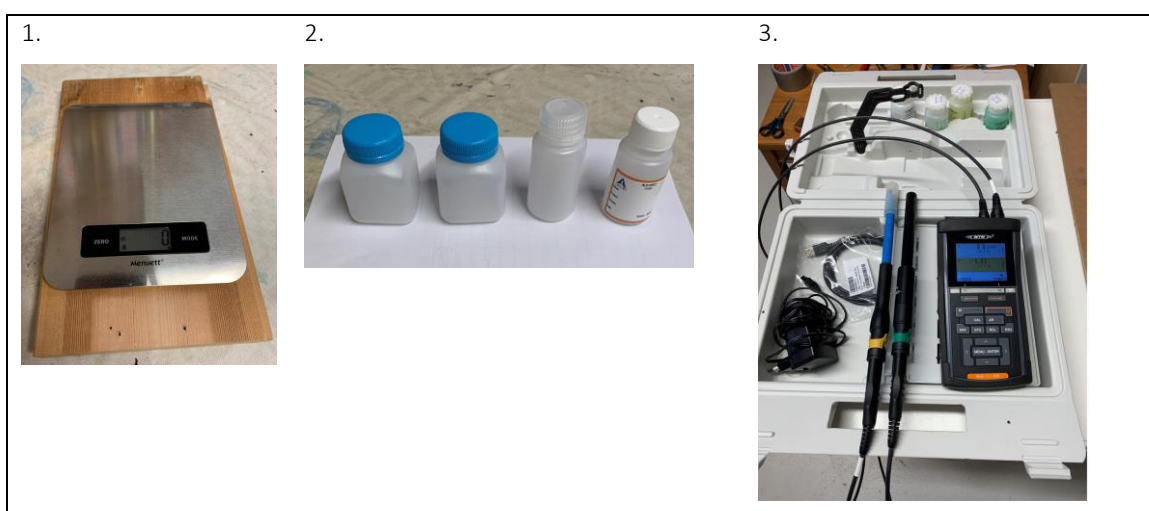
Figure 6: Equipment and bottles used during the analyses:

- *1 – Kitchen scale to weigh the materials and the water eluate.*
- *2 – Sample bottles sent to ALS: From the left; storage for eventually later analysis/control (100 ml), DOC (100 ml), total Cr (60 ml), Cr(VI) (60 ml preserved).*
- *3 – pH/conductivity meter (WTW Multi 3620 IDS)*

Table 3: Overview of analytical methods for parameters analysed in water eluates in situ and at ALS. Limit of reporting (LOR) and measurement uncertainty (MU) for analyses at ALS is described.

Parameter	Method
pH	In situ: With use of a multi-parameter portable meter (WTW Multi 3620 IDS) (Figure 6).
Temp, °C	
Conductivity	
Total Cr	ALS: (US EPA 200.7, ISO 11885, CSN EN 16192, US EPA 6010, SM 3120, CSN 75 7358 samples prepared in accordance with method for Cr(VI) analysis). Determination of elements at AES with ICP and stoichiometric calculations of the concentration of current compounds from measured values including calculation of total mineralization and calculation of the sum Ca + Mg. The sample was homogenized and mineralized with nitric acid in an autoclave below high pressure and temperature before analysis. MU: 10 %. LOR: 0.002 mg L^{-1}.
Cr(VI)	ALS: (CSN EN 16192, EPA 7199, SM 3500-Cr). Determination of hexavalent chromium by ion chromatography with spectrophotometric detection. MU: 10 %. LOR: 0.4 µg L^{-1}.
Cr(III)	ALS: Cr (III) is determined by calculation: Cr tot - Cr (VI) = Cr (III).
DOC	ALS: (CSN EN 1484, CSN EN 16192, SM 5310) Determination of total organic carbon (TOC), dissolved organic carbon (DOC), total inorganic carbon (TIC) and total carbon (TC) by IR detection. MU: 20 %. LOR: 0.5 mg L^{-1}.

Precipitate

From start and throughout the test period a precipitate was observed in all the water eluate samples, except from the blanks "Sand" and "Soil". After filling the bottles to be sent to ALS, the remaining water eluate was filtered. The precipitate was analysed for total Cr content at USN, Campus Bø. The analytical method is shown in Table 4.

Table 4: Analytical method for total Cr content in precipitate at USN, Campus Bø.

Parameter	Method
Total Cr	The precipitate was dissolved in 0,1 N HNO₃, solution (pH = 1) for 24 hrs. before being analyzed for total Cr by ICPMS (ICP-MS Agilent 7850 ICP-MS).

2.8 Data analysis

The analysis results were transferred to excel where tables and diagrams were prepared. Mean and standard deviation for the triplicates columns was calculated except for pH where only the mean was calculated. The mean for pH was found by converting the pH values into concentrations of hydrogen ions, then finding the means for the hydrogen concentrations and then reconverting into pH values. To use data reported as below the LOR in diagrams, the < sign was removed, and the number was divided by 2. To calculate released substances cumulatively per kg of material (dry weight) for all eluate fractions formula 3 was used (NS-EN 14405:2017):

$$U_i = \frac{(V_i \; x \; c_i)}{(m_0)} \tag{3}$$

Where

U_i *Released quantity of a component per quantity of sample for analysis in water eluate fractions, i, in mg kg⁻¹ dry matter*

V_i *Volume of the water eluate fraction, i, in L*

c_i *Concentration of the component concerned in the water eluate fraction i, in mg L⁻¹*

m_0 *Dry mass of the solid type(s) in the column, in kg*

2.9 Quality control and quality assurance

Before the concrete (RCA) (12 kg) was sieved to ≤ 4 mm, it was mixed by hand to avoid loss of fines (C. Eckbo (NGI), pers.com.). In total, 15 kg of organic soil (peat) was taken out from 8 different sites within an outdoor bin at Sundland Torv og Jord AS, before

approximately 2 kg was subsampled for use in column tests. The sand was used as received.

After the column was set up and before they were filled with materials, they were tested by pumping water through them. The test was important to check for possible leaks in fittings and tubes and gain experience in setting the speed of the pumps. Based on available equipment, the water eluate was weighed rather than measured in volume. The density of water is very close to 1, i.e. 1 mL = 1 g. Any discrepancy that may have resulted due to using water weight rather than volume is therefore negligible.

To increase the representativeness, all columns with same order of distribution between concrete (RCA), sand or soil were set up in triplicates. One column with only soil, only concrete or only sand (3 columns) was used as control columns, i.e blanks. Milli-Q water produced by reverse osmosis was used in the test to avoid possible influence from macro chemical components in the water.

Deviation from the standard

L/S ratios should be calculated based on the dry weight of materials according to standard NS-EN 14405:2017. While running the test, the ratios were based on wet weights and adjusted to dry weights after all flow through experiments were finished. The adjustments led to some increase in the L/S ratios, negligible for the lower ones, but somewhat higher at higher ratios, as shown in Annex 2. For the blank "Soil" the deviation was high which also was the case before the adjustments due to the low density of the soil. Thus, when comparing against the other blanks and triplicates in figures it was only possible to show the analysis results at L/S 2.4 and L/S 18 for the blank "Soil". For the research results presented here, the deviations in L/S ratios have had little significance.

According to standard NS-EN 14405:2017 the total accumulated amount of water eluate should be analysed. This was not possible for the samples taken every two weeks. When the speed was increased to 1000 mL water per week, the bottles were emptied between two and six times before the last analysis. Owing to this there may be some slight deviations compared to the standard method description.

For columns with an inner diameter of 5 cm, the water velocity should be set at 15 cm/day, or approximately 2000 mL per week according to NS-EN 14405:2017. In this experiment, the water velocity was 500 mL/week which was increased to 1000 mL/week towards the end of the experiment. The lower water velocity may have given different physical or chemical leaching conditions and a different result compared with the standard setup.

Possible sources of error

In the first 4 weeks of the test, the membrane filters at the top of the columns 4, 5 and 6 (triplicates of "B. Concrete + soil under") became clogged. The biggest problem was for column No. 5. When the filters became clogged, they had to be replaced. When the filters were replaced, some water had to be drained out at the bottom of the columns. The lost water drained was included in the calculation of the L/S ratios but was not analysed. To solve the problems with clogging, a 1.5 cm thick layer of the sand used in the experiment was placed between the filter and the soil layer. The clogging probably has given some changed conditions compared to the other triplicates at least in the early stage of the test. The 1.5 cm thick sand layer has probably given somewhat changed conditions compared to the other triplicates. The changes are assumed to be small due to the neutral chemical properties of the sand.

For column No. 4, 5 and 6 (triplicate "B. Concrete + soil under") the LOR for Cr(VI) concentration in the water eluate varied between < 0.4 µg L^{-1} and < 4.0 µg L^{-1} in samples taken out at day 2, week 1 and week 2. The elevated LOR is described as matrix interference in the analysis reports from ALS (Annex 5). In some cases, the concentration of Cr(VI) was just above the concentration for total Cr. ALS assumed this discrepancy being a result of different analytical methods for the analysis of Cr(VI) and total Cr (M. V. Larsen (ALS), pers.com.).

3 Results

3.1 Description of solid materials and L/S ratios

Physical and chemical parameters for the solid materials used in the column test are shown in Table 5.

Table 5: Physical and chemical parameters of the solid materials used in the column test. Wet weight is shown as ww. Dry weight is shown as dw.

	ww	dw	pH	TOC	$K_{25°C}$	Total Cr	Cr(III)	Cr(VI)
Material	kg L^{-1}	%		% of dw	mS cm^{-1}	µg kg^{-1} dw	µg kg^{-1} dw	µg kg^{-1} dw
Concrete (RCA)[1]	1.55	95.7	12.0	0.17	5.6	21000	17000	4100
Sand	1.44	99.6	6.9	< 0,10	0.095	310	310	< 200
Soil	0.20	22.1	4.5	23	0.085	1400	1400	< 200

[1] Concentration of total Cr, Cr(III) and Cr(VI) for concrete (RCA) is at ww.

Annex 1 shows the volume and weight (wet weight) of the materials packed in the columns. The total weight of the materials in the columns after packing was approximately 900 g for the triplicates "A. Concrete + sand under" and "C. Concrete + sand over" and 715 - 730 g for the triplicates "B. Concrete + soil under" and "D. Concrete + soil over". For the blanks the weights were approximately 910, 850 and 130 g for "Concrete", "Sand", and "Soil" respectively.

Annex 2 shows the results of the calculation converting the wet weight of the materials to dry weight after the column test was finished. In Table 6 (Annex 3 for each column) the L/S ratios is shown for the triplicates and the blanks based on dry weight for the materials. Sampling week for the water eluate and added amount of water per week is also shown. Table 6 shows that the lowest L/S ratio for the blank "Soil" was 2.4 and the highest was 316.8, whilst for the other columns the lowest L/S ratios was 0.1 and the highest in the range L/S 12 to 15. Thus, the blank "Soil" was very different to the other columns. When comparing the analysis results for "Soil" against the results for the other columns in diagrams, it was only possible to consider the results at L/S 2.4 and L/S 18 for "Soil". The triplicates "B. Concrete + soil under" and "D. Concrete + soil over" had higher

standard deviations than Triplicate "A. Concrete + sand under" and "C. Concrete + sand over" (Table 6).

Table 6: Calculated L/S ratios at each sampling time (given by week number from start). The ratios are given with mean and standard deviation for the triplicates. The ratios are based on dry weight of the materials. The volume of added water per week is also shown. It was not taken out samples at L/S 9.8 for triplicate D.

	Name	No.		Sampling week										
				(day 2)	1	2	3	4	6	8	10	12	14	16
				L/S ratios										
Triplicates	A. Concrete + sand under	1, 2, 3	Mean	0.1	0.7	1.3	1.8	2.5	3.8	5.2	6.4	8.9	10.4	12.6
			Std. dev.	0.0	0.0	0.1	0.2	0.3	0.1	0.2	0.3	0.3	0.1	0.2
	B. Concrete + soil under	4, 5, 6	Mean	0.1	0.9	1.6	2.3	3.1	4.6	6.1	7.4	10.0	10.8	14.2
			Std. dev.	0.0	0.2	0.3	0.6	0.4	0.2	0.2	0.3	0.3	0.0	0.1
	C. Concrete + sand over	7, 8, 9	Mean	0.1	0.6	1.1	1.6	2.3	3.5	4.8	6.0	8.5	10.4	12.2
			Std. dev.	0.0	0.1	0.1	0.3	0.4	0.2	0.1	0.1	0.2	0.0	0.1
	D. Concrete + soil over	10, 11, 12	Mean	0.1	0.8	1.6	2.4	3.1	4.8	6.6	8.2	9.8	10.8	15.2
			Std. dev.	0.0	0.2	0.0	0.1	0.2	0.3	0.5	0.7	0.2	0.1	0.7
Blanks	Concrete	13		0.1	0.5	1.2	1.8	2.4	3.5	4.7	6.1	8.6	10.5	12.2
	Sand	14		0.1	0.7	1.3	2.0	2.7	4.0	5.4	6.6	8.9	10.0	12.5
	Soil	15		2.4	18.0	34.0	40.3	46.6	65.0	100.3	133.5	168.4	230.8	316.8
				Added water, mL/week										
				500								1000		

3.2 Water eluate results

Table 7 show the results for the analysed parameters for the triplicates water eluate at the respective L/S ratios. Relative standard deviation ≥ 30 % and concentrations below the limit of reporting (LOR) are marked. The concentration of Cr(III) was mainly < LOR for all triplicates except those samples taken in the first weeks for "B. Concrete + soil under". Chromium was thus primarily present as Cr(VI) in almost all samples. "B. Concrete + soil under" differs from the other triplicates as it had several relative standard deviations ≥ 30 %, especially for total Cr and Cr(VI).

Table 7: pH, conductivity ($K_{25°C}$) and concentrations of total Cr, Cr(III), Cr(VI) and DOC in water eluate sampled at various L/S ratios based on triplicates. Light blue colour indicates concentrations below limit of reporting (LOR) for all three columns in a triplicate. Dark blue colour indicates that one or two columns is < LOR. When results were below the LOR, shown with < symbol in the analysis reports, < was removed, and the number divided by 2. Yellow colour indicates relative standard deviation ≥ 30 %.

Name	No.	L/S ratio	pH	$K_{25°C}$ mS cm⁻¹		Total Cr µg L⁻¹		Cr(III) µg L⁻¹		Cr(VI) µg L⁻¹		DOC mg L⁻¹	
		Mean	Mean	Mean	Std. dev.	Mean	Std. dev.	Mean	Std. dev.	Mean	Std. dev.	Mean	Std. dev.
A. Concrete + sand under	1, 2, 3	0.1	12.4	9.3	1.1	48	17	1.0	0	48	18	105	35
		0.7	12.6	8.1	0.1	22	1.7	1.0	0	23	1.8	24	1.6
		1.3	12.5	6.6	0.1	25	2.8	1.0	0	26	2.2	4.1	0.3
		1.8	12.5	6.3	0.2	21	2.2	1.0	0	22	2.7	1.3	0.0
		2.5	12.5	6.7	0.1	19	1.1	1.0	0	19	2.4	1.3	0.0
		3.8	12.4	6.5	0.3	17	1.8	1.0	0	19	1.8	1.5	0.4
		5.2	12.5	6.5	0.2	16	1.7	1.0	0	19	1.0	1.4	0.1
		6.4	12.5	6.2	0.4	15	0.8	1.0	0	16	1.0	1.8	0.9
		8.9	12.4	6.0	0.9	16	3.1	1.0	0	18	3.4	1.1	0.1
		10.4	12.4	5.1	0.8	18	4.9	1.0	0	20	4.4	0.8	0.1
		12.6	12.1	4.0	0.4	23	2.8	1.0	0	25	2.4	1.1	0.2
B. Concrete + soil under	4, 5, 6	0.1	4.4	1.1	0.1	41	20	41	20	0.8	1.0	150	65
		0.9	6.1	0.9	0.0	20	3.8	20	3.8	2.0	0.0	768	101
		1.6	11.3	2.4	1.1	21	4.4	21	4.4	2.0	0.0	373	83
		2.3	12.1	3.9	1.8	7.6	8.1	4.4	5.9	3.8	1.7	153	131
		3.1	12.4	5.2	0.6	3.8	0.3	1.0	0	4.5	0.2	92	24
		4.6	12.4	6.2	0.1	3.9	0.4	1.0	0	3.0	0.9	46	8.5
		6.1	12.5	5.9	0.8	2.6	2.7	1.0	0	3.0	1.2	36	4.1
		7.4	12.4	5.4	1.1	7.0	7.9	5.2	7.2	2.6	1.4	29	2.6
		10.0	12.4	5.5	1.3	3.6	2.4	1.0	0	4.4	3.0	18	3.1
		10.8	12.3	4.8	1.4	3.3	2.1	1.0	0	3.6	2.3	18	3.1
		14.2	12.1	4.0	1.0	2.7	2.9	1.0	0	3.5	3.8	15	1.5
C. Concrete + sand over	7, 8, 9	0.1	12.4	13	0.4	44	0.6	1.0	0	43	2.0	94	4.9
		0.6	12.6	8.5	0.2	23	2.4	1.5	0.8	22	0.9	28	4.7
		1.1	12.5	6.6	0.4	20	1.6	1.4	0.8	20	0.4	6.0	1.5
		1.6	12.5	6.3	0.5	17	1.3	1.0	0.0	18	1.7	3.1	0.7
		2.3	12.5	6.8	0.3	14	1.2	1.0	0	16	0.6	1.7	0.8
		3.5	12.5	6.6	0.2	13	0.1	1.0	0	16	0.4	1.9	0.3
		4.8	12.5	6.7	0.0	13	0.8	1.0	0	16	1.1	1.4	0.0
		6.0	12.5	6.4	0.3	13	0.6	1.0	0	13	0.7	1.3	0.0
		8.5	12.4	5.6	0.5	18	4.6	1.0	0	19	3.1	1.2	0.1
		10.4	12.4	4.7	0.4	25	1.4	1.0	0	26	0.5	1.4	0.5
		12.2	12.1	4.0	0.3	29	0.8	1.0	0	31	0.6	1.3	0.5
D. Concrete + soil over	10, 11, 12	0.1	12.5	14	1.4	52	7.6	1.0	0	53	7.0	105	17
		0.8	12.6	8.6	0.2	24	3.1	2.4	2.4	22	0.9	27	3.1
		1.6	12.4	6.8	0.3	18	0.4	1.0	0	20	0.9	5.7	0.1
		2.4	12.5	6.8	0.2	15	0.5	1.0	0	16	0.5	2.4	2.0
		3.1	12.5	6.4	0.4	15	2.5	1.0	0	17	2.0	2.2	1.7
		4.8	12.5	6.8	0.3	13	0.9	1.0	0	15	1.5	2.2	0.5
		6.6	12.5	6.8	0.2	12	0.8	1.0	0	14	0.3	1.6	0.4
		8.2	12.5	6.5	0.5	11	1.4	1.0	0	11	1.7	1.3	0.0
		10.8	12.4	6.0	0.6	13	2.6	1.0	0	16	3.6	1.7	0.3
		15.2	12.2	4.3	0.5	22	13	1.0	0	24	14	1.3	0.1

Table 8 show the results for the analysed parameters for the blanks water eluate at the respective L/S ratios. Concentrations < LOR are marked. For "Concrete" chromium was present mainly as Cr(VI). The Cr(III) concentration for "Concrete" was < LOR for all samples taken except at L/S 0.1 and for two slight outliers at L/S 1.2 and L/S 10.5. The concentration of Cr(VI) was < LOR during the whole test for "Sand" and "Soil". And the concentration of Cr(III) was < LOR, except at L/S 0.1 and L/S 2 for "Sand" and at L/S 2.4 to 40.3 for "Soil".

Table 8: pH, conductivity ($K_{25°C}$) and concentrations of total Cr, Cr(III), Cr(VI) and DOC for the water eluate sampled at the respective L/S ratios for the blanks. Light blue colour indicates concentrations < LOR. When results were below the LOR, shown with < symbol in the analysis reports, < was removed, and the number divided by 2.

Name	No.	L/S ratio	pH	$K_{25°C}$ mS cm^{-1}	Total Cr µg L^{-1}	Cr(III) µg L^{-1}	Cr(VI) µg L^{-1}	DOC mg L^{-1}
Concrete	13	0.1	12.5	15	62	5.8	56	120
		0.5	12.6	9.2	27	1.0	29	32
		1.2	12.5	7.1	20	2.2	18	5.9
		1.8	12.5	6.8	17	1.0	17	3.0
		2.4	12.5	6.7	16	1.0	18	1.3
		3.5	12.5	6.7	14	1.0	17	2.2
		4.7	12.4	6.9	14	1.0	16	1.4
		6.1	12.5	7.0	13	1.0	14	1.3
		8.6	12.5	7.1	13	1.0	16	1.5
		10.5	12.6	6.9	14	2.6	11	1.2
		12.2	12.3	6.5	13	1.0	18	1.5
Sand	14	0.1	7.3	1.1	9.7	9.7	0.2	53
		0.7	6.9	0.2	1.0	1.0	0.2	3.4
		1.3	9.3	0.03	1.0	1.0	0.2	3.1
		2.0	8.4	0.01	13	13	0.2	2.2
		2.7	9.5	0.01	1.0	1.0	0.2	1.0
		4.0	8.8	0.01	1.0	1.0	0.2	1.6
		5.4	8.2	0.01	1.0	1.0	0.2	1.1
		6.6	8.9	0.01	1.0	1.0	0.2	1.0
		8.9	8.8	0.01	1.0	1.0	0.2	0.6
		10.0	8.1	0.01	1.0	1.0	0.2	0.3
		12.5	9.4	0.01	1.0	1.0	0.2	0.5
Soil	15	2.4	7.1	1.3	6.9	6.9	0.2	71
		18.0	4.9	0.06	7.5	7.5	0.2	57
		34.0	8.5	0.03	2.4	2.4	0.2	36
		40.3	5.7	0.05	8.4	8.4	0.2	32
		46.6	5.7	0.02	1.0	1.0	0.2	18
		65.0	8.5	0.02	1.0	1.0	0.2	19
		100.3	7.8	0.02	1.0	1.0	0.2	9.5
		133.5	7.8	0.01	1.0	1.0	0.2	14
		168.4	7.2	0.01	1.0	1.0	0.2	10
		230.8	6.8	0.01	1.0	1.0	0.2	4.6
		316.8	6.3	0.01	1.0	1.0	0.2	4.1

As shown in Figure 7 and Table 7 the water eluate from "A. Concrete + sand under", "C. Concrete + sand over" and "D. Concrete + soil over" had a high pH of about 12.5 at L/S 0.1, with a decrease in pH over time, reaching pH ≈ 12 at the end the test period. For triplicate "B. Concrete + soil under" the pH was low, 4.4 – 6.1, during the initial phase, L/S 0.1 – 0.9, before increasing to the same pH-levels as the other triplicates from L/S 3.1.

The Cr(VI) concentration (Figure 7) (Table 7) in the water eluate from triplicate "A. Concrete + sand under", "C. Concrete + sand over" and "D. Concrete + soil over" showed similar patterns over time with high concentrations at L/S 0.1 (43 – 53 µg L^{-1}) followed by a large decrease at L/S 0.6 – 0.8, a minor decrease at L/S 6 – 8 followed by an increase at the end of the test (24 to 31 µg L^{-1}). The increase in concentration after L/S 6 – 8 corresponded with the change in the amount of water added, from 500 mL/week to 1000 mL/week (Table 6). The Cr(III) concentrations in the same triplicates (Figure 7) (Table 7) were < LOR during the 4 month test period, except for some small outliers in the early part for "C." and "D.".

The Cr(VI) concentrations in the water eluate from "B. Concrete + soil under" showed a completely different pattern over time (Figure 7) (Table 7), with concentrations < LOR from L/S 0.1 to L/S 1.6, followed by an increase in concentration to 3.8 µg L^{-1} at L/S 2.3, then mainly decreasing to 2.6 µg L^{-1} at L/S 7.4, then increasing at first (4.4 µg L^{-1}) and then decreasing to 3.5 µg L^{-1} at the end of the test period. The increase after L/S 7.4 corresponds with the increase in amount of water added from 500 mL/week to 1000 mL/week. The Cr(III) concentration (Figure 7) (Table 7) in the same triplicate was high at L/S 0.1 (41 µg L^{-1}) followed by a large decrease at L/S 2.3 before being < LOR throughout the remaining test period, except for one unexplained outlier (5.2 µg L^{-1} at L/S 7.4) probably due to analytical reasons.
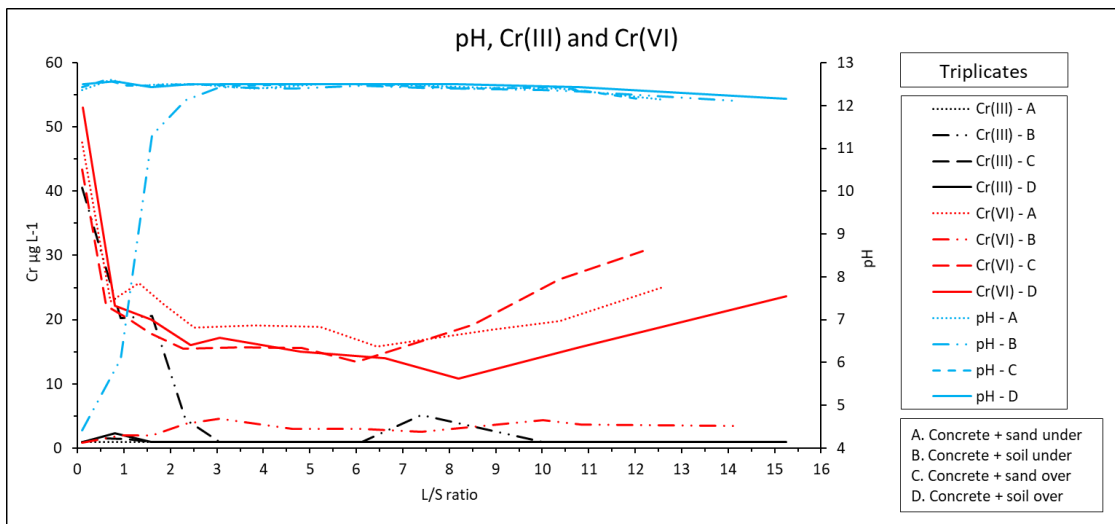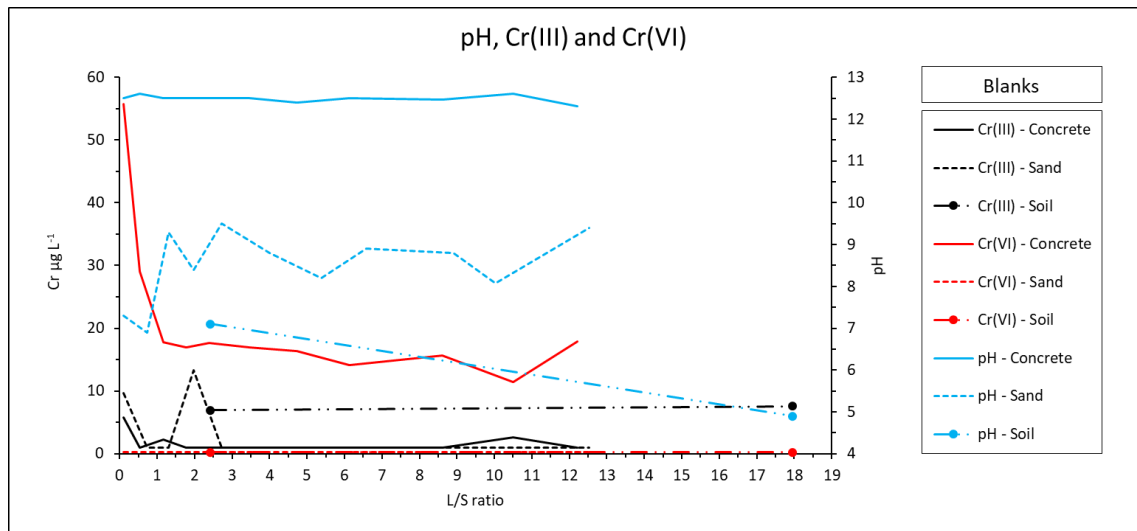
Figure 7: pH and concentrations of Cr(III) and Cr(VI) at the respective L/S ratios for the 4 triplicates, A, B, C and D.

As shown in Figure 8 and Table 8 the pH was about 12.5 in the water eluate during the total test period for the blank "Concrete". In the water eluate from the blank "Sand", pH fluctuated between 7.3 to 9.4 throughout the test, while pH in the water eluate from the blank "Soil" decreased from pH 7.1 at L/S 2.4 to pH 4.9 at L/S 18.

The Cr(VI) concentration (Figure 8) (Table 8) in the water eluate from the blank "Concrete" was high at L/S 0.1 (56 µg L^{-1}) followed by a large decrease to 18 µg L^{-1} (L/S 1.2), then slightly decreasing with fluctuations to 11 µg L^{-1} (L/S 10.5), then increasing to 18 µg L^{-1} at the end of the test. The Cr(VI) concentration for "Concrete" increased with fluctuations when the amount of added water was increased from 500 mL/week to 1000 mL/week after L/S 6.1. At L/S 0.1 the Cr(III) concentration was 5.8 µg L^{-1} in the water eluate from "Concrete", then decreasing to < LOR, remaining there throughout the test, except for some outliers probably due to analytical reasons. The Cr(VI) concentration was < LOR in the water eluate from both the blanks "Sand" and "Soil" (Figure 8) (Table 8) during the test period. The Cr(III) concentration in the water eluate from "Sand" followed the same pattern as for "Concrete" with a concentration of 9.7 µg L^{-1} at L/S 0.1 before decreasing to < LOR from L/S 0.7 and remained there except from one outlier at L/S 2.0 (13 µg L^{-1}) probably due to analytical reasons. For the blank "Soil" the Cr(III) concentration

in the water eluate was relatively constant, i.e. 6.9 µg L^{-1} at L/S 2.4 and 7.5 µg L^{-1} at L/S 18.

Figure 8: pH and concentrations of Cr(III) and Cr(VI) at the respective L/S ratios for the 3 blanks, "Concrete", "Sand" and "Soil". For "Soil" the results are shown in the graph only for L/S 2.4 and L/S 18 caused by the deviation in L/S ratio compared to the other blanks. Since L/S 18 is much higher than maximum ratios for the other blanks the lines for "Soil" span a greater L/S ratio.

The results for the triplicate concentrations of DOC in the water eluate is shown in Figure 9 and Table 7. The DOC concentrations for triplicate "A. Concrete + sand under", "C. Concrete + sand over" and "D. Concrete + soil over" showed similar patterns over time for the concentrations of DOC, from ≈ 100 mg L^{-1} at L/S 0.1, with a significant decrease to 1 – 2 mg L^{-1} at L/S 2 - 3 and remained at this level to the end of the test period. For triplicate "B. Concrete + soil under" the DOC concentration followed a very different pattern with a concentration of 150 mg L^{-1} at L/S 0.1 followed by significant increase to a peak with concentration 768 mg L^{-1} at L/S 0.9, before decreasing steeply to 153 mg L^{-1} at L/S 2.3 followed by a slight decrease to 15 mg L^{-1} at the end of the test period.
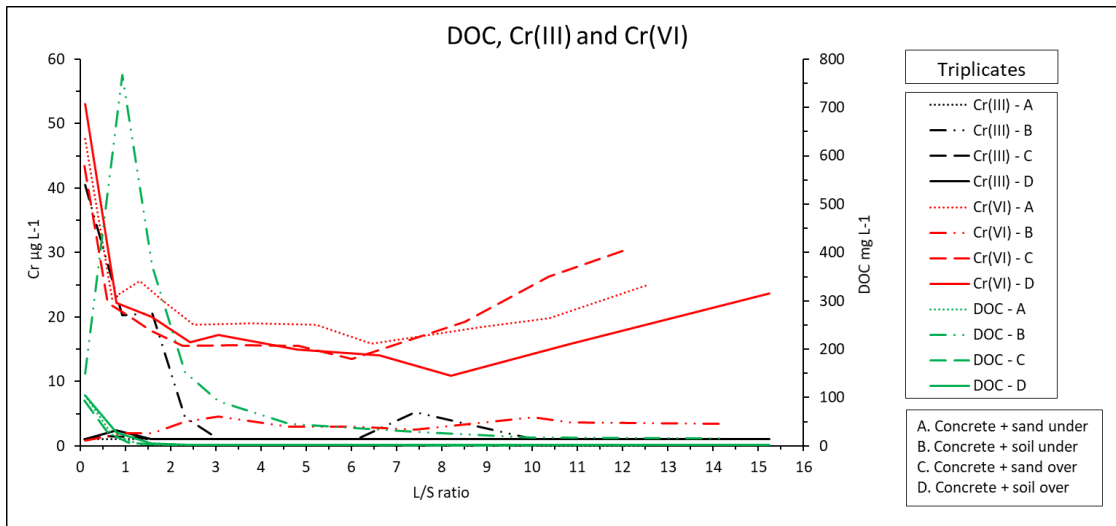
Figure 9: Concentrations of DOC, Cr(III) and Cr(VI) at the respective L/S ratios for the 4 triplicates A, B, C and D.

The results for the blank concentrations of DOC in the water eluate is shown in Figure 10 and Table 8. For the blank "Concrete" the DOC concentration in the water eluate was 120 mg L^{-1} at L/S 0.1, before decreasing significantly to 5.9 mg L^{-1} at L/S 1.2 and a smaller decrease down to 1.5 mg L^{-1} at the end of the test. The DOC concentration in the water eluate for the blank "Sand" was 53 mg L^{-1} at L/S 0.1, before decreasing significantly to 3.4 mg L^{-1} at L/S 0.7 and a further decrease to 0.5 mg L^{-1} at the end of the test. The blank "Soil" showed a relatively stable DOC pattern by time, i.e. 71 mg L^{-1} at L/S 2.4 and 57 mg L^{-1} at L/S 18.
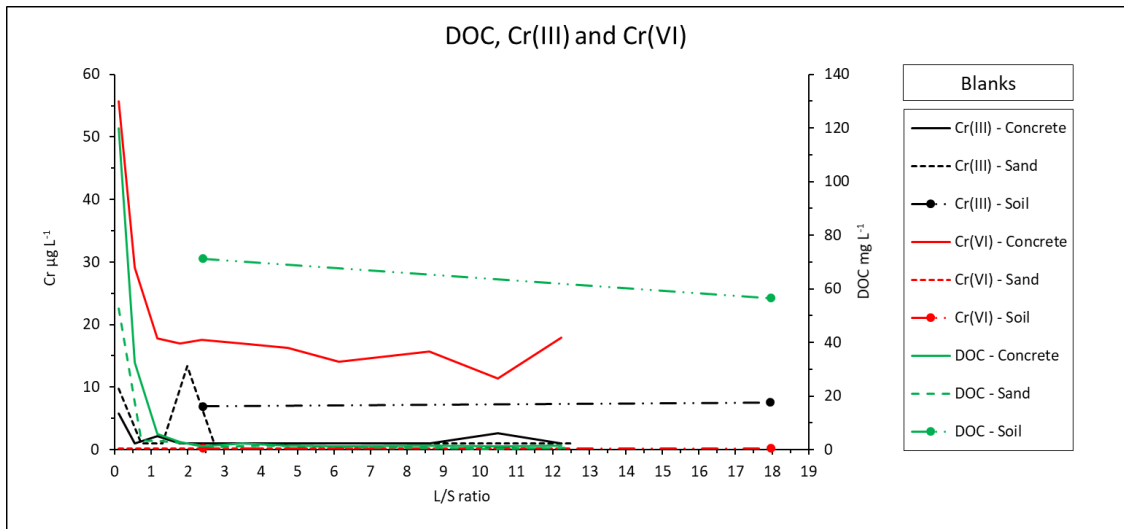
Figure 10: Concentrations of DOC, Cr(III) and Cr(VI) at the respective L/S ratios for the 3 blanks, "Concrete", "Sand" and "Soil". For "Soil" the results are shown in the graph only for L/S 2.4 and L/S 18 caused by the deviation in L/S ratio compared to the other blanks. Since L/S 18 is much higher than maximum ratios for the other blanks the lines for "Soil" span a greater L/S ratio.

The conductivity values ($K_{25°C}$) in water eluates from the different triplicates are shown in Figure 11 and Table 7. The conductivity in the water eluate for "A. Concrete + sand under", "C. Concrete + sand over" and "D. Concrete + soil over" followed almost similar patterns over time. The conductivity began high with 13 and 14 mS cm^{-1} for "C." and "D." and lower for "A." with 9.3 mS cm^{-1}, before decreasing to 6 - 7 mS cm^{-1} at L/S 1 − 2, remained almost constant for a certain period before slightly decreasing to ≈ 4 mS cm^{-1} at the end of the test period. For "B. Concrete + soil under" the initial conductivity, i.e. at L/S 0.1, was much lower (1.1 mS cm^{-1}) than for the other 3 triplicates. The conductivity remained constant from L/S 0.1 to L/S 0.9 before increasing to 6.2 mS cm^{-1} at L/S 4.6, and thereafter following almost the same pattern over time as the three other triplicates.

Figure 11: Conductivity ($K_{25°C}$) and concentrations of Cr(III) and Cr(VI) at the respective L/S ratios for the 4 triplicates, A, B, C and D.

The conductivity ($K_{25°C}$) in the water eluate from the blanks is shown in Figure 12 and Table 8. The conductivity for the blank "Concrete" was 15 mS cm^{-1} at L/S 0.1, decreasing to 7.1 mS cm^{-1} at L/S 1.2 and remained in the range 6.5 – 7.0 mS cm^{-1} to the end of the test period. As for the blank "Concrete" the blank "Sand" showed an elevated conductivity at L/S 0.1 but much less, 1.1 mS cm^{-1}. Thereafter the conductivity decreased significantly to 0.01 mS cm^{-1} at L/S 2.0 and then remained at that level to the end of the test period. For "Soil" the conductivity was 1.3 mS cm^{-1} at L/S 2.4, i.e. almost at the same level as "Sand", before decreasing to 0.06 mS cm^{-1} at L/S 18.

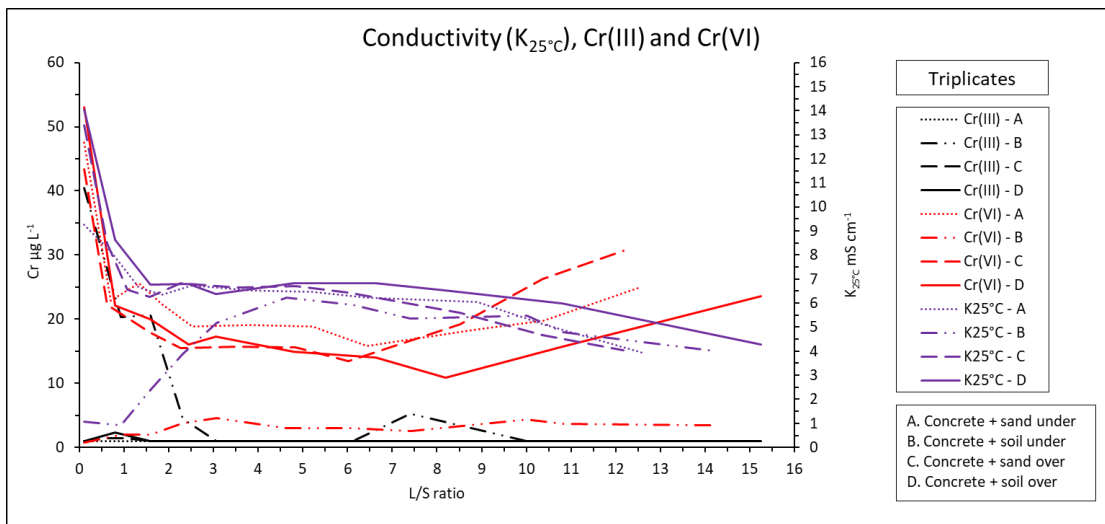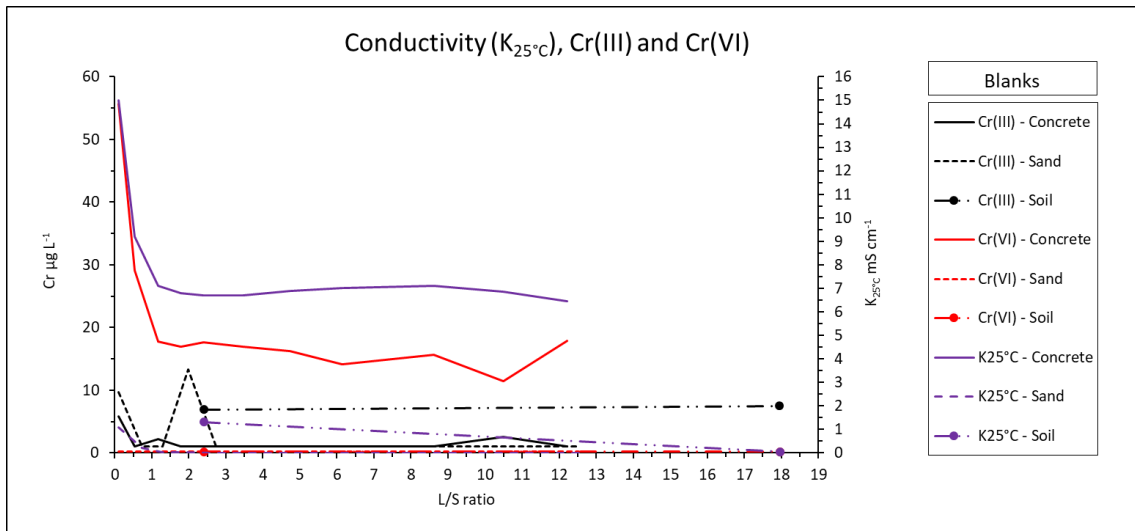*Figure 12: Conductivity ($K_{25°C}$) and concentrations of Cr(III) and Cr(VI) at the respective L/S ratios for the 3 blanks, "Concrete", "Sand" and "Soil". For "Soil" the results are shown in the graph only for L/S 2.4 and L/S 18 caused by the deviation in L/S ratio compared to the other blanks. Since L/S 18 is much higher than maximum ratios for the other blanks the lines for "Soil" span a greater L/S ratio.*

Annex 4 shows the cumulatively released quantities of total Cr, Cr(III), Cr(VI) and DOC in mg kg^{-1} dw (Ch. 2.8, formula 3) for the triplicates and blanks. Cumulatively released quantities of Cr(VI) for the triplicates and the blank "Concrete" are shown in Figure 13. Triplicate "A. Concrete + sand under", "C. Concrete + sand over", "D. Concrete + soil over" and the blank "Concrete" followed the same pattern over time from the start with a relatively large increase in cumulatively release of Cr(VI) from L/S 0.1 up to L/S 2 - 3. From there, the increase was less steep to L/S 8 − 9. After L/S 8 − 9, which correspond with increase in amount of added water from 500 mL/week to 1000 mL/week, it was a significant increase for "C.", a significant lower increase for "A." and "Concrete" and negligible increase for "D.". "B. Concrete + soil under" followed the same pattern as "D." but with much lower levels. The total cumulatively release of Cr(VI) were 0.17 ("A."), 0.03 ("B."), 0.18 ("C."), 0.15 ("D.") and 0.14 ("Concrete") mg Cr(VI) kg^{-1} dw (Annex 4).
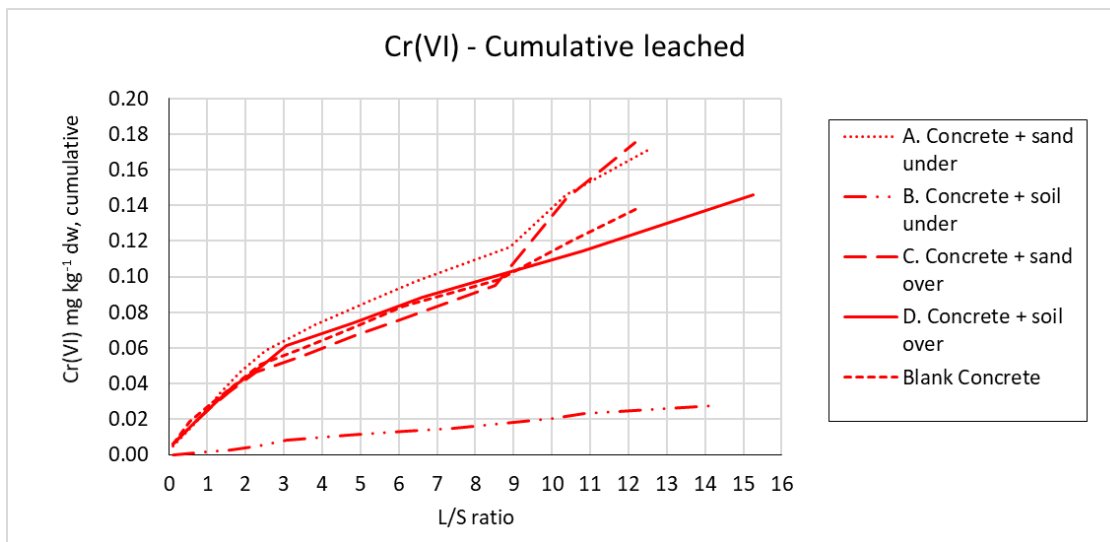
Figure 13: Cumulatively released quantities of Cr(VI) in mg kg⁻¹ dw at the respective L/S ratios for the triplicates A, B, C, D and for the blank Concrete.

In the water eluate (Figure 14) from all the columns, except from the blanks "Sand" and "Soil", the formation of a film was observed which latter formed into flakes, before precipitating to the bottom of the sampling bottles. A coating was also formed on the walls of the bottles. When the water eluate was put in a beaker a film was formed spontaneously (Figure 14). Analysis result of the precipitate shows a total Cr concentration of 1.4 µg g⁻¹ $CaCO_3$. $CaCO_3$ was not analysed but assumed to be the main precipitate.

Figure 14: A film (right) was observed on top of the water eluate bottles (except for the blanks "Sand" and "Soil") which formed flakes before precipitating to the bottom of the bottles. A coating was also formed inside the bottle walls. A film (left) was formed spontaneously when the water eluate was transferred to a beaker.

4 Discussion

4.1 Description of solid materials

The concrete (RCA) used in the column test was dated to be from 1969. Eckbo (2020a) reported ages between 1940 to 1980 for RCA from 104 different demolitions projects in Norway. Thus, the age of our RCA is within the same period.

The Cr(VI) concentration in our RCA was 4100 µg kg^{-1} (Table 5), and thus very comparable with Eckbo (2020a) who analysed 58 RCA samples from demolition projects in Norway and found an average concentration of 5200 µg kg^{-1} Cr(VI) with a standard deviation of 5500 µg kg^{-1}.

4.2 Speciation of chromium in the RCA leachate

In the water eluate from the blank "Concrete", chromium was present mainly as Cr(VI) and with a pH of about 12.5 (Figure 8). This result supports hypothesis a, *"Most of the chromium that leaches from the RCA will be in the hexavalent form (Cr-VI)",* and is due to the higher solubility of Cr(VI) than Cr(III) in leachate from alkaline wastes (Cornelis et al., 2008; Serclérat et al., 2000). Eckbo et al. (2022) presented similar findings for RCA leachate in a similar column test set up (NS-EN 14405:2017). In the leachate from five different RCA, they found that Cr(VI) made up an average of 63, 93 and 96 % of the total chromium content at L/S 0.1, 2 and 10 respectively. pH in the water eluates was > 12. The authors also report batch shake (NS-EN 12457-2, 2002) and layered leaching tests (top-down leaching, unsaturated), where they reported chromium mainly present as Cr(VI) (water eluate pH > 12) in RCA eluates. Serclérat et al. (2000) reported similar findings in leaching tests on mortar bars where chromium was in the hexavalent form as chromate (CrO_4^{-2}) in the leachate. Cr(III) was found to not leach because it was bound in the mortars. Butera et al. (2015a) also presented similar findings in an up-flow column test (similar as here) and a down-flow lysimeter test on RCA. They found that the chromium in the water eluate was mainly present as Cr(VI) and pH in the range 12 – 13.

The total leached amount of Cr(VI) from the blank "Concrete" was 140 µg kg^{-1} dw (Annex 4). The Cr(VI) content in the solid concrete (RCA) was 4100 (Table 5), meaning that only 3.4 percent of Cr(VI) leached out during the column test. The result agrees with Eckbo et al. (2022) who found a low amount (7 %) of Cr(VI) in the solid RCA had leached out during the experimental period. Our result shows that Cr(VI) in RCA represent a long-term source of Cr(VI) leaching, also concluded by Eckbo et al. (2022).

The observed precipitate in the eluate bottles (Figure 14) from all the columns except from the blanks "Sand" and "Soil" is probably caused by a carbonation process. Carbonation occurs when CO_2 dissolves in concrete pore water and the carbonate species H_2CO_3/HCO_3^-/CO_3^{2-} are formed. CO_3^{2-} reacts with Ca^{2+} and $CaCO_3$ is precipitated (Engelsen et al., 2012b). When Butera et al. (2015a) conducted a similar column test as in our study, using demolition waste (also RCA), they covered the water eluate bottles with a plastic film to reduce the effect of carbonation. In our test he bottles were not covered and therefore exposed to carbonation. The concentration of total Cr in the precipitate was found to be 1.4 µg g^{-1} $CaCO_3$. $CaCO_3$ was not analyzed but assumed to be the main precipitate.

4.3 The effect of SOM on the leaching of Cr(VI) from the RCA

Initially (L/S 0.1 to L/S 1.6) 100 % of the Cr(VI) in the water eluate from triplicate "B. Concrete + soil under" was reduced to Cr(III) (Figure 7). Based on this, hypothesis b, "*The concentration of Cr(VI) in the leachate water will decrease when the RCA is in contact with SOM",* was verified when RCA leachate percolated SOM. The result is in agreement with Bartlett and Kimble (1976) and Kožuh et al. (2000) who observed rapid reduction of Cr(VI) to Cr(III) in solutions with SOM. Eckbo et al. (2022) presented similar findings in a down-flow column test as Cr(VI) concentrations were mainly < LOQ when RCA leachate had percolated soils with either 3.2 or 5.3 % TOC. Butera et al. (2015b) also found Cr(VI) retention when RCA leachate was mixed in a solution with soils in a batch test. The retention efficiency varied between 30 to 97 %. The SOM content in the soils was 0.3 − 0.4 %. Similar findings were also shown in two large scale field tests at Jølsen Miljøpark and Lindum Oredalen (Eckbo, 2020b). At Jølsen Miljøpark the RCA leachate percolated soil with 15 % TOC. In the first 2 months (test period of 4 months) chromium was found

in the water eluate mainly as Cr(VI). In the last 2 months Cr(VI) was < LOR. The leaching of Cr(VI) in the first two months was explained by reduced contact between the RCA leachate and the soil at the beginning of the test. At Lindum Oredalen (15 weeks test period), RCA leachate percolated soils with either $0.8 - 1.4$ % (low TOC), $2.7 - 3.2$ % (medium TOC) or $6.0 - 7.1$ % (high TOC). The Cr(VI) reduction was about 95 % for the soils with medium and high TOC contents and about 85 % for the soil with low TOC content. Thus, numerous reports document significant effects of TOC (or SOM) on reduction of Cr(VI) to Cr(III).

After running the test for longer time period (i.e. after L/S 1.6) for the set-up "B. Concrete + soil under" (Figure 7), the reduction of Cr(VI) to Cr(III) decreased to approximately 85 %. The decrease corresponded with elevated leaching of DOC (Figure 9) and increase in pH (> 12) (Figure 7). This shows that the DOC leaching lowered the SOM capacity to reduce Cr(VI) to Cr(III). The increase in pH probably also contributed to the elevated leaching of Cr(VI), as release of Cr(VI) anions is pH depend, i.e. highest release at high pH (Van der Sloot & Kosson, 2010). The elevated leaching of DOC for "B. Concrete + soil under"(Figure 7) can be explained by that humic and fulvic acids neutralized the strong base $Ca(OH)_2$ (Engelsen et al., 2012a) in the RCA leachate by emitting H^+ ions (Lydersen et al., 2002). This deprotonation of organic acids (release of H^+ ions to the water) increased the water solubility of soil organic matter. The significant deprotonation of organic acids caused by the high pH in the RCA leachate exceeded the pH buffering capacity of the SOM present, and pH increased from 4.4 to > 12 after a relatively short time.

The total leached amount of DOC from "B. Concrete + soil under" during the column test was 1088 mg kg^{-1} dw and the soils dry weight was 7 gram in "B. Concrete + soil under" (Annex 4). When adjusting against the soils dry weight in "B." the total leached amount was 104 000 mg kg^{-1} dw (Annex 4). The TOC amount in the soil was 23 % of dw (Table 5) or 223 000 mg kg^{-1} dw, which means that about 50 % of SOM in "B." leached out during the column test. This shows that peat can be vulnerable in contact with RCA where lowering in the reduction capacity of Cr(VI) might occur when DOC is leaving out of the soil system. Eckbo et al. (2022) did not observe elevated leaching of DOC in the down

flow column test with RCA and two soils (3.2 or 5.3 % TOC). Their soils had higher density (about 1 kg/dm^3) and 80 % dry weight. The soil in our test had a density of 0.20 kg/dm^3 and 22 % dry weight (Table 5). Moreover Eckbo et al. (2022) had a filling ratio of soil/concrete of 1 in the columns, whilst the ratio in our test was soil/concrete of 0.25. Those differences are probably a part of the explanation why Eckbo et al. (2022) did not observe elevated leaching of DOC as we did.

At the initial L/S ratios (L/S 0.1 to L/S 1.6) the reduced Cr(III) from Cr(VI) was not adsorbed in the soil for "B. Concrete + soil under" (Figure 7). This can be assumed to be because the Cr(III) pattern for "B." followed the Cr(VI) patterns for the three other triplicates, where no Cr(VI) reduction was observed (Figure 7), indicating that nearly all the Cr(VI) reduced to Cr(III) for "B." was soluble. This occurred at pH 4.4 − 6.1 (Table 7). Further in the test (after L/S 1.6) for "B.", Cr(III) was no longer found in the water eluate (Figure 7). The result shows that all the Cr(VI) reduced to Cr(III) was precipitated or adsorbed in the soil. This occurred at pH > 12 (Table 7). Eckbo et al. (2022) reported similar findings in their down flow test with RCA and two soils (3.2 or 5.3 % TOC). They found relatively high concentration of Cr(III) in water eluate with pH 4.4 − 6.3 from the columns with RCA and 5.3 % TOC soil, but no Cr(III) in the water eluate with pH 7.3 − 7.8 from the columns with RCA and 3.2 % TOC soil. Similar findings have earlier been published by James and Bartlett (1983) who observed that Cr(III) was soluble in water at pH up to pH 7 - 7.5 when complexed to organic ligands. Without organic ligands the Cr(III) precipitation started at pH 4.5 - 5.5.

The fact that all the chromium was present as Cr(VI) in the water eluate from "D. Concrete + soil over" (Figure 7) shows that the leachate water from the soil percolating the RCA has no effect on Cr(VI). Eckbo et al. (2022) found a slight increase in Cr(VI) reduction with increasing DOC concentrations (67 to 301 mg L^{-1}) in a static batch test, while Kožuh et al. (2000) observed no reduction of Cr(VI) by DOC. In our test no Cr(VI) reduction by DOC percolating the RCA leached from the soil in "D." (Figure 9) was found either. Not even at the elevated leaching of DOC for "B. Concrete + soil under" (Figure 9) it was observed an effect of Cr(VI) reduction by DOC. Based on this, hypothesis b, "*The concentration of*

Cr(VI) in the leachate water will decrease when the RCA is in contact with SOM", was not verified when SOM leachate percolated RCA.

The sand in the triplicates "A. Concrete + sand under" and "C. Concrete + sand over" had no effect on Cr(VI) reduction (Figure 7). This was expected due to the low organic content in sand, neutral pH in suspension (Table 5) and low cation exchange capacity (Foth, 1984). This was also in accordance with a previous leaching tests carried out by Eckbo (2019) where sand was mixed with RCA (Sand/concrete ratio – 20/80, L/S ratio - 10) carried out using end-over-end agitation. They found that the sand had no buffering capacity against the high pH resulting from the concrete in solution and thus no effect on the chromium chemistry.

4.4 The effect of simulated precipitation volumes on the leaching of Cr(VI) from the RCA

The elevated conductivity and concentrations of Cr(III), Cr(VI) and DOC at L/S 0.1, mainly followed by significant reductions, observed both for the triplicates and the blanks (Table 7) (Table 8), can be explained by a first flush effect. This result supports <u>hypothesis c</u>, *"The leaching of Cr(VI) from the RCA will increase with increasing simulated precipitation volumes".* The first flush effect is often explained by high initial concentrations of accumulated elements in the runoff from surfaces during intense rainfall events (Deletic, 1998). In the down-flow leaching test with RCA and two soils Eckbo et al. (2022) also observed a similar first flush effect.

After the amount of added water was increased from 500 mL/week to 1000 mL/week (Table 6), it was an increase in Cr(VI) concentrations in the water eluate from the 3 triplicates "A. Concrete + sand under", "C. Concrete + sand over", "D. Concrete + soil over" (Figure 7) (Figure 13) and the blank "Concrete" (Figure 8) (Figure 13). The result supports <u>hypothesis c</u>, *"The leaching of Cr(VI) from the RCA will increase with increasing simulated precipitation volumes".* The result is supported by Butera et al. (2015b) which also found increased leaching of Cr(VI) from RCA with increased amounts of water percolating the RCA. Takahashi et al. (2007) reported similar findings with increase in

accumulated leached amounts of chromium with increased L/S ratios in tank leaching experiments on mortar. They suggested that increased concentrations of chromium in the water eluate restricted further diffusion of chromium from the mortar. Also for "B. Concrete + soil under" (Figure 7) (Figure 13) there was an increase in Cr(VI) concentration when the amount of water added was increased from 500 mL/week to 1000 mL/week (Table 6), but unlike the other triplicates and the blank "Concrete" the increase was followed by an decrease against the end for the test period. This, together with much lower Cr(VI) concentrations for "B." than for other triplicates and the blank "Concrete", the increase in Cr(VI) concentration because of elevated amounts of water added was negligible. The result shows that elevated Cr(VI) leaching from RCA because of increased amounts of water added will be buffered (reduced to (Cr-III)) when RCA leachate percolate SOM. The triplicates "A. Concrete + sand under" and "C. Concrete + sand over" had the highest total cumulatively release of Cr(VI), 0.17 and 0.18 mg kg^{-1} dw, respectively, also higher than for the blank "Concrete", 0.14 mg kg^{-1} dw (Figure 13) (Annex 4). Differences in permeability between sand, soil and RCA and amounts of water added to the columns may be a part of the explanation of why "A." and "C." had the highest total cumulatively release of Cr(VI).

A slight decrease in pH from 12.5 to about 12 at the end of the test was observed for all the triplicates, also for "B. Concrete + soil under" after the pH had reached the same level as the others (Figure 7). The decrease corresponded to some extent with a slight decrease in conductivity ($K_{25°C}$) against the end of the test period (Figure 11). This can be explained by decreasing concentration of Ca^{2+} dissolved from solid Ca-compounds by time, and subsequent reduction in OH^- concentrations, identified by the decrease in pH from pH 12.5 to 12.0. The same effect was observed for the blank "Concrete" but here was both the decrease in pH and conductivity ($K_{25°C}$) lower (Figure 8) (Figure 12).

5 Conclusion

The results showed that most of the chromium present in recycled concrete aggregates (RCA) will leach out as Cr(VI) if no countermeasures were implemented. In addition, the result shows that Cr(VI) in RCA presents a long-term source of Cr(VI) leaching. The only countermeasure tested which achieved significant Cr(VI) reduction to Cr(III) was shown to occur when simulated precipitation first percolated RCA before percolating soil organic matter (SOM), while no reduction occurred when simulated precipitation first percolated SOM before percolating RCA.

The results showed that elevated leaching of Cr(VI) because of increased simulated precipitation percolating the RCA, will be buffered (reduced to (Cr-III)) when the RCA leachate percolating SOM.

To avoid leaching of Cr(III) from the reactions described above, the results indicates that soil water pH should be above neutral or higher.

Future research should focus on qualitative and quantitative interaction between SOM and RCA, including differences in permeability or residence time of water in the different SOM and RCA layers, to optimize Cr(VI) reduction and the subsequent retention of Cr(III), in order to protect the external environment.

In a reuse scenario where a 0.5 m layer (density, 1.5 tonnes/m^3) of RCA is used in a construction, the L/S rations of 0.1, 2 and 10 represent about one month, one and half year and eight years of precipitation[6]. Under such scenarios it is imperative that measures are taken to avoid any negative effects on the environment because of leaching of chromium. Thus, such studies are highly important to meet the Norwegian goal of 70 % recycling of demolition waste.

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[6] The calculation is based on an average annual precipitation amount of 889 mm yr^{-1} (2016 – 2020) at Blindern weather station in Oslo (SN 18700) (Norsk Klimaservicesenter, 2022), where all the precipitation infiltrate through the RCA.

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Annexes

Annex 1 - Packing of materials in the columns

Table 9: Packing of materials in the 15 columns. Calculated weight before packing and actual weight after packing. Filling height of materials is also shown. The weights are based on wet weight.

						Calculated			Actual	
	Name	No.	Layers	Propor-tion	Height, cm	Volume, mL	Mass, g ml^{-1}	Wet weight, g	Wet weight, g	Height, cm
Triplicates	A. Concrete + sand under	1	Sand	1/4	7.5	147	1.44	212	215	8.0
			Concrete	3/4	22.5	442	1.55	685	695	22.6
			Sum		30.0	589		897	910	30.6
		2	Sand	1/4	7.5	147	1.44	212	215	7.7
			Concrete	3/4	22.5	442	1.55	685	689	22
			Sum		30.0	589		897	904	29.7
		3	Sand	1/4	7.5	147	1.44	212	215	8.1
			Concrete	3/4	22.5	442	1.55	685	691	22.2
			Sum		30.0	589		897	906	30.3
	B. Concrete + soil under	4	Soil	1/4	7.5	147	0.20	29	34	5.5
			Concrete	3/4	22.5	442	1.55	685	690	23
			Sum		30.0	589		714	724	28.5
		5	Soil	1/4	7.5	147	0.20	29	36	5.8
			Concrete	3/4	22.5	442	1.55	685	696	23
			Sum		30.0	589		714	732	28.8
		6	Soil	1/4	7.5	147	0.20	29	33	5.5
			Concrete	3/4	22.5	442	1.55	685	696	23.2
			Sum		30.0	589		714	729	28.7
	C. Concrete + sand over	7	Concrete	3/4	22.5	442	1.55	685	688	22.4
			Sand	1/4	7.5	147	1.44	212	216	8.3
			Sum		30.0	589		897	904	30.7
		8	Concrete	3/4	22.5	442	1.55	685	692	23
			Sand	1/4	7.5	147	1.44	212	214	8.1
			Sum		30.0	589		897	906	31.1
		9	Concrete	3/4	22.5	442	1.55	685	692	22.8
			Sand	1/4	7.5	147	1.44	212	213	8
			Sum		30.0	589		897	905	30.8
	D. Concrete + soil over	10	Concrete	3/4	22.5	442	1.55	685	686	22.4
			Soil	1/4	7.5	147	0.20	29	33	5
			Sum		30.0	589		714	719	27.4
		11	Concrete	3/4	22.5	442	1.55	685	684	22.6
			Soil	1/4	7.5	147	0.20	29	31	4.5
			Sum		30.0	589		714	715	27.1
		12	Concrete	3/4	22.5	442	1.55	685	684	22.5
			Soil	1/4	7.5	147	0.20	29	32	4.7
			Sum		30.0	589		714	716	27.2
Blanks	Concrete	13	Concrete	1	30.0	589	1.55	913	913	30.5
	Sand	14	Sand	1	30.0	589	1.44	848	850	31.2
	Soil	15	Soil	1	30.0	589	0.20	118	131	20.0

Annex 2 - Adjustment from wet weight to dry weight of the materials

Table 10: Adjustment from wet weight (ww) to dry weight (dw) of the materials in the 15 columns. The change in L/S ratio at L/S 0.1 and L/S 10 caused by the adjustment from ww to dw is shown.

	Name	No.	Layers	Wet weight (ww)	Dry weight (dw)	Dry weight (dw)	L/S 0.1			LS 10		
							Added water	L/S (ww)	L/S (dw)	Added water	L/S (ww)	L/S (dw)
				g	%	g	mL			mL		
Triplicates	A. Concrete + sand under	1	Sand	215	99.6	214						
			Concrete	695	95.7	665						
			Sum	910		879	91	0.10	0.10	9110	10.01	10.36
		2	Sand	215	99.6	214						
			Concrete	689	95.7	659						
			Sum	904		874	90	0.10	0.10	9127	10.10	10.45
		3	Sand	215	99.6	214						
			Concrete	691	95.7	661						
			Sum	906		875	93	0.10	0.11	9041	9.98	10.33
	B. Concrete + soil under	4	Soil	34	22.1	8						
			Concrete	690	95.7	660						
			Sum	724		668	71	0.10	0.11	7254	10.02	10.86
		5	Soil	36	22.1	8						
			Concrete	696	95.7	666						
			Sum	732		674	73	0.10	0.11	7308	9.98	10.84
		6	Soil	33	22.1	7						
			Concrete	696	95.7	666						
			Sum	729		673	70	0.10	0.10	7263	9.96	10.79
	C. Concrete + sand over	7	Concrete	688	95.7	658						
			Sand	216	99.6	215						
			Sum	904		874	86	0.10	0.10	9051	10.01	10.36
		8	Concrete	692	95.7	662						
			Sand	214	99.6	213						
			Sum	906		875	90	0.10	0.10	9098	10.04	10.39
		9	Concrete	692	95.7	662						
			Sand	213	99.6	212						
			Sum	905		874	90	0.10	0.10	9046	10.00	10.35
	D. Concrete + soil over	10	Concrete	686	95.7	657						
			Soil	33	22.1	7						
			Sum	719		664	71	0.10	0.11	7149	9.94	10.77
		11	Concrete	684	95.7	655						
			Soil	31	22.1	7						
			Sum	715		661	79	0.11	0.12	7101	9.93	10.74
		12	Concrete	684	95.7	655						
			Soil	32	22.1	7						
			Sum	716		662	70	0.10	0.11	7187	10.04	10.86
Blanks	Concrete	13	Concrete	913	95.7	874	94	0.10	0.11	9170	10.04	10.50
	Sand	14	Sand	850	99.6	847	86	0.10	0.10	8494	9.99	10.03
	Soil	15	Soil	131	22.1	29	70	0.53	2.42	1350	10.31	46.63

Annex 3 – L/S ratios based on dry weight of the materials

Table 11: (shown over two pages) Calculated L/S ratios at each sampling time (week) of the water eluate based on the dry weight (dw) of the materials (S) and accumulated weight of the water eluate (L) in the 15 columns. Shown with mean and standard deviation for the triplicates.

Page 1				Sampling week											
				(day 2)		1		2		3		4		6	
	Name	No.	Mater-ials (dw)	El-uate	L/S	El-uate	L/S	El-uate	L/S	El-uate	L/S	El-uate	L/S	El-uate	L/S
			g	g		g		g		g		g		g	
Triplicates	A. Concrete + sand under	1	879	91	0.1	649	0.7	1298	1.5	1803	2.1	2444	2.8	3347	3.8
		2	874	90	0.1	629	0.7	1129	1.3	1513	1.7	2182	2.5	3385	3.9
		3	875	93	0.1	585	0.7	1062	1.2	1547	1.8	1989	2.3	3280	3.7
	Mean				0.1		0.7		1.3		1.8		2.5		3.8
	Std. dev.				0.0		0.0		0.1		0.2		0.3		0.1
	B. Concrete + soil under	4	668	71	0.1	572	0.9	1096	1.6	1528	2.3	1971	3.0	2973	4.5
		5	674	73	0.1	542	0.8	827	1.2	1197	1.8	1842	2.7	3059	4.5
		6	673	70	0.1	759	1.1	1294	1.9	1949	2.9	2360	3.5	3301	4.9
	Mean				0.1		0.9		1.6		2.3		3.1		4.6
	Std. dev.				0.0		0.2		0.3		0.6		0.4		0.2
	C. Concrete + sand over	7	874	86	0.1	426	0.5	845	1.0	1121	1.3	1557	1.8	2860	3.3
		8	875	90	0.1	584	0.7	947	1.1	1392	1.6	2137	2.4	3106	3.5
		9	874	90	0.1	609	0.7	1035	1.2	1599	1.8	2264	2.6	3250	3.7
	Mean				0.1		0.6		1.1		1.6		2.3		3.5
	Std. dev.				0.0		0.1		0.1		0.3		0.4		0.2
	D. Concrete + soil over	10	664	71	0.1	506	0.8	1046	1.6	1622	2.4	1864	2.8	3344	5.0
		11	661	79	0.1	643	1.0	1072	1.6	1558	2.4	2117	3.2	3194	4.8
		12	662	70	0.1	440	0.7	1033	1.6	1670	2.5	2101	3.2	3002	4.5
	Mean				0.1		0.8		1.6		2.4		3.1		4.8
	Std. dev.				0.0		0.2		0.0		0.1		0.2		0.3
Blanks	Concrete	13	874	94	0.1	470	0.5	1022	1.2	1555	1.8	2088	2.4	3037	3.5
	Sand	14	847	86	0.1	621	0.7	1099	1.3	1674	2.0	2311	2.7	3395	4.0
	Soil	15	29	70	2.4	520	18.0	983	34.0	1350	46.6	1882	65.0	2904	100.3

	Name	No.	Materials (dw)	Sampling week								
				8		**10**		**12**		**14**		**16**	
				El-uate	L/S	El-uate	L/S	El-uate	L/S	El-uate	L/S	El-uate	L/S
			g	g		g		g		g		g	
Triplicates	A. Concrete + sand under	1	879	4489	5.1	5713	6.5	7983	9.1	9110	10.4	11113	12.6
		2	874	4757	5.4	5828	6.7	7873	9.0	9127	10.4	10826	12.4
		3	875	4464	5.1	5388	6.2	7543	8.6	9041	10.3	11110	12.7
	Mean				5.2		6.4		8.9		10.4		12.6
	Std. dev.				0.2		0.3		0.3		0.1		0.2
	B. Concrete + soil under	4	668	4061	6.1	5001	7.5	6850	10.3	7254	10.9	9576	14.3
		5	674	3987	5.9	4792	7.1	6525	9.7	7308	10.8	9598	14.2
		6	673	4284	6.4	5134	7.6	6812	10.1	7263	10.8	9465	14.1
	Mean				6.1		7.4		10.0		10.8		14.2
	Std. dev.				0.2		0.3		0.3		0.0		0.1
	C. Concrete + sand over	7	874	4216	4.8	5360	6.1	7576	8.7	9051	10.4	10704	12.3
		8	875	4189	4.8	5217	6.0	7269	8.3	9098	10.4	10574	12.1
		9	874	4287	4.9	5194	5.9	7488	8.6	9046	10.3	10638	12.2
	Mean				4.8		6.0		8.5		10.4		12.2
	Std. dev.				0.1		0.1		0.2		0.0		0.1
	D. Concrete + soil over	10	664	4780	7.2	5947	9.0	6631	10.0	7149	10.8	9795	14.8
		11	661	4331	6.5	5327	8.1	6397	9.7	7101	10.7	9856	14.9
		12	662	4050	6.1	5022	7.6	6379	9.6	7187	10.9	10638	16.1
	Mean				6.6		8.2		9.8		10.8		15.2
	Std. dev.				0.5		0.7		0.2		0.1		0.7
Blanks	Concrete	13	874	4142	4.7	5360	6.1	7526	8.6	9170	10.5	10678	12.2
	Sand	14	847	4553	5.4	5565	6.6	7537	8.9	8494	10.0	10606	12.5
	Soil	15	29	3866	133.5	4874	168.4	6681	230.8	6681	230.8	9173	316.8

Annex 4 – Cumulatively released quantities in mg kg⁻¹ dw

Table 12: Cumulatively released quantities of total Cr, Cr(III), Cr(VI) and DOC for the triplicates and the blanks in mg kg^{-1} dw at the respective L/S ratios.

Triplicate "A. Concrete + sand under" (mg kg^{-1} dw)

L/S ratio	Total Cr		Cr(III)		Cr(VI)		DOC	
	mg kg^{-1} dw	acc.	mg kg^{-1} dw	acc.	mg kg^{-1} dw	acc.	mg kg^{-1} dw	acc.
0.1	0.005	0.00	0.000	0.000	0.005	0.00	11	11
0.7	0.01	0.02	0.001	0.001	0.01	0.02	14	25
1.3	0.02	0.03	0.001	0.001	0.02	0.03	2.5	28
1.8	0.01	0.04	0.001	0.002	0.01	0.05	0.7	29
2.5	0.01	0.06	0.001	0.003	0.01	0.06	0.8	29
3.8	0.01	0.07	0.001	0.003	0.01	0.07	1.1	31
5.2	0.01	0.08	0.001	0.004	0.01	0.09	1.0	32
6.4	0.01	0.09	0.001	0.005	0.01	0.10	1.2	33
8.9	0.02	0.11	0.001	0.006	0.02	0.12	1.1	34
10.4	0.03	0.13	0.001	0.007	0.03	0.15	1.2	35
12.6	0.02	0.16	0.001	0.008	0.03	0.17	1.1	36

Triplicate "B. Concrete + soil under"

L/S ratio	Total Cr		Cr(III)		Cr(VI)		DOC	
	mg kg^{-1} dw	acc.	mg kg^{-1} dw	acc.	mg kg^{-1} dw	acc.	mg kg^{-1} dw	acc.
0.1	0.004	0.004	0.004	0.004	0.0001	0.00008	16	16
0.9	0.01	0.02	0.01	0.02	0.001	0.001	537	553
1.6	0.01	0.03	0.01	0.03	0.001	0.003	230	783
2.3	0.005	0.04	0.003	0.03	0.002	0.005	96	879
3.1	0.003	0.04	0.001	0.03	0.003	0.008	65	944
4.6	0.003	0.04	0.001	0.04	0.003	0.01	40	983
6.1	0.002	0.04	0.001	0.04	0.002	0.01	28	1011
7.4	0.005	0.05	0.004	0.04	0.002	0.02	21	1032
10.0	0.004	0.05	0.001	0.04	0.005	0.02	21	1053
10.8	0.003	0.06	0.001	0.04	0.003	0.02	14	1067
14.2	0.004	0.06	0.001	0.04	0.005	0.03	21	1088
							*	104393

*Accumulated amount of DOC leaching based on the dry weight of the soil alone

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Triplicate "C. Concrete + sand over"

L/S ratio	Total Cr		Cr(III)		Cr(VI)		DOC	
	mg kg^{-1} dw	acc.	mg kg^{-1} dw	acc.	mg kg^{-1} dw	acc.	mg kg^{-1} dw	acc.
0.1	0.01	0.01	0.0001	0.0001	0.01	0.01	13	13
0.6	0.01	0.02	0.001	0.001	0.01	0.02	14	27
1.1	0.01	0.03	0.001	0.002	0.01	0.03	2.8	30
1.6	0.01	0.04	0.000	0.002	0.01	0.04	1.5	31
2.3	0.01	0.05	0.001	0.003	0.01	0.05	1.2	32
3.5	0.01	0.05	0.001	0.003	0.01	0.06	1.2	34
4.8	0.01	0.06	0.001	0.004	0.01	0.07	1.0	35
6.0	0.01	0.07	0.001	0.005	0.01	0.08	0.8	35
8.5	0.02	0.09	0.001	0.006	0.02	0.10	1.3	37
10.4	0.05	0.1	0.002	0.008	0.05	0.14	2.7	39
12.2	0.03	0.2	0.001	0.009	0.03	0.18	1.4	41

Triplicate "D. Concrete + soil over"

L/S ratio	Total Cr		Cr(III)		Cr(VI)		DOC	
	mg kg^{-1} dw	acc.	mg kg^{-1} dw	acc.	mg kg^{-1} dw	acc.	mg kg^{-1} dw	acc.
0.1	0.01	0.01	0.000	0.000	0.01	0.01	12	12
0.8	0.02	0.02	0.002	0.002	0.02	0.02	19	30
1.6	0.01	0.04	0.001	0.003	0.02	0.04	4.5	35
2.4	0.01	0.05	0.001	0.003	0.01	0.05	2.1	37
3.1	0.01	0.06	0.001	0.004	0.01	0.06	1.4	38
4.8	0.01	0.07	0.001	0.005	0.01	0.07	1.9	40
6.6	0.01	0.08	0.001	0.006	0.01	0.09	1.6	42
8.2	0.01	0.09	0.001	0.007	0.01	0.10	1.2	43
10.8	0.01	0.11	0.001	0.008	0.02	0.11	1.8	45
15.2	0.03	0.14	0.001	0.009	0.03	0.15	1.7	46

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Blank "Concrete"

L/S ratio	Total Cr		Cr(III)		Cr(VI)		DOC	
	mg kg⁻¹ dw	acc.	mg kg⁻¹ dw	acc.	mg kg⁻¹ dw	acc.	mg kg⁻¹ dw	acc.
0.1	0.01	0.01	0.001	0.001	0.01	0.01	13	13
0.5	0.01	0.02	0.000	0.001	0.01	0.02	14	27
1.2	0.01	0.03	0.001	0.002	0.01	0.03	3.7	31
1.8	0.01	0.04	0.001	0.003	0.01	0.04	1.8	32
2.4	0.01	0.05	0.001	0.004	0.01	0.05	0.8	33
3.5	0.01	0.06	0.001	0.004	0.01	0.06	1.2	34
4.7	0.01	0.07	0.001	0.005	0.01	0.07	1.0	35
6.1	0.01	0.08	0.001	0.006	0.01	0.08	1.1	36
8.6	0.01	0.09	0.001	0.007	0.01	0.10	1.4	38
10	0.03	0.12	0.005	0.012	0.02	0.12	2.3	40
12	0.01	0.13	0.001	0.013	0.02	0.14	1.5	42

Blank "Sand"

L/S ratio	Total Cr		Cr(III)		Cr(VI)		DOC	
	mg kg⁻¹ dw	acc.	mg kg⁻¹ dw	acc.	mg kg⁻¹ dw	acc.	mg kg⁻¹ dw	acc.
0.1	0.001	0.00	0.001	0.001	0.00002	0.00002	5.3	5.3
0.7	0.001	0.00	0.001	0.002	0.0001	0.0001	2.2	7.5
1.3	0.001	0.00	0.001	0.002	0.0001	0.0003	1.7	9.2
2.0	0.009	0.01	0.009	0.011	0.0001	0.0004	1.5	11
2.7	0.001	0.01	0.001	0.012	0.0002	0.0005	0.8	12
4.0	0.001	0.01	0.001	0.013	0.0001	0.0007	1.0	13
5.4	0.001	0.01	0.001	0.013	0.0001	0.0008	0.8	13
6.6	0.001	0.01	0.001	0.014	0.0001	0.0009	0.6	14
8.9	0.001	0.02	0.001	0.015	0.0002	0.0012	0.6	15
10.0	0.001	0.02	0.001	0.016	0.0002	0.0014	0.3	15
12.5	0.001	0.02	0.001	0.017	0.0002	0.0016	0.6	15

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Blank "Soil"

L/S ratio	Total Cr		Cr(III)		Cr(VI)		DOC	
	mg kg^{-1} dw	acc.	mg kg^{-1} dw	acc.	mg kg^{-1} dw	acc.	mg kg^{-1} dw	acc.
2.4	0.02	0.02	0.02	0.02	0.0005	0.0005	172	172
18.0	0.12	0.1	0.12	0.1	0.003	0.004	880	1052
34.0	0.04	0.2	0.04	0.2	0.003	0.007	576	1628
40.3	0.05	0.2	0.05	0.2	0.001	0.008	203	1831
46.6	0.01	0.2	0.01	0.2	0.001	0.009	115	1946
65.0	0.02	0.2	0.02	0.2	0.004	0.01	353	2299
100.3	0.02	0.3	0.02	0.3	0.003	0.02	167	2466
133.5	0.02	0.3	0.02	0.3	0.003	0.02	241	2706
168.4	0.02	0.3	0.02	0.3	0.004	0.02	190	2896
230.8	0.02	0.3	0.02	0.3	0.005	0.03	115	3011
316.8	0.03	0.4	0.03	0.4	0.006	0.03	124	3135

Annex 5 – Analysis reports from ALS (own electronic document)