



# Soil retention of hexavalent chromium released from construction and demolition waste in a road-base-application scenario



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## HIGHLIGHTS

- Cr(VI) from alkaline sources retained in subsoils with low organic matter.
- Slow Cr(VI) reduction followed by precipitation as Cr(III).
- Modelled Cr(VI) migration contained within 70 cm depth.
- High water infiltration and temperate climates might result in migration up to 2 m.

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## ABSTRACT

We investigated the retention of Cr(VI) in three subsoils with low organic matter content in laboratory experiments at concentration levels relevant to represent leachates from construction and demolition waste (C&DW) reused as unbound material in road construction. The retention mechanism appeared to be reduction and subsequent precipitation as Cr(III) on the soil. The reduction process was slow and in several experiments it was still proceeding at the end of the six-month experimental period. The overall retention reaction fit well with a second-order reaction governed by actual Cr(VI) concentration and reduction capacity of the soil. The experimentally determined reduction capacities and second-order kinetic parameters were used to model, for a 100-year period, the one-dimensional migration of Cr(VI) in the subsoil under a layer of C&DW. The resulting Cr(VI) concentration would be negligible below 7–70 cm depth. However, in rigid climates and with high water infiltration through the road pavement, the reduction reaction could be so slow that Cr(VI) might migrate as deep as 200 cm under the road. The reaction parameters and the model can form the basis for systematically assessing under which scenarios Cr(VI) from C&DW could lead to an environmental issue for ground- and receiving surface waters.

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## 1. Introduction

When waste materials are utilized in geotechnical applications, hazardous compound leaching to the underlying soil is one of the main environmental concerns, with potential consequences as severe as groundwater pollution and impaired drinking-water resources. Quantifying the leaching is paramount, and numerous studies concern leaching from different materials that can be

reused in geotechnical scenarios. Most studies address municipal solid waste incineration bottom ash (MSWI BA), showing that copper, lead, zinc, cadmium, chromium, molybdenum, and antimony [1–5] are of the most concern in the leachate. In the case of construction and demolition waste (C&DW), primarily oxyanion-forming elements such as chromium and selenium are problematic in the leachate [6–9]. Estimation of leaching of, among others, Cr from C&DW can be found in e.g., Engelsen et al. [10], Butera et al. [6], and Butera et al. [7]. Chromium, which is naturally present in cementitious materials as a consequence of use of different fuels during clinker production, grinding and use of additives in cement or concrete production, is believed to be released in hexavalent form [6,11].

However, characterizing the leachate constitutes only the first step of a full assessment of the overall environmental impact. The fate of the emitted contaminants in the subsoil must be taken into account, as interactions with the soil might involve processes such

*Abbreviations:* C&DW, construction and demolition waste; CEC, cation exchange capacity; HFO/AIO, hydrous iron/aluminum oxides; ICP-OES, inductively coupled plasma optical emission spectrometry; L/S, liquid to solid ratio; LOD, limit of detection; MSWI BA, municipal solid waste incineration bottom ash; SFA, soil fulvic acid; SHA, soil humic acid; SOM, soil organic matter; TS, total solid.

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**Table 1**  
Selected chemical parameters of the three experimental soils (TS: total solid).

		S1	S2	S3
Depth		50–100	55–100	65–100
Horizon		B-horizon	C-horizon	C-horizon
Texture <sup>a</sup>		Sand with clay	Coarse sand	Sand with clay and silt
Clay (<0.002 mm)	%	8.1	2.4	14
Silt (0.002–0.02 mm)	%	8	0.8	11
Fine sand (0.02–0.2 mm)	%	49	16	49
Coarse sand (0.2–2 mm)	%	35	80	26
Soil organic matter (SOM) <sup>b</sup>	%	0.3	0.4	0.3
Cation exchange capacity (CEC) <sup>c</sup>	meq·kg <sup>-1</sup> TS	47	26	80
pH (CaCl <sub>2</sub> ) <sup>d</sup>	–	4.4	5.3	6.4

<sup>a</sup> Soil texture was determined according to the hydrometer analysis, which uses a Bouyoucus hydrometer with a scale 0–60 g·L<sup>-1</sup>. The effective depth and the time of settling are calculated according to the theory of Day [Hansen L. (1961). Hydrometermetoden til bestemmelse af jordens tekstur. Grundförbättring 1961 (3) 177–188. In Danish].

<sup>b</sup> Dry combustion by Fisher induction carbon apparatus [Jackson, M.L. (1962). Soil Chemical Analysis. 3rd edition. Prentice Hall Inc. US, pg 208].

<sup>c</sup> Determination with ammonium following extraction of exchangeable cations [Jackson, M.L. (1962). Soil Chemical Analysis. 3rd edition. Prentice Hall Inc. US, pg 66; Peech, M. (1945). Determination of exchangeable cations and exchange capacity. Soil Science 59, 25–28].

<sup>d</sup> Instrumental method for the routine determination of pH using a glass electrode in a 1:5 (volume fraction) suspension of soil in 0.01 mol/l calcium chloride solution (pH in CaCl<sub>2</sub>). ISO (2005) [ISO standard procedure 10.390:2005 Soil quality – Determination of pH].

as precipitation, sorption, or complexation affecting the contaminant mobility or bioavailability. In the case of chromium, both mobility and toxicity greatly depend upon the oxidation state of chromium, which might be affected by interactions with the soil. Trivalent Cr is a nutrient [12], sorbs strongly to soils, and has low solubility above pH 5.5 [13]. Cr in its hexavalent form is toxic, mutagenic, and carcinogenic, with significant solubility and mobility [14,15]. Therefore, detailed knowledge about potential chromium interactions with subsoils is important for assessing the consequences of leaching from C&DW reused for geotechnical purposes.

Chromium chemistry and redox behavior have been extensively studied, also in terms of possible soil interactions. While Cr(VI) adsorption is disfavored at high pH, owing to repulsion between negatively charged soil particles and chromate anions, adsorption was found still to play a significant role at pH as high as 7–8 [16] with hydrous iron oxides (HFO) being the strongest adsorbent compared with aluminum oxides (AlO) and clay. Cr(III) can be oxidized by dissolved oxygen at pH > 9, and by MnO<sub>2</sub>, increasingly with decreasing pH [17]. Possible electron donors for Cr(VI) reduction include organic matter [18–21], Fe(II) [22], and sulfides [17]. Cr(VI) reduction is favored by acidic environments [23,24], and subsequent precipitation of Cr(III) is instantaneous [19]. The factors affecting Cr(VI) immobilization are well-known from a theoretical standpoint. Some insight on Cr(VI) retention efficiency and kinetics exists for soils with a high content of organic matter under acidic conditions [18,25,26]. However, scenarios relevant for C&DW reuse are more likely characterized by subsoils that are poor in organic matter and in contact with highly alkaline leachates containing other anions potentially competing for sorption sites.

This study assesses the chromium retention capacity and kinetics of three subsoils under conditions relevant for reuse of C&DW in an unbound road base. The objectives were to: (i) assess the retention capacity of three typical subsoils with respect to Cr(VI) released by C&DW in realistic concentrations (ii) assess which processes control Cr retention, (iii) experimentally determine the retention kinetics, and (iv) simulate the migration of Cr(VI) in the subsoil.

## 2. Materials and methods

### 2.1. Materials

Three Danish subsoils from depths between 50 cm and 100 cm were used for the test. The soils were slightly acidic, with pH in 10<sup>-2</sup> M CaCl<sub>2</sub> between 4.4 and 6.4, and low soil organic matter (SOM) content (0.3–0.4%). Table 1 presents the main features of the soils (S1, S2, S3). S1 had the lowest pH, and S2 had the highest

SOM. S3 had the highest cation-exchange capacity (CEC) and clay content (see Table S1 in Supplementary data for further details).

Leachates from C&DW were obtained from shaking crushed (0–4 mm) C&DW with distilled water for 24 h. A standard batch test was used (EN 12,457 parts 1 and 2 [27,28]) at liquid-to-solid ratios (L/S) of 2 and 10 L·kg<sup>-1</sup> TS to obtain leachates with two different ionic strengths. The C&DW was sampled from full-scale C&DW recycling plants in Denmark investigated in an earlier study [6]. The leachates (pH ≈ 11) were spiked to different levels of Cr(VI) to represent typical high, medium, and low concentrations of Cr(VI) expected in leachates from crushed C&DW used in an unbound road-base layer: 500, 200, and 100 µg·L<sup>-1</sup>, respectively [7]. As a reference, Danish limit values for total Cr from utilization of residues in road construction correspond to 500 µg·L<sup>-1</sup> in restricted applications [29]. Table S2 (Supplementary data) shows the compositions of the leachates, together with the soil–leachate composition as measured after 48 h of contact time with each of the three soils.

### 2.2. Methods

#### 2.2.1. Experimental

Batch tests were carried out in PE bottles by mixing in full factorial design each subsoil with each of the six leachate types at an L/S ratio of 10 L·kg<sup>-1</sup> TS (65 g of soil to 650 ml of leachate). We performed all tests in duplicate, and in addition we ran 9 blank experiments, for a total of 45 bottles. Table S3 (Supplementary data) provides an overview of the test design. The bottles were rotated during the whole test, i.e., for approximately six months. Seven samples were collected from each bottle at specific times (0d, 2d, 10d, 30d, 80d, 130d, 180d). As a result of the samplings, the L/S ratio decreased approximately 15 percent during the experiment. Although care was used during the sampling to minimize atmospheric oxygen exposure, the experiments were not anaerobic. After filtration, Cr<sub>TOTAL</sub> was determined by ICP-OES (limit of detection, LOD: 1.5 µg·L<sup>-1</sup>). Additionally, cationic Cr was removed in a cation-exchange cartridge (Maxi-Clean 1.5 ml IC-H Alltech) as Ball and McCleskey describe [30]. At the existing pH conditions, all Cr(VI) was present in anionic form (chromate/dichromate), so we could subsequently determine Cr(VI) by ICP-OES (LOD 0.75 µg·L<sup>-1</sup>). Statistical evaluation of data (analysis of variances) was done using the computer software R.

#### 2.2.2. Cr reduction model

We hypothesized that Cr(VI) would be reduced by the soil and precipitate as Cr(III) on the soil particles. This would result in decreasing Cr(VI) concentrations in the liquid phase of the batch

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