



The effects of soil organic matter on leaching of hexavalent chromium from concrete waste: Batch and column experiments

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ABSTRACT

Concrete is one of the most common building materials in the world and in accordance with the world's shift to a circular economy, there is a need of an increase in concrete reuse and recycling. One of the environmental concerns linked to concrete recycling is the leaching and spread of hexavalent chromium (Cr(VI)). In the present study the Cr(VI) leaching from crushed concrete waste and the effects of soil organic matter (SOM) on chromium (Cr) speciation has been investigated in realistic reuse scenarios by the means of batch shale tests and layered column tests. The effects of concrete properties (pH, grain size and age) on Cr(VI) leaching was also studied. Cr leaching from concrete alone is mainly in the form of Cr(VI), with the pH of the leachate being >10. The smaller the grainsize of the concrete, the higher the Cr(VI) concentration is in the leachate. There was no correlation between the age of the concrete and concrete leaching. When exposed to SOM the Total-Cr concentration in the leachate was reduced. The reduction increased with higher TOC level, with a 99% reduction at very high TOC (25%). The results indicate that Cr(VI) leaching from recycled concrete waste can be mitigated by exposing it to SOM in the desired recycling scenario.

The European Commission's Circular Economy Action Plan includes a legislative proposal on waste and provides long-term targets to reduce landfilling and increase recycling and reuse (European Commission, 2020). The action plan aims to lead the way towards a circular economy for the whole of Europe, by putting measures into place that reduce waste generation and ensure a competitive market for secondary raw materials. In 2008, the 2008/1998/EC Waste Framework Directive (European Union, 2008) outlined the goal of achieving a 70% reuse of materials in construction and demolition (C&D) projects by 2020. According to Eurostat data from 2016, the construction sector is responsible for more than 35% of total waste generation in the EU (European Commission, 2020). Concrete makes up a large fraction of this waste and in Norway, for example, 60% of total concrete and brick waste (916 000 tons) is produced by the C&D sector (Statistics Norway, 2020b). Of the total concrete and brick waste generated in Norway per year (1 474 000 tons) only 35% is recycled (70 000 tons), with the remainder ending up at landfill sites (Statistics Norway, 2020a).

Chromium (Cr) is a common element in the raw materials used in cement production (which include limestone, clay and iron agents) (Roskovic et al., 2011). The most stable forms of Cr are trivalent (Cr(III)) and hexavalent (Cr(VI)) (Dayan and Paine, 2001). Cr(III) is an important

trace element in mammals and has a low bioavailability and water solubility. Cr(VI) is water soluble and is hazardous to the environment and humans (IARC, 1990; World Health Organisation (WHO), 1988) and is classified as carcinogenic and allergenic under the EU REACH regulation (EC No 1907/2006 of the European Parliament and of the Council of 18 December 2006 concerning the Registration, Evaluation, 2006). During the production of cement, the raw material is processed in a 1450 °C rotary oven, which results in most of the Cr(III) in the raw material being oxidised to Cr(VI) (Roskovic et al., 2011). This Cr(VI) can cause cement dermatitis in construction workers upon exposure (Kjuus H, 2003). The 2003/53/EC European Directive concerning restrictions and use of certain dangerous substances and preparations therefor limits use and distribution of cement containing more than 2 mg/kg water soluble Cr(VI) (Directive 2003/53/EC, 2003). Reducing agents such as iron (Fe) (II) sulphate (FeSO₄) are added to the cement to reduce Cr(VI) to Cr(III) (Fregert et al., 1979; Roskovic et al., 2011).

In the environment, Cr(VI) is mainly present as the oxyanions CrO₄²⁻ (chromate), HCrO₄⁻ (hydrogen chromate) and Cr₂O₇²⁻ (dichromate) (Banks et al., 2006; Estokova et al., 2018). Under neutral or basic conditions, chromium is mainly present as Cr(OH)₃ and has limited mobility (Apte et al., 2006). Cr(VI) can be reduced to Cr(III) in soils and

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subsequently immobilized. In the environment, the mobility and availability of Cr(VI) is influenced by the reduction and oxidation of Cr, which again depends on soil physicochemical properties such as soil organic matter (SOM), Fe(II), Mn(II) and Mn-oxides and pH (Banks et al., 2006; Bartlett and James, 1979; Buerge and Hug, 1997; Butera et al., 2015; Eary and Rai, 1987; Bruce R. James & R. J. Bartlett, 1983; N. Kozuh et al., 2000; Wittbrodt and Palmer, 1996). Only Manganese is known to oxidize Cr(III) to Cr(VI) (Eary, 1987). Previous studies have shown that Cr(VI) reduction increases with increasing soil total organic carbon (TOC) content (Apte et al., 2006; Bartlett and Kimble, 1976; Choppala et al., 2018; Bruce R. James & R. J. Bartlett, 1983; Mari Pansar-Kallio et al., 2001; N. Kozuh et al., 2000; Wittbrodt and Palmer, 1995).

Concrete is highly alkaline (Barnes and Bensted, 2001) and the Cr contained in its matrix is most often released as Cr(VI) when concrete comes in to contact with water (Estokova et al., 2016, 2018; Sercl erat et al., 2000; Van Der Sloot, 2000). Several previous studies have investigated the leaching of chromium from C&D waste (Butera et al., 2014, 2015; Costeri et al., 2016; Engelsen et al., 2012, 2017; Jung et al., 2014; Kayhanian et al., 2009; Krol, 2020; Loehr, 1996; Saca et al., 2015), but the majority of these studies focused on total Cr concentrations in leachate water and not the speciation of Cr. Studies can also be found related to Cr in cement and the subsequent leaching of Cr(VI) (Estokova et al., 2018; Estokova et al., 2016; Fregert et al., 1979; M. Frias and Sancez Rojas, 1994; Roskovic et al., 2011; Takahashi et al., 2007; Van Der Sloot, 2000; Yamaguchi et al., 2006). However, there is a scarcity of studies that have investigated more realistic scenarios where crushed concrete is utilized in scenarios such as road construction or as a filling material and subsequently leading to leaching of Cr(VI) to soil and groundwater. Existing literature related to remediated waste-sites and waste-materials have not addressed the leaching behaviour of metal ions under persistent rainfall conditions. Engelsen et al. (2012) carried out field trials to simulate a reuse scenario for a road construction and investigated the leaching behaviour of concrete. Recycled concrete aggregates were compared to natural aggregates and used as a sub-base layer in a road construction with and without an asphalt cover. Concentrations of metals (including total-Cr) in leachate water were monitored and were observed to vary between 1 and 100 $\mu\text{g/L}$ for Cr throughout the test period (4 years). The trials showed that the release of Cr was higher from the concrete aggregates than for natural aggregates over the first 100 days of exposure. The pH of the leachate water decreased from over 12 to 10 during a period of two and a half years due to carbonation of the concrete, a process whereby CO_2 dissolves and CaCO_3 is precipitated at high pH (Engelsen et al., 2012). Butera et al. (2015) conducted laboratory batch experiments to investigate the leaching of Cr(VI) from C&D waste in contact with three subsoils with low TOC contents. Based on the results, the authors suggested that the main mechanism for a reduction in Cr(VI) leaching (between 30 and 99.6% depending on experimental duration and soil type and subsequent pH) caused by the soil was a redox reaction with SOM, followed by the precipitation of Cr(III).

Many countries have understood that finding more sustainable uses for waste materials is important and this is now supported by policies that promote recycling and reuse (Inyang, 2003). Increased recycling of waste materials reduces pressure on landfills and virgin materials, reduces the amount of waste needing to be stored, reduces transport costs, and provides more possibilities for sustainable city development. In order to increase the reuse of concrete, negative environmental effects must be avoided. Assessing realistic reuse scenarios where by different soils and different concretes are tested, provides a wealth of data to address this. Therefore, the aim of the present study was to investigate leaching of Cr species from concrete in the presence and absence of soil with different total organic carbon (TOC) contents. To this end, batch shake tests and column tests were conducted with different concrete wastes and soils to mimic realistic reuse scenarios. This is the first study of its kind to investigate the effect of soil TOC content on Cr speciation in

leachate water from concrete waste. The aims of the study were to investigate: 1) the effects of concrete properties such as pH, grain size and age on the amount of Cr(VI) that leaches from concrete waste and 2) how the presence of soil organic carbon affects Cr speciation and fate and transport of Cr(VI) from concrete waste.

1. Materials and method

1.1. Concrete and soil

A total of 147 concrete samples have been analysed in this study, referred to as C1–C147 in the text and Supplementary Material (SM) (see Table 1 in the Supplementary Material (SM) for details about the samples). Sixty concrete waste samples were obtained between 2018 and 2020 from four different C&D projects, one cement producer and two landfill sites. In addition, an unpublished dataset consisting of 87 data points of total solid Cr(VI) concentration in waste concrete from different C&D projects, were provided from the organisation Forum for Milj okartlegging (Forum for Environmental Mapping - FEM) (Werner, 2018). Different sampling methods were used to obtain the samples depending on practicalities and logistics, however they were all collected either using a sledge hammer or a core (40 cm length and 10 cm diameter). Samples were transported to the laboratory in polypropylene plastic buckets and stored cold until further analysis.

Four soils (S1–S4) with: very high (25%), high (5.3%), medium (3.2%) and low (1.6%) TOC contents were used in the experiments described below (Table 1). In addition, a second very high TOC soil (35%) was used in tests with total dissolved organic carbon (DOC). A total of 20 kg was sampled of the different soil types and transported to the laboratory in plastic buckets for further processing. All soil samples were mixed by hand and sieved to ≤ 4 mm before being stored cold for further use. More details are given in Tables S1 and S2 in the SI.

1.2. Concrete sample preparation

Concrete samples (14 kg) were crushed to approximately 40 mm and subsequently ≤ 4 mm first using a sledge hammer and then a jaw-breaker with an adjustable aperture (Retsch Germany jaw breaker type BB100 Wolframcarbide). The 14 kg samples were mixed together by hand in order to avoid loss of fines before being sieved to ≤ 4 mm and 4–40 mm grain size. The moisture content of the concrete and soil samples was determined via oven-drying and loss following heating according to ISO-standard 17 892-1 (ISO17892-1, 2014). Results are shown in Table S4 in the SI.

1.3. Batch shake tests

Batch shake test for leaching of granular waste materials were carried out as a one stage batch test with a liquid to solid (L/S) ratio of 10 (L/S 10) (NS-EN12457-2, 2002). Polyethylene (PE) bottles of different sizes were used in the tests. More details of the test setup are presented in Table S4 in the SI. Shake test numbers 1–27 were carried out in two steps. First an end-over-end shake test with concrete only (test numbers 1–9) was carried out for 24 h to obtain concrete leachate water. Then an end-over-end shake test (test numbers 10–27) was carried out using the obtained concrete leachate water mixed with the soils S1 (25% TOC) and

Table 1
Soil used in experiments describes in this paper. 1.5 column fitting image.

Soil number	TOC (%)	Soil type
S1	25	Compost soil
S2	5.3	Podzol
S3	3.2	Produced soil from landfill site
S4	1.6	Till soil
S5	35	Compost soil

S4 (1.6% TOC) representing very high and low TOC soils, for seven days. After seven days, the samples were filtrated through a 0.45 µm Whatman glass fibre filter using vacuum. Shake test numbers 28–41 were conducted at the external accredited laboratory ALS Laboratory Group Norway AS using the same method as for samples 1–9 (NS-EN12457-2, 2002).

A DOC influence test (test number 82–100) was conducted to investigate the effect of DOC on Cr(VI) in concrete leachate. The test was set up by mixing concrete leachate (from C2, 855 µg/l Cr(VI)) with water containing DOC (from soil S5, 301 mg/L DOC) in a 1:10 ratio. The DOC water was diluted with distilled water in ratios of 1:4, 1:2, 3:4 and 9:10, in addition to a control batch without DOC (Table S4 in the SI). The water containing DOC and the concrete leachate was then made in a single stage batch end-over-end shake test with L/S = 10 (NS-EN12457-2, 2002) for 24 h. The DOC water was filtered in three steps, first through a fine mesh sieve and then through 2 µm and 0.45 µm Whatman glass fibre filter using vacuum. The concrete leachate was filtered through a 0.45 µm Whatman glass fibre filter using vacuum. PE bottles of different sizes was used in the tests. The DOC water and concrete leachate was mixed together in different ratios and were static for seven days only turned (end over end) once a day. One of the samples was used to investigate the effect of time on Cr speciation by letting it stand for 24 h. All batches (except for the 24 h sample (one replicate) and the 1:4 DOC samples (two replicates)) were prepared in triplicates.

1.4. Column tests

Column tests (test numbers 42–81 in Table S4 in the SI) were conducted using two different methods. One set of column tests (test number 54–81) was carried out with up-flow water percolation according to standard NS-EN 14405 with a concrete particle size of ≤ 4 mm and a L/S ratio of 0.1 (test number 54–67) or a L/S ratio of 2 and 10 (test number 68–81). The other set of column tests were run with top-down percolation (test numbers 42–53) where water was added from the top using a PE bottle with holes in the lid to evenly distribute the water. Layered columns with soil at the bottom and concrete at the top were set up to simulate a realistic reuse scenario as well as to investigate the effect of soil organic carbon on the leaching of Cr from concrete. The concrete used (C5) was from a demolition project in Norway and soil samples with 5.3% (S2) and 3.2% (S3) TOC were used (Table S1 in the SI) representing high and medium TOC soils. Soil and concrete were packed into glass columns (50 cm length, 10 cm diameter) with a 5 cm thick non-metal seal with an O-ring at the bottom. For details of the column setup see Fig. S1 in the SI. The columns received 400 mL of Milli-Q water each week for a period of 9 weeks. This volume was chosen in order to collect enough leachate water to carry out weekly sampling and represents approximately three times the normal mean annual precipitation in the Oslo area for the given column surface area (not accounting for evapotranspiration) (Norwegian Climate Centre, 2021). This volume can be considered as a “worst case” scenario as it is likely that the soil/concrete mixture would be exposed to less water than this under a realistic scenario. Prior to starting the experiment, an additional 500 mL of water was added to saturate the columns. The columns were left to stand for two days before starting the experiment. After saturation, water was observed to flow relatively quickly through the columns and over time this flow was reduced likely due to hydration and hardening of the concrete. Sampling started after the second week. The leachate was not filtered before analysis to replicate as closely as possible, leaching in a realistic concrete reuse scenario.

1.5. Solid analysis

Cr(VI) in concrete and soil samples were determined with alkaline digestion using NaOH and Na₂CO₃ and ion chromatography with spectrophotometric detection (ISO15192, 2010). Total Cr was determined using nitric acid digestion that was boiled at 120 °C for 30 min. Extracts

from the digestion was diluted with milliQ water and then analysed by Inductively Coupled Plasma (ICP) (DS-259, 2003). Cr(III) was determined by subtracting Cr(VI) from total Cr. Concentrations of metals and organic pollutants were also analysed. Details can be found in Table 2 the SI.

1.6. Leachate analysis

Leachate from the column and shake tests were sent to ALS laboratory Group Norway AS for analysis of total chromium (Cr-total), Cr(III) (calculated as the difference between Cr-tot and Cr(VI)), Cr(VI) and DOC. Concentrations of Cr(VI) were analysed by introducing filtered sample (0.45 µm) into an ion chromatograph (IC). A guard column removed the organics from the sample and the absorbance of the purple complex of Cr(VI) with 1,5-diphenylcarbohydrazide at 530 nm was measured using a UV spectrophotometer (EPA-7199, 1996; NS-EN16192:2011, 2011). All leachate samples for Cr(VI) analysis were sampled in vials containing an additional preservative ((NH₄)₂SO₂ + NH₄OH). Total Cr was determined by inductively coupled plasma optical emission spectrometry (ICP-OES) (NS-EN16192:2011, 2011; NS-EN-ISO11885, 2009). DOC was determined by loss following incineration (Shimadzu incinerator) (NS-EN1484, 1997). Conductivity and pH were measured using a pH-meter (inoLab, pH level 2) and a conductivity meter (WTW LF 538).

1.7. Quality control and quality assurance

In order to obtain representative concrete samples, approximately 14 kg was mixed by hand and a subsample of 2 kg was taken for each test. All analyses carried out by ALS Laboratory Norway AS were accredited. The limit of quantification (LOQ) of all analytical methods for the solid samples were 0.2 mg/kg. ALS states an uncertainty of 30–40% for the Cr(VI) solid concentration analysis. The LOQ for the analyses of DOC, Cr-total and Cr(VI) in leachate were 0.15 mg/l, 2 µg/l and 0.4 µg/l respectively. Cr(III) was calculated as the difference between Cr-total and Cr(VI). Analytical uncertainties of the DOC analysis were 15%. The uncertainties for the Cr analysis was 10%. Duplicate and triplicate samples were used as far as possible (in tests 1–27, 42–53 and 83–100) and blanks with only soil and only concrete were also run in tests 1–9, 42–43, 44–47 and 82–85. Background concentrations of Cr(VI) in the soil were between 4 and 8% of the concentrations in the concrete in the cases where Cr(VI) was detected. These background concentrations were not corrected for. Jamovi statistical computer software was used for data analysis (correlation and Pearson’s linear regression) (R Core R Core Team, 2020; The jamovi The Jamovi project, 2021).

Table 2

Leached Cr(VI) (µg/l) concentration, percentage Cr(VI) reduction after soil exposure and pH from 0 to 4 mm concrete (C2) and 4–40 mm concrete (C2) mixed with no soil (reference), 25% TOC soil (S1) and 1.6% TOC soil (S4) in a batch end-over-end shake test. 2 column fitting image.

	Concrete reference 0% TOC		Concrete mixed with Soil (S1) 25% TOC		Concrete mixed with Soil (S4) 1.6% TOC	
	0–4 mm	4–40 mm	0–4 mm	4–40 mm	0–4 mm	4–40 mm
	Leached Cr(VI) (µg/l)	464	299	0.51 ± 0.43	0.46 ± 0.19	457.3 ± 22.4
Percentage reduction of Cr (VI) (%)			± 0.09	± 0.06	2.7	6.2
pH	12.2	12.2	6.1 ± 0.1	6.1 ± 0.2	11.4 ± 0.05	11.7 ± 0.09

2. Results and discussion

2.1. Hexavalent chromium in solid concrete

Fig. 1 shows the concentration of Cr(VI) (1 A) and total Cr (1 B) in concrete samples C1–C18 and C61–C87 with known age (year of production) (For details of the samples see Table S3 the SI). There was no significant correlation between concrete age and total Cr(VI) concentration ($R^2 = 0.15$, $p = 0.128$) or total Cr concentration ($R = 0.104$, $p = 0.358$). Over time, concrete can be subject to carbonation which occurs when concrete is exposed to carbon dioxide (CO_2). When the calcium hydroxide (CaOH) in the cement comes into contact with CO_2 , calcium carbonate is formed (CaCO_3) leading to a pH reduction in the concrete porewater to below 10 and a subsequently breakdown of the hydrate phases in the cement. (Hartmann et al., 1999; Lagerblad, 2005; Macias et al., 1997; Mulugeta et al., 2011). Although not directly correlated with the concrete age, the solubility of Cr has been reported to increase as concrete is carbonized (Alba et al., 2001; Demars and Benoit, 2019; Macias et al., 1997; Mulugeta et al., 2011). Results from leaching tests on recycled concrete aggregates with variable degrees of carbonation showed enhanced release of elements such as arsenic (As), molybdenum (Mo), antimony (Sb), selenium (Se), vanadium (V) and Cr upon carbonation. The elements leached were mostly in the form of oxyanion and it can be expected that for Cr the dominating species was Cr(VI) (Mulugeta et al., 2011).

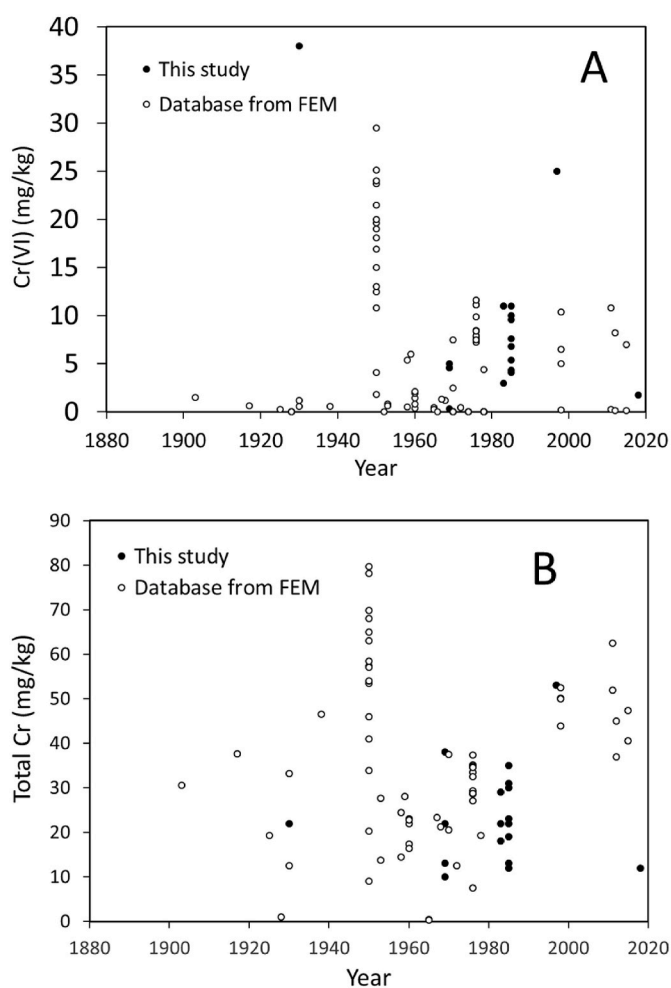


Fig. 1. Concentration of Cr(VI) (mg/kg) (A) and total Cr (B) in concrete samples compared to their age (production year). The figure shows concrete samples (C1–C18) from this study (black) and from previous work (Werner, 2018) (white). 1.5 column fitting image.

There are many factors that can affect the concentration of Cr(VI) in concrete including the sample matrix (the aggregate/cement ratio), sampling method and equipment used as well as analytical method and sample preparation method used (Borai et al., 2002; Huo and Kingston, 2000). The samples included in Fig. 1 were sampled using either a hammer and chisel, a concrete core or a sledgehammer. These different sampling methods result in samples with different representativeness of the concrete structure they were taken from (aggregate/cement ratio) as well as different degrees of crushing before storage. Previously, finely crushed material (obtained using a hammer and chisel) has been shown to be more exposed to air and other external conditions than monolithic structures (obtained using a core) which affects the Cr speciation of the sample (Shin and Paik, 2000). Thus, it is likely that sampling method may have affected the lack of correlation observed here.

When determining total solid concentration of Cr(VI), (i) soluble, (ii) sparingly soluble and (iii) insoluble Cr(VI) has to be extracted (Grabarczyk et al., 2006). This extraction is challenging due to possible changes in the Cr oxidation state during the course of the analytical process combined with concentrations that are often close to the analytical detection limit (Borai et al., 2002; Huo and Kingston, 2000; Korolczuk and Grabarczyk, 2005). There are many different analytical methods that can be used for Cr(VI) determination in solid samples. The US Environmental Protection Agency (EPA) accepts the following methods; 7195 precipitation, 7196 colorimetric with diphenyl carbazide, 7197 chelation with ammonium pyrrolidine dithiocarbamate (APDC) and extraction with methyl isobutyl ketone, 7198 differential pulse polarography and 3060 A alkaline digestion using NaOH and Na_2CO_3 (Borai et al., 2002; US EPA). The results presented in Fig. 1 were analysed using ISO 15192 with alkaline digestion (similar to EPA 3060 A using NaOH and Na_2CO_3) and ion chromatography with spectrophotometric detection.

Literature related to concentrations of trace elements including Cr (and Cr(VI)) in concrete of different ages is scarce. Costeri et al. (2016) analysed 18 samples for Cr(VI), total-Cr and other elements in cement from 1925 to 1940 sampled from walls and pillars of old industrial and residential buildings and new samples from commercial cements. Cement and aggregates in the old concrete was separated using a screw press, sieved and crushed to a fine fraction ($<63 \mu\text{m}$) then analysed using acid extraction (HCL 2 N). Cr(VI) concentrations in the old and new cement samples were in the range of 8–15 mg/kg and no clear correlation with age was observed. Most of the old concrete samples studied by Costeri et al. (2016) are older than the samples shown in Fig. 1 of this study and the sampling preparation (only cement vs concrete with cement and aggregates) and analytical methods (acid extraction vs alkaline digestion) are also different. Despite this both studies show it is challenging to relate Cr(VI) concentration to concrete age.

EU Directive 2003/53/EC which stipulates a maximum concentration of 2 mg/kg of water soluble Cr(VI) in cement is often met by cement producers by adding iron sulphate (FeSO_4) to the cement that can reduce Cr(VI) to Cr(III) (European Union, 2002; Fregert et al., 1979). However, Geelhoed et al. (2003) reported an increased leaching of Cr(VI) following the addition of FeSO_4 to column experiments due to the precipitation of Fe(II) at high pH that failed to reduce the Cr(VI) to Cr(III). Based on the data presented in this study there is no clear correlation between total Cr(VI) concentration in concrete before and after the practise of adding FeSO_4 to the cement assuming the practise started when the EU Directive 2003/53/EC came into effect in 2005, which can indicate that the FeSO_4 loses some effect with time.

2.2. Chromium speciation and mobility in crushed waste concrete

Results from the crushed waste concrete (C5) unsaturated leaching tests showed that 99.9% of the Cr leached from the concrete alone was in the form of Cr(VI). Fig. 2 shows the leaching of Cr(VI) with increasing L/S (0.3–2.4) from the two concrete unsaturated column tests. Substantial

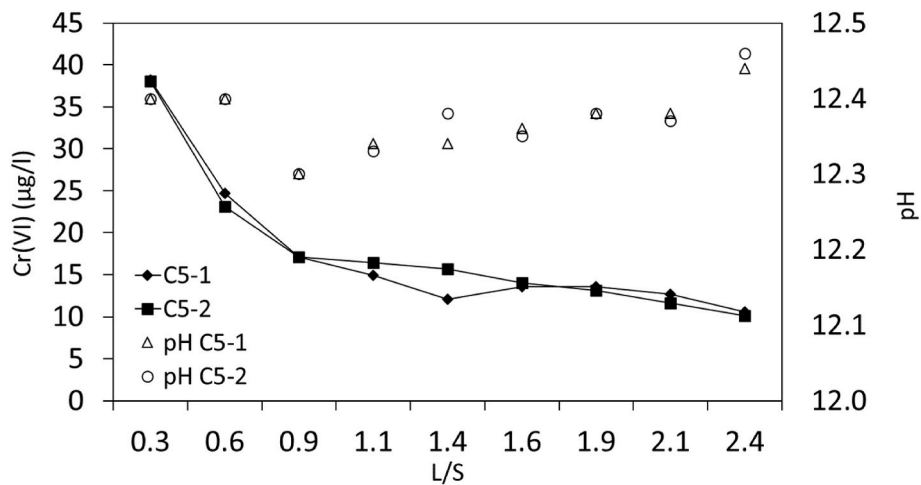


Fig. 2. Leaching of Cr(VI) (µg/l) and leachate pH with increasing L/S from waste concrete from a tunnel rehabilitation project (C5) in a duplicate unsaturated column test. 2 column fitting image.

leaching was seen in the first week (38.2 µg/l ± 0.05). Leaching reduced rapidly with increased L/S (to 17.1 ± 0.0 µg/l) and then stabilised at around 15 µg/l with a slow decline up to L/S 2.4–10.4 µg/l ± 0.25. The pH in both columns throughout the test was stable at around 12.4 (all values are reported in Table S5 in the SI) and the results showed no correlation between pH and Cr(VI) leaching (R² = 0.031).

High Cr(VI) concentrations at low L/S (0.3) with a subsequent rapid reduction over time is most likely due to a first flush effect and the presence of preferential flow paths which results in elevated concentrations upon exposure to water, which then level off quite rapidly and stabilise (Deletic, 1998; Geelhoed et al., 2003). Fig. 3 shows leaching of Cr(VI) from waste concrete from a demolition project (C10, C59, C7, C8) and from the landfill (C19) in a saturated column tests with L/S 0.1, 2.0 and 10. Overall, the concentrations in the saturated column tests are higher than in the unsaturated column test, with concentrations ranging from 36.2 µg/l to 187 µg/l at the lowest L/S (0.1) and 7.2 µg/l to 74 µg/l at the highest L/S (10) (all values are reported in Table S10 in the SI). The trend is however similar to the unsaturated column with high Cr(VI) concentrations at low L/S and a rapid reduction with increased L/S.

The first flush effect is often related to rainfall events where a larger

volume of water results in a higher concentration of contaminants at the start of the event than later in the rainfall event (Deletic, 1998; Kayhanian and Streenstrom, 2005). Preferential flow paths, defined as flow paths in the matrix where the water moves faster and more easily (Gerke, 2006; Hendrickx and Flury, 2001), may affect Cr(VI) concentrations as water is only in contact with a portion of the concrete throughout the test period. The variable size of the concrete aggregates that were used in the columns (0–4 mm) increases the chances of preferential flow paths. The high Cr(VI) leached concentration in the first sample, followed by a rapid decline is in line with previous literature reporting leaching tests (Saca et al., 2015; Geelhoed et al., 2003; Takahashi et al., 2007; Zhang and Lin, 2020; Overmann et al., 2021). Geelhoed et al. (2003) conducted column experiments to investigate Cr(VI) leaching from Chromite Ore Processing Residue (COPR) using NaCl as a leaching agent and adding FeSO₄ to look at the effect of FeSO₄ on Cr(VI) conversion and leaching. COPR has similar chemical properties as cement (mineral composition, pH, Cr levels). Leaching of Cr(VI) from COPR with NaCl showed a similar leaching curve with this study, with higher leaching the first week (approximately 32 mg/l Cr(VI)) and a strong decrease after the first pore volume (approximately 23 mg/l Cr

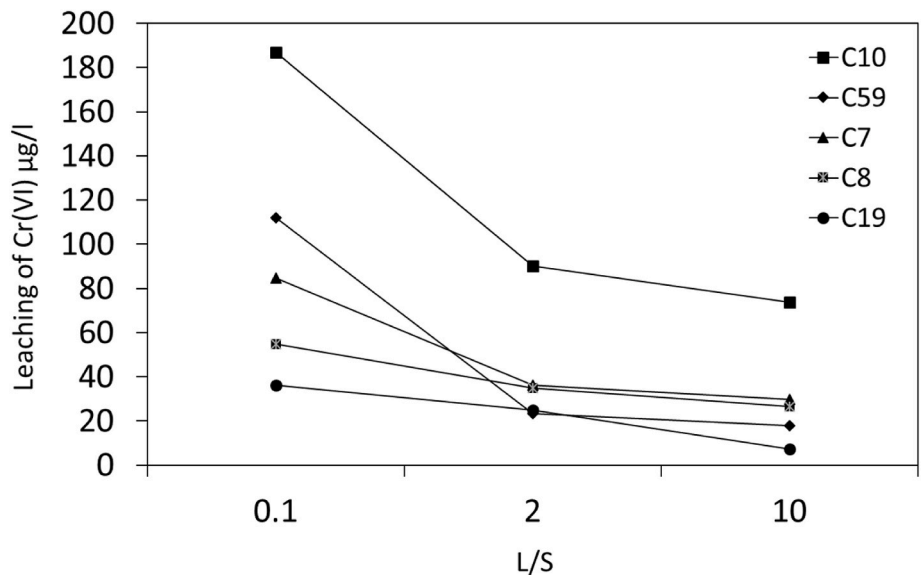


Fig. 3. Leaching of hexavalent chromium (Cr(VI)) (µg/l) from waste concrete from a demolition project (C10, C59, C7, C8) and from landfill (C19) in a saturated column tests with L/S 0.1, 2.0 and 10.2 column fitting image.

(VI)), as well as only a slight decrease in pH (11.8–11.5 after 70 pore volumes). The results showed that after 70 pore volumes analysed, only 3% of the total Cr(VI) in the COPR had leached. This is in line with the results presented here showing that only 7% of the total Cr(VI) in the concrete leached over the experimental period (total amount (μg) of Cr(VI) leached from the columns as a percent of the total solid concentration (μg) of Cr(VI) in the columns, see eqns. (1) and (2) in the SI), indicating that concrete may represent a long term source of Cr(VI) when exposed to water. Takahashi et al., 2007 conducted tank leaching tests, where a monolithic concrete structure was added to a tank with distilled water and leaching of Cr(VI) from the concrete was determined. The authors reported elevated leaching of Cr(VI) at the start of the experiment, followed by a decreased with subsequent rainfall (to 4–8 $\mu\text{g/l}$ Cr(VI)), concluding that the overall impact of Cr(VI) leaching was low in their experimental setup. This conclusion may not hold true if external conditions are altered (i.e. pH).

The concentration of Cr oxyanions in concrete and cement peaks at neutral to mild alkaline pH and leachability is lower at both low and high pH (Loehr, 1996; Van Der Sloot, 2000). At high pH (over 10) phases such as ettringite and calcium silicate hydrate (C–S–H phases, which is the primary binding phase in cement based materials) can play a role to decrease the leachability of Cr (Mulugeta et al., 2011; Van Der Sloot, 2000). In the column test presented here, the pH was always above 12, however it is plausible that leaching would increase with reducing pH values and then decrease again below pH 6.

2.3. Cr(VI) leaching from crushed concrete waste vs total solid concentration of Cr(VI)

The Cr(VI) concentrations in the leachate from the batch shake tests (L/S 10) and the column test (L/S 0.1) (Tables S10 and S11 in the SI), are plotted against the total solid concentration of Cr(VI) in Fig. 4. The results showed no clear correlation between solid concentration and leachate concentration from the batch shake test (Fig. 4A, $R^2 = 0.049$) or the column test (Fig. 4B, $R^2 = 0.07$). This is in line with studies carried out by Van Der Sloot (2000) who investigated concentrations in cement mortars which were crushed to <4 mm with a jaw crusher and used in leaching tests and crushed again to <125 μm for availability tests according to standard NEN 7341:1994 (NEN-7341, 1994). The results showed a poor correlation between total and potentially leachable Cr in the cement mortars (R^2 not given). The fraction of the total concentration of Cr in the cements that was potentially leachable in the Van Der Sloot (2006) experiment was determined to be approximately 35%. These results highlight the shortcoming in trying to estimate environmental effects of Cr(VI) from concrete just based on the total solid concentration of Cr(VI) in concrete and/or cement (Van Der Sloot, 2000).

2.4. Effect of soil organic carbon on Cr(VI) leaching from concrete

In this study batch end-over-end shake tests, layered column tests with concrete and soil were conducted to look at the effect of soil organic carbon on Cr(VI) leaching from crushed concrete waste. A static batch test was also carried out whereby concrete leachate was mixed with water containing various amounts of DOC to investigate the effects of DOC on Cr(VI) leaching. Table 2 shows leaching and percentage reduction of Cr(VI) from concrete C2 (demolition project) at ≤ 4 mm and 4–40 mm fractions following the end-over-end shake test with concrete alone, 25% (S1) and 1.6% (S4) TOC soil. The concentration of Cr(VI) in the leachate from concrete alone was higher in the ≤ 4 mm concrete fraction (564 $\mu\text{g/l}$) than the ≤ 40 mm concrete fraction indicating a possible effect of concrete aggregate size on leaching. The Cr(VI) concentration was reduced in all samples mixed with soil except one, and there was a significant negative correlation between TOC level and Cr(VI) leaching ($R^2: -0.92$, $P < 0.001$), meaning that the Cr(VI) leaching is reduced with increasing TOC. Mixing the ≤ 4 mm concrete fraction with

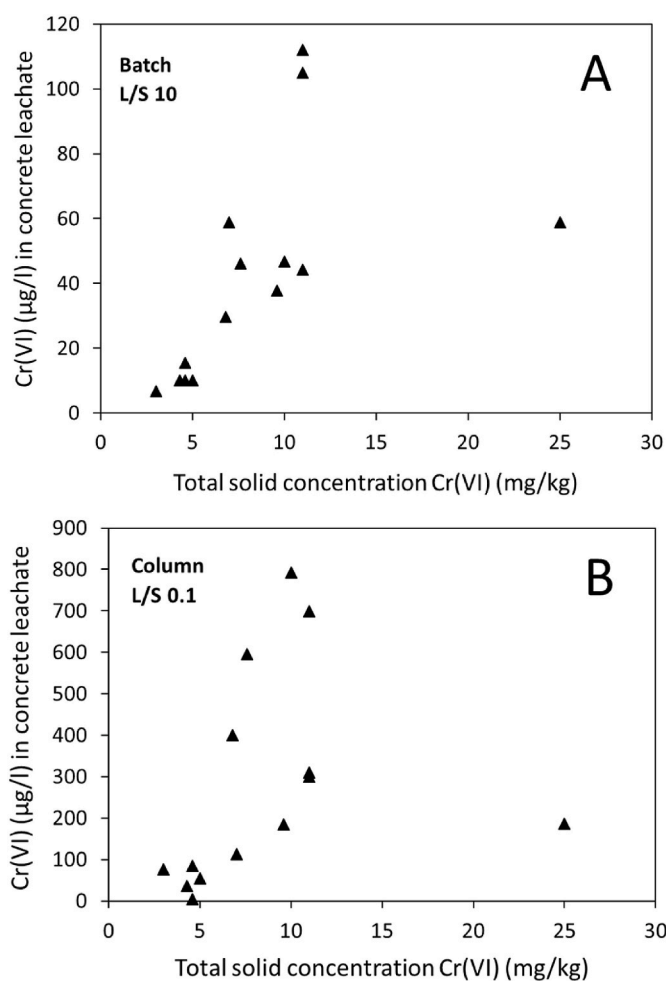


Fig. 4. Leached Cr(VI) ($\mu\text{g/l}$) concentration from batch shake test (L/S 10) (A) and column test (L/S 0.1) (B) of concrete aggregates compared with total solid concentration of Cr(VI) (mg/kg) in the same concrete aggregate samples. The LOQ for the analytical methods was 20 $\mu\text{g/l}$ Cr(VI). Concentrations below the LOQ were removed from the data set. 1.5 column fitting image.

low TOC soil had little effect on the Cr(VI) leachate concentrations and the maximum reduction achieved was 7%. The reduction was higher in the leachate from the 4–40 mm concrete fraction (8–23% reduction). The pH was relatively stable throughout the test, ranging from 11.4 to 11.8. When exposed to soil S1 (25% TOC) the percentage reduction of Cr(VI) in the leachate was close to 100% and the pH was in the range of 5.9–6.3. There is a significant positive correlation between pH and Cr(VI) leaching ($R^2: 0.67$, $P = 0.003$). There was no leaching of Cr(VI) from concrete C1 (newly casted) before or after exposure to soils S1 and S4 (Table S12 in the SI).

The layered column experiments were carried out with concrete sample C5 and 3.2% (S3) and 5.3% TOC soil (S2). The leached concentrations of Cr(VI) from the layered column tests are reported in Fig. 5 and Tables S6–9 in the SI. There was a reduction of Cr(VI) leaching after exposure to both types of soil. There was no Cr(VI) leaching from the soil alone. In the layered columns with 3.2% TOC soil and concrete (Fig. 5A) the leachate Cr(VI) concentrations was on average 2.1 $\mu\text{g/l}$ (± 0.096) after one week. By week two, concentrations were below the LOQ (0.4 $\mu\text{g/l}$). Leaching of Cr-total was low (ranging from 1 to 3.4 $\mu\text{g/l}$) and most of it was in the form of Cr(III). pH was stable, ranging from 7.3 to 7.8. The leachate Cr(VI) concentrations in the column with the 5.3% TOC soil (Fig. 5B) were below the LOQ at all L/S values. The pH dropped from 5 to 4 the first week when adding water before it steadily increased to an average of 5.4. These pH-variations did not seem to have an effect

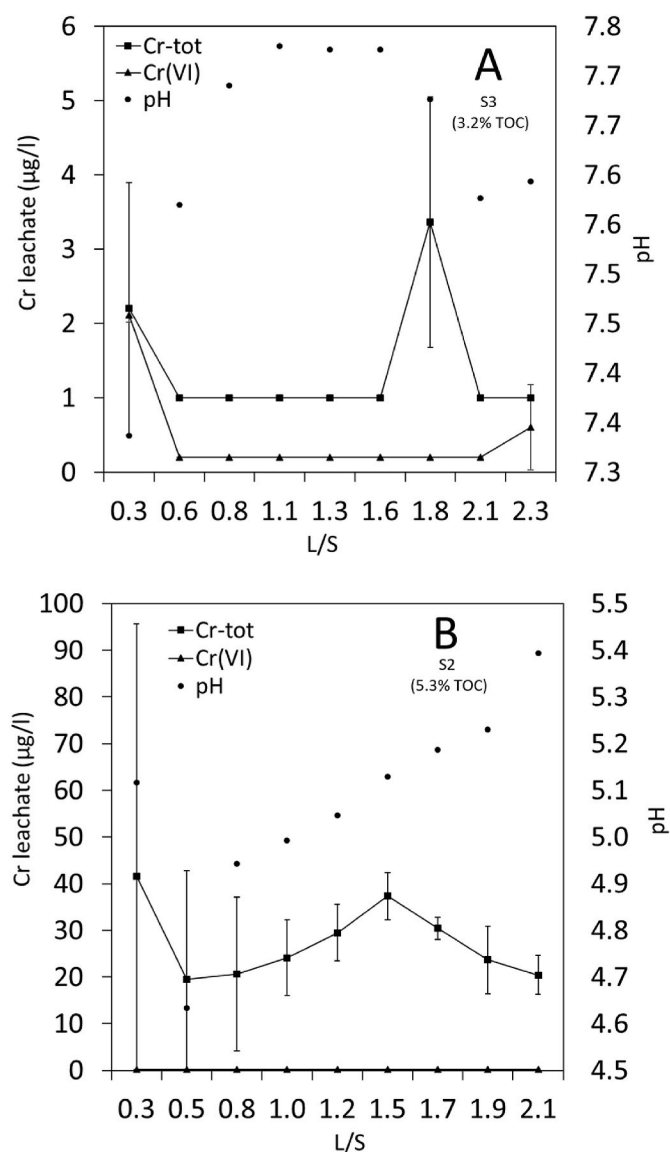


Fig. 5. Leaching of Cr(VI) ($\mu\text{g/l}$) and leachate pH with increasing L/S from waste concrete (C5) in an unsaturated layered column test with A) medium TOC soil (S3, 3.2% TOC) and B) high TOC soil (S2, 5.3% TOC). The lines show average value from triplicates with SD. Cr(VI) and Cr(III) concentration below LOQ is used as half of the LOQ (0.4 $\mu\text{g/l}$ and 2 $\mu\text{g/l}$ respectively). 1.5 column fitting image.

on Cr speciation. The concentration of Cr-total (in the form of Cr(III)) in leachate water varied from below the analytical detection limit to 118 $\mu\text{g/l}$. The results showed more Cr(III) leaching from the layered columns with 5.3% TOC than from concrete alone. The soil alone had little leaching of Cr(III) (from <2–5.6 $\mu\text{g/l}$), so a possible explanation can be the higher acidity (ranging from 4.2 to 6.3) and organic content in the soil which results in the concrete being dissolved to a greater degree right at the interface between the soil and the concrete. Organic complexed Cr(III) in soils are often more soluble compared to the Cr(III) metal ion, that is quickly adsorbed (B. R. James & R. J. Bartlett, 1983). Bruce R. James and R. J. Bartlett (1983) found in their experiments that added citric acid to soil spiked with Cr(VI) ($\text{K}_2\text{Cr}_2\text{O}_7$) resulted in the formation of soluble Cr(III) complexes after the reduction of Cr(VI). Similarly, our results show a clear correlation between total Cr and DOC in the leachate from concrete mixed with high TOC soil ($R^2 = 0.90$, $P < 0.001$) and can explain the increased Cr(III) concentration in the leachate. A decrease in Cr(VI) in the soil can be due to direct adsorption

of Cr(VI) to soil particles and to the reduction of Cr(VI) to Cr(III) and subsequently immobilisation through adsorption. Ferrous iron, humic substances and sulphur are able to reduce Cr(VI) to Cr(III) in soil (Bartlett and Kimble, 1976; Eary and Rai, 1991; Pettine et al., 1998; Wittbrodt and Palmer, 1995). Previous studies have shown that the reduction of Cr(VI) to Cr(III) is often the dominate process and that this process is fast, reaching a maximum in just a few days (Bolan et al., 2003; James, 1995). This is in line with the results presented in this study that showed a rapid decline of Cr(VI) in the concrete leachate when exposed to SOM. The redox potential of the soil also plays a big role in the Cr speciation and potential release of Cr(VI). Zhang and Lin (2020) investigated the influence of redox conditions on Cr release from cement-based stabilisation of soil and reported an increase of Cr leaching with increasing redox potential (Eh). Cr leaching was high during the first two days before it steadily declined for all redox potentials used in the study (0, 200 and 400 mV). This is in line with the results presented in this paper where Cr leaching is initially high, before it steadily drops. Zhang and Lin (2020) concluded that the initial high leaching of Cr was due to diffusion controlled leaching of Cr in unstable phases.

Soil pH is an important factor determining Cr sorption in soils. In alkaline soils Cr(III) is often adsorbed on negatively charged soil surface sites or precipitated as $\text{Cr}(\text{OH})_3$ (Bartlett, 1991; Bradl, 2004; Choppala et al., 2018). As soil pH is increased Cr(VI) reduction to Cr(III) is reduced as well as increasing the negative charges on the soil surface causing a decrease in Cr(VI) sorption (Jiang et al., 2008). This was confirmed in a study looking at sorption of Cr species where lime and elemental sulphur was used to alter pH (Choppala et al., 2018). In acidic soils Cr(VI) sorption is higher due to the increase of positively charged soil surfaces (Bradl, 2004). The Cr(VI) ion is adsorbed to the soil due to surface complexation reactions with hydroxyl sites (Ajouyed et al., 2010; Choppala et al., 2018; Eary and Rai, 1991). Fe(II) oxide is also known to increase the sorption of Cr(VI) to soils due to its addition of positive charge to the soil and through complexation with hydroxyl species of iron oxides (Choppala et al., 2018). According to Zachara et al. (1987) minerals with proton specific surface sites and high point of zero charge (PZC), like Fe and aluminium (Al) oxides, can adsorb Cr(VI) at pH 2–7. However due to the presence of other anions competing for adsorption sites, chromate mobility can still be high (Wittbrodt and Palmer, 1995). Bartlett and Kimble (1976) found in their experiments that in a solution containing Al and Cr(VI), the Cr(VI) precipitated at pH 6 and was almost completely insoluble until the pH was above 8. Thus, in the layered columns with 5.3% TOC soil presented in this study, where the pH range was between 4.2 and 6.3 the Cr could be adsorbed as Cr(VI) in the soil. This could explain the high Cr(III) leaching from these columns.

In the static batch test with DOC and concrete leachate, the percentage Cr(VI) of the total Cr in the samples was still high but decreased with increasing DOC content (83.4 with no DOC and 67.1 $\mu\text{g/l}$ with 9:10 DOC) (Fig. S3). Total Cr remained almost constant. These findings are in line with existing literature showing that DOC is one of the potential drivers of Cr(VI) reduction (Jardine et al., 1999). Detailed results from the static DOC batch test is reported in Table S13 and Fig. S3 in the SI. Soil humic substances which make up the majority of organic matter in soils, is a source of electron donors for the reduction of Cr(VI) (Wittbrodt and Palmer, 1996). Thus humic and/or fulvic acids can effectively reduce Cr(VI) to Cr(III) at acid pH (Wittbrodt and Palmer, 1995, 1996, 1997). Jardine et al. (1999) studied the impact of hydrologic and geochemical processes on the fate and transport of Cr(VI) in undisturbed soil cores with acidic inceptisol and found that the Cr(VI) mobility significantly decreased in the presence of DOC in the soil. Batch mixtures of DOC (mainly consisting of humic and fulvic acids) and Cr(VI) without solid phase material showed a decrease in aqueous phase Cr(VI) exponentially with time (DOC concentration was stable), while total Cr remained stable. This indicates that the dominant reaction occurring in the study presented by Jardine et al. (1999) was reduction of Cr(VI) to Cr(III) when the leachate water was exposed to the soil. Bolan et al. (2003) studied the reduction of Cr(VI) in a mineral soil with low TOC

(0.9%) after adding different organic amendments. Soil samples were mixed with a solution containing known Cr concentrations (60 mg/l of added $K_2Cr_2O_7$), and then subjected to end-over-end shaking batch test for 16 h. Both the concentration of Cr(III) and Cr(VI) were reduced in solution (from 60 mg/l to 53.1 and 55.7 mg/l respectively) following amendment. They also found a significant positive linear relationship between the extent of Cr(VI) reduction and the amount of DOC in the soil. The authors concluded that the decrease of Cr(III) was due to adsorption and precipitation and that the decrease of Cr(VI) was due to reduction to Cr(III) in the presence of organic material and subsequently adsorption. They found no evidence of Cr(VI) adsorption to the soil. This is consistent with the results presented in this study that showed a reduction of both Cr(VI) and Cr(III) concentrations in all of the leaching tests except the layered column with 5.3% TOC where Cr(III) leaching increased. In this case most of the Cr(VI) from the concrete was most likely reduced to Cr(III) due to the high TOC levels, but the sorption capacity of the soil for Cr(III) was low (due to low pH) resulting in Cr(III) leaching. The concentration of Cr(VI) was reduced the most in soil with medium to very high TOC levels which is consistent with existing literature (Banks et al., 2006). This can indicate that the Cr(VI) released from the concrete was reduced to Cr(III) when exposed to TOC and in most cases subsequently immobilized in the soil.

3. Conclusion

Concrete batch shake and layered leaching tests have shown that Cr is mainly in the form of Cr(VI) when leached from crushed concrete alone. Speciation analysis showed that chromium is mainly in the form of Cr(VI) when pH is > 10 (as is the case for concrete leachate) and reduces to Cr(III) as the pH lowers. The results in this study show no clear effects of pH fluctuations when below neutral. Cr(VI) in concrete leachate is high at first due to a first flush effect, but levels out with time. The leaching increased with reduced concrete grain size due to increased specific surface area. This means that although Cr(VI) leaching from larger grain sizes and monolithic structures might not be of environmental concern, crushed concrete waste aggregates can leach Cr(VI) when used alone and in environments where the chemical properties such as pH remain unchanged.

Results showed that there was no clear correlation between concrete age and Cr(VI) solid concentration or between total solid concentration of Cr(VI) in the concrete and Cr(VI) leached. Literature shows that there could however be a link between concrete carbonation and Cr(VI) leaching, which is somewhat connected to aging. Hence it is mainly soil organic carbon content and the redox potential of the soil, rather than concrete age that will affect the Cr speciation in the leachates and carbonation/calcination in concrete-debris after entering the soil system. It is these factors that determine how hazardous a crushed concrete waste can be for a specific disposal site.

Column leaching tests showed that when concrete leachate is percolated through soil containing different levels of SOM, Cr(VI) is in most cases reduced to Cr(III) and subsequently immobilized in the soil. Reduction increases with higher TOC level. DOC was an important reducing agent in this study, but the reduction of Cr(VI) is most likely controlled by many different properties in the soil such as Fe and/or Mn content, SOM and acidity. Long term field experiments are necessary to look at the effects of the sorption and reduction capacity of the soil over time.

Concrete is the one of the most common building materials in the world and subsequently huge amounts of concrete waste is generated every day. If the building and construction industry are to move towards a circular economy and reduce their environmental footprint more of this concrete waste will need to be recycled and reused. This work has demonstrated potential environmental concerns related to Cr(VI) leaching from crushed waste concrete. However, this may be amended by changing the physiochemical properties surrounding the concrete to reduce the leaching. Examples of this could be to avoid placing crushed

concrete directly on bedrock to minimize the concentration of Cr(VI) in leachate which could present a risk to the environment. Road constructions, car parks and other typical applications of recycled crushed concrete should be designed with a layer of soil below the concrete back filling (if the technical specifications allow) that can reduce and immobilise the Cr(VI). In many cases soil will be present in the digging pit which could be used as backfill.

For the next chapter in this research, a pH dependence tests on concrete mortar with different cement types and carbonation degrees is planned to look at the pH effect on Cr speciation in different types of concrete. To address the possible long-term leaching potential of Cr(VI) in the concrete dynamic leaching tests (or tank-tests) could be carried out on crushed concrete to determine the release rates of chromium under diffusion controlled conditions over time. Obtained results may provide a stronger indication of whether a relationship exists between concrete age and Cr leaching. Further research should also look at Cr(VI) reducing capacity of the soils and their vulnerability to future changes in pH.

Credit author statement

Cathrine Eckbo: Conceptualization, Methodology, Formal analysis, Investigation, Writing – original draft, Writing – review & editing, Visualisation, Project administration, **Gudny Okkenhaug:** Conceptualization, Methodology, Validation, Writing – review & editing, Supervision, Funding acquisition, **Sarah Hale:** Conceptualization, Methodology, Validation, Writing – review & editing, Supervision

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jenvman.2022.114708>.

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