Leaching of Heavy Metals from Contaminated Soil Stabilised by Portland Cement and Slag Bremen

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LEACHING OF HEAVY METALS FROM CONTAMINATED SOIL STABILISED BY PORTLAND CEMENT AND SLAG BREMEN

Abstract: Leaching behaviour is an important evidence of soil quality. The assessment of leaching of heavy metals from the contaminated soil is vital for environmental applications. However, leaching may differ in soil stabilised by various ratios of binders. In this study we measured leaching behaviour of soil contaminated by As, Cd, Co, Cr, Cu, Hg, Ni, Pb, V, Zn, methyl Hg, aliphatic compounds of hydrocarbons, polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs) and dissolved organic carbon (DOC). To evaluate leaching of these substances we tested the effects of changed amount of binder (120 kg and 150 kg) and binder ratios (70/30 %, 50/50 % and 30/70 %) added to soil samples. Soil was dredged from several stations in Ostrand area, SCA Sundsvall Ortvikens Pappersbruk. The results demonstrated a systematically decreasing leaching with the increased slag. The contribution of this research include: (i) devising systematic approach to extract information on leaching from stabilised soil collected from the coastal area of Bothnian Bay, (ii) developing a workflow for stabilising soils by various combination of Portland cement Basement CEM II/A-V (SS EN 197-1) and ground granulated blast furnace slag (GGBFS), Bremen type (SS EN 15167-1), (iii) determining water ratio and density for the untreated and stabilised soil and performing comparative analysis, (iv) evaluating chemical content of pollutants and toxic elements in the aggregated soil samples. Treatment of the contaminated soil by binders improved its parameters by the increased strength and decreased leaching of heavy metals and toxic elements.

Keywords: leaching, sediments, Baltic Sea, stabilisation, binder

Introduction

Solidification/stabilisation of soil by cement, cementitious products or other binders is one of the most representative geotechnical techniques in civil engineering aimed at improving properties and performance of soil [1-4]. Soil stabilisation consists in treatment of soil by stabilising agents aimed to increase its strength, maximise durability, stability and compression characteristics, as required by safety engineering standards. During the past decades, soil stabilisation has been applied to a wide range of areas in geotechnical engineering, from coastal environmental engineering [5], industrial works, such as highway construction [6] or remediation of contaminated groundwater [7], to agricultural application, e.g., simulating moisture ingress/digress into subsoils from rainfall events for modelling leaching cycles and field precipitation and moisture infiltration processes [8].

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Soil stabilisation can be performed using various methods and techniques [9, 10]. Various binders can be used as stabilisers for soil treatment, e.g., cement, cementitious by-products, slag, sludge, flying ash, lime, to mention a few [11]. Characteristics of the stabilised soil can be assessed by measuring its physical and mechanical properties, e.g., compressive strength, compaction, porosity, as applied in many geotechnical tasks [12-16].

Chemical stabilisation of soil is beneficial for leaching of the environmentally toxic elements. Examples of leaching from the stabilised soils include immobilisation of heavy metals [17, 18], rare earth elements [19, 20], or removing environmentally dangerous pollutants [21]. By stabilising organic soils with supplementary cementitious material, such as high carbon fly ashes, such treatment helps improving the geotechnical characteristics, performance and environmental properties of soil [22-24]. An important aim of leaching tests is to see if treated soil is acceptable, i.e., to examine environmental effects, impacts and consequences of use in construction applications in civil engineering [25-28]. Contamination of soil by pollutants may be caused by various chemical elements, e.g., compounds of hydrocarbons, polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs). PAHs originate from the anthropogenic activities: industrial emissions, petroleum combustion, emissions from coal and fossil fuels and domestic activities [29].

Soil stabilisation performed using binders enables to remove dangerous chemical elements originating from groundwater pollutants, mostly generated by the industrial, agricultural, and urban processes or stormwater discharge [30]. Although the existing methods of soil stabilisation, either using cement or other agents, are effective in many geotechnical applications for improving soil quality, they have limitations. The first limitation is the type of binder which has the effects on soil stabilisation [31]. The second one is the ratio of binders used in various proportions. For instance, various binder combinations may have different effect on soil treatment as to make them more or less prominent, which requires an empirical testing of the optimal ratio of binders [32-34]. Optimal ratio of binders obtained from a series of tests enables to define the best practical setting for chemical leaching of toxic and dangerous elements from the contaminated soil.

To address the aforementioned limitations of leaching performance in stabilised soil, we present a series of the experiments performed on various combinations of stabilising agents selected in different combinations. Two binders were used in this study: cement Basement *CEM II/A-V* (SS EN 197-1) [35] and ground granulated blast furnace slag (*GGBFS*), Bremen type (SS EN 15167-1) [36]. We tested 3 ratios of cement/slag to evaluate their effect on leaching: 70/30 %, 50/50 % and 30/70 %. Our analyses are based on the three aspects:

1. First, the properties of binders were modified by changing their ratio: adding more Portland cement and decreasing amount of slag, and vice versa. This makes out the results flexibly applicable in case where the amount of binder varies.
2. Second, the amount of leached chemical elements was quantitatively examined in all the three cases of soil stabilisation. The results of leaching are summarised in plots and tables for each element: As, Cd, Co, Cr, Cu, Hg, Ni, Pb, V, Zn, Methyl Hg, aliphatic compounds of hydrocarbons, polycyclic aromatic hydrocarbons (PAHs), and polychlorinated biphenyls (PCBs).
3. Third, water ratio and density for untreated and treated (stabilised) soil were evaluated for several test sample stations: A4, B4, B6b and B6B. Besides, for each of these tests, the amount of binder added into the soil mass changed (120 kg and 150 kg,

respectively). This enabled to assess both characteristics: local properties of soil in the test sites and the effects of the amount of binder on the properties of the stabilised soil.

Experiment

This section describes the experiment workflow and presents the research scope and objectives. Sampling methods performed by the Sundsvall Ortvikens Pappersbruk (SCA) including the description of binders and workflow of specimen processing is included with provided details regarding the environmental and technical aspects of leaching experiments.

Research scope and objectives

The research has been performed on behalf of the SCA paper mill by Swedish Geotechnical Institute (SGI). The data were collected in the Sundsvall region, located in coastal area of Bothnian Bay of the Baltic Sea, Sweden (Fig. 1).

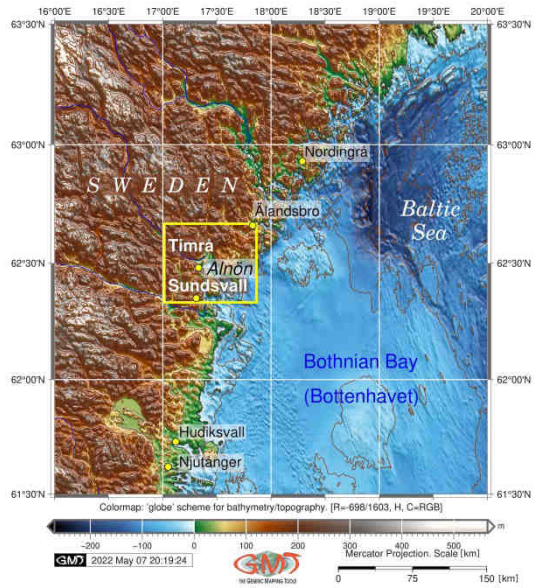

Fig. 1. Topographic map showing study area in the coastal area Bothnian Bay, Sweden. Data: GEBCO DEM. Sample locations of taking soil samples in Sundsvall area are from the test sites A4 and B4 within the location of SCA (62°24′0″N, 17°19′0″E). Cartographic source: authors' own map

The main goal was to evaluate how the choice of binder ratio affects leaching of chemical elements in soil samples. The practical objective to evaluating how the binder combinations changes leaching of toxic elements from soil: heavy metals (As, Cd, Co, Cr, Cu, Hg, Ni, Pb, V, Zn), methyl Hg, aliphatic compounds of hydrocarbons, PAHs, PCBs as well as *BTEX* chemical compounds (benzene, toluene, ethylbenzene and xylenes). The geochemical and environmental properties in soil samples were evaluated through technical processing of stabilisation.

The general purpose of this work was to analyse soil performance on leaching of heavy metals by changing combination of binders. The experiments were performed according to

the geotechnical and environmental quality standards of soil. The practical aim was to achieve the improved soil properties: strength and stability. Stability, i.e., settlement in the stabilised soil should not exceed 5 cm after the two years from stabilisation. The quality objective regarding the permeability constituted geotechnical and environmental requirements. The permeability should be below 10^{-8} m/s when finished structure can be considered as a monolith with low water permeability. This reduces the risk of leaching of the contaminants and shortens service life due to weathering or corrosion.

Soil sampling by SCA

The SCA was responsible for collecting representative soil samples of the dredged sediments and groundwater that were used as leachate in experiments. The location of the study area and sampling points are shown in Figure 1. The sampling was made during June 2017 fieldwork using a self-developed sampler, a so-called "Jonhuggare", developed by Sweco in the test point B6b. The test points were selected because of the specifics of the soil in this place: high pollution of sediments and a high content of mesa and fibre. The reason for choosing samples containing mesa/fibre was that laboratory experiments needed to be performed using samples which composition and structure represent natural conditions, i.e., sediments consist not only naturally occurring materials but also external inclusions. Eight different samples were taken at different bottom depths (0, 100) cm. The samples were put into the 10 L buckets and sent to the SGI. The groundwater was extracted on 6 February 2018 in a test point GVV1 by Sweco, a total of approximately 5 litres. When sampling groundwater, the pH level was evaluated, and the electrical conductivity (*EC*) was 0.43 mS/cm.

Binder

The following binders were chosen for soil tests in SGI:

- Portland cement Basement type *CEM II / A-V* (SS EN 197-1) [35].
- Ground granulated blast furnace slag (*GGBFS*), type Bremen (SS EN 15167-1) [36].

The reason of selecting these two binders is that both of them (or comparable ones) are available on the commercial market for purchase and qualify for geotechnical tests. Both binders have been successfully used in similar previous projects in the SGI, including the Arendal geotechnical works on soil in the port of Gothenburg. Besides, Portland cement *CEM II* / *A-V* replaces previously used Byggcementen *CEM II* / *A-L*.

Manufacturing specimens

The specimens were manufactured in the laboratory of SGI from the dredged soil sampled by SCA. The material was stored in a climate room with a temperature of 7 °C. The aggregated sample selected from the B6b site was divided into the two barrels with tight-fitting lids. After that, one of the barrels was mixed with more added water, to simulate higher water ratio. Afterwards, specimens were mixed and thinned by mechanical stirring for 5 minutes. This enabled to receive a homogeneous mixture of binder and soil.

After mixing, soil was poured into the sampling sleeves with a lid attached to the bottom. The sleeves had a diameter of ca. 50 mm and a height of ca. 170 mm. During "casting" of specimens, the sleeve was tapped into the table to loosen the trapped air pores. The sleeves were filled up, then placed in a water bath. The water level in a bath was kept constantly above the upper edge of sleeves and the excess water formed during mixing was

recorded. The samples were stored in a water bath for 28 days, afterwards, extruded out of the sleeves and trimmed to the correct length to 100 mm. The specimens were stored in a plastic bag in a climate-controlled room with a temperature of (20 ± 1) °C. An amount of paper moistened with water was placed in a plastic bag, to prevent specimens from drying. The curing process was ensured according to the SIS standards [37-41]. A compressive strength was evaluated before leaching according to the SIS standards [42].

Laboratory stabilisation included weighing the required amount of soil to produce the two test specimens. The weighing took place on a scale with a measurement accuracy of 0.1 g. The permissible tolerance was set to the target weight ± 0.25 %. The binders were weighed up. Experimental changes in binder amount have practical reasons for geotechnical works. Thus, an optimal amount of binder that can provide a sufficient quality for soil stabilisation. Besides, lower amount of binder is favourable for economic and environmental reasons. These include low cost production and reduced CO_2 emissions related to the use of cement. A scale with a measurement accuracy of 0.01 g was used. Each mixture consisted of soil for two specimens. The specimens were divided into the A and B samples, where A were used for geotechnical measurements of compressive strength, while B - for the environmental and chemical experiments on leaching. This procedure was repeated to obtain duplicate experiments. Leaching properties of the contaminated soil from the scaled-up experiment demonstrated agreement with the laboratory experiment.

Leaching tests

The leaching properties of the stabilised material have been tested using three different forms of leaching tests:

1. one-step shaking test L/S 10 according to SS-EN 12457-2 (Phase 1 and 2) [37];
2. surface leaching tests according to standard SS-EN 15863 (Phase 1) [38, 39];
3. reduced surface leaching experiments that were modified from standard to be adapted to the project's issues (Phase 1).

Table 1

Research questions, purpose of work and setup of leaching test experiments

No.	Question	Purpose	Leaching test
1	How is the diffusion-controlled leaching of pollutants from the surfaces affected depending on chosen proportions?	Finding the best solution for binder ratios with as low leaching as possible	Six (6) reduced surface leaching trials with water withdrawal on days 9 and 36. The 3 ratios in two different scenarios ("worst case" and "best case") were tested. A surface leaching test with seven of the eight-time steps* according to the standard SS-EN 15863 [39]
2	Which equilibrium concentrations of binders affects the material and how do they relate to the chemical composition of the local groundwater?	Worst-case assessment of which pore water levels may arise	A one-step shaking test at leaching solution to solid material (L/S) = 10 according to the SS-EN 12457-2 [37], leachate MilliQ
3	Do differences arise in the equilibrium chemistry depending on which water is in contact with the material? How is the pH value of the leachate affected by contact with natural water?	Guidance on interpretation of uncertainties in the lab results in lab scale versus pilot scale	A one-step shaking test at L/S 10 according to SS-EN 12457-2 [37], leachate groundwater from the area
4	Expected leaching according to Phase 1 is affected by the optimisation in Phase 2 of the experiment	Support for final recommendation of ratio of binders	Three one-step shaking attempts at L/S 10 according to SS-EN 12457-2 [37]

*The last time step at 64 days was deselected due to the lack of time

The majority of the leaching experiments have been done using chemically pure water as a leaching liquid (MilliQ). Another test run in parallel was made for the leachate consisted of groundwater from the area for the large-scale application. Table 1 shows the test setup and describes the issues that form a basis for each test series. The emphasis was set on surface leaching tests. The environmental technical quality requirement was that a permeability should be lower 10^{-8} m/s. Shake tests on crushed soil do not reproduce expected leaching mechanisms, i.e., leaching from surfaces where the soil is a monolith.

Workflow of leaching experiments

Table 2 describes the scope of the analysis.

Table 2

Summary of leaching methods and scope of analysis in the environmental technical evaluation

Leaching method	Scope of analysis
Reduced surface leaching trials with water withdrawal 9 and 36 days. Modification of the standard SS-EN 15863 [39]	Heavy metals incl. Hg, DOC at both time steps
Surface leaching tests with seven of eight-time steps according to the standard SS-EN 15863 [39]	Heavy metals incl. Hg, DOC in seven-time steps
One-step shaking attempt at L/S 10 according to SS-EN 12457-2 [37]	Heavy metals incl. Hg, DOC, *BTEX*, PCB, PAH 16 Mineral oil (C10-C40)

Various equipment and analytical methods exist for determining heavy metals in soil. These include, for instance, Inductively Coupled Plasma - Mass Spectrometry (ICP-MS), Atomic Absorption Spectroscopy (AAS), Gas Chromatography - Mass Spectrometry (GC-MS). In this study the leached elements were evaluated according to the SGI technical standards using the ICP-MS as the most optimal for the estimation of heavy metals [42, 43]. For the majority of elements (As, Cd, Co, Cr, Cu, Ni, Pb, V, Zn) we used the existing ISO 17294-2:2016/SS-EN ISO 17294-2:2016 standard [45] for determination of chemical elements and heavy metals in soil and assessing water quality. The Hg was assessed using standard ISO 17852:2006 / SS-EN ISO 17852:2008 [46] for determination of mercury using atomic fluorescence spectrometry. The determination of dissolved organic carbon (DOC) was performed using standard SS EN 1484:1997 [47].

Results

The results of this study based on soil sampling performed in the laboratory of SGI include the assessment of surface leaching of heavy metals from soil for the worst and best cases and evaluating the effects from binder/water ratio measurements on performance of the stabilised soil. The shake tests were made for evaluating the equilibrium concentrations of heavy metals in stabilised soil.

Water ratio and density evaluated in samples processed in laboratory of SGI

Soil samples collected from testing sites A4 and B4 in Ostrand area were mixed to a homogeneous mass by stirring in the laboratory of SGI. This was done to minimise the effects from the sedimentation that occurs during transport. The water ratio of the material was determined upon the completion of the homogenisation process (Fig. 2).

Figure 2a shows water ratio for the untreated soil from test points A4 and B4. For the sample collected in site A4, the mean value for 8 determinations of the water ratio was about 135 % (excess) and the standard deviation was 0.83. The equivalent for the test point B4 was a water ratio of about 69 % and a standard deviation of 0.27. Test point B6b is included as a connection to the original laboratory tests. The subplot in Figure 2b shows the density of the untreated soil from test points A4 and B4. For sample A4, the mean value for 8 tests of density was 1.36 Mg/m^3 (Mg = 10^6 g = tonne) and standard deviation 0.0052. The equivalent for test point B4 was a density of ca. 1.6 Mg/m^3 and standard deviation 0.0053. Test point B6b was included as a connection to the original laboratory tests.

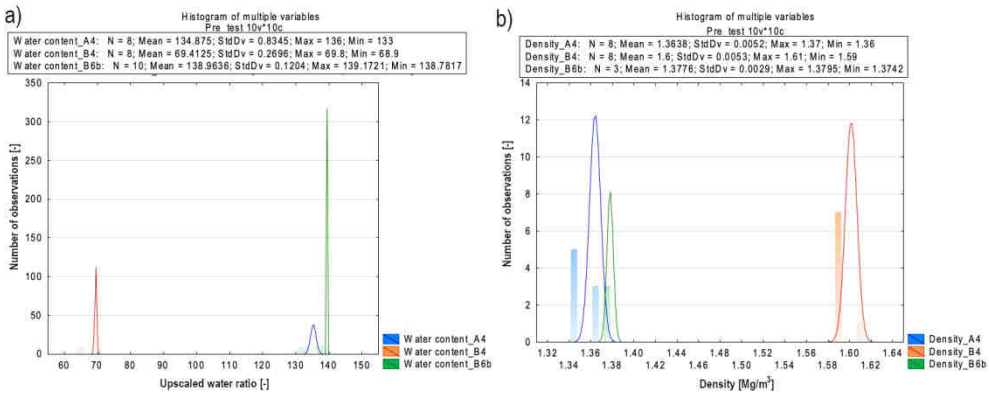

Fig. 2. a) Upscaled water ratio for the untreated soil (A4 and B4), b) density of the untreated soil (A4 and B4)

Water ratio between the samples was large, but for material from the site A4 it had the same ratio as for materials from B6b, as included previous laboratory experiments. The density shown the same pattern in case of water ratio: samples collected from the site A4 had the same value of range as those from the site B6b. The differences between the samples A4 and B4 demonstrated the same pattern for the water ratio. The difference between the masses from various test points includes density and water ratio. Soil collected from the sampling points in the scaled-up experiment cover samples from the point B6b. After sampling the contaminated soil samples were stabilised.

Water ratio and density evaluated in stabilised sediments

The stabilised samples were stored in the SGI in Linköping for curing and testing. Soil samples were processed under water to simulate field conditions. Density and water ratio of the specimens of stabilised soil were assessed (Fig. 3). The results of the B6b were taken from the SGI laboratory report. The results of the scaled-up experiment show the expected results between the samples from A4 and B4, i.e., higher strength for B4 with lower water ratio. The results also showed that performance of samples from B4 site have a good agreement with the results from laboratory tests. The difference in water ratio in samples from site B6b, stabilised by 120 vs 150 kg of binder, was evaluated to study the effects of large water mixture (worst case). Testing B6b in a laboratory was done by ratio *HWLB*.

Figure 3a shows water ratio of the stabilised soil collected from sites A4 and B4. Samples from A4 show that 120 kg of binder gives a slightly higher water ratio, compared

to those containing 150 kg of binder. The results from the test site B4 show a deviating pattern of values, however, in general, the values are within normal distribution range. The samples within each group were treated, processed and analysed for density. The density of the stabilised specimens follows the same pattern as for the water quotas (Fig. 3b).

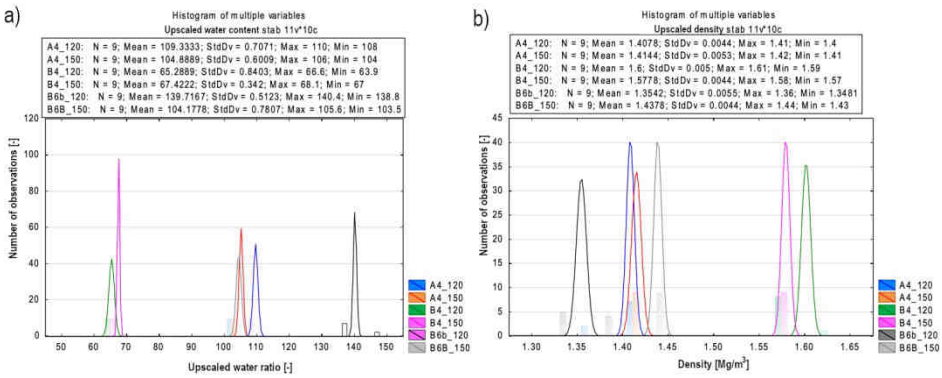

Fig. 3. a) Upscaled water ratio in stabilised soil (A4 and B4), b) density in stabilised soil (A4 and B4)

The samples from B4 (lowest water ratio) have the highest density. The differences between the B6b_120 (ratio *HWLB*) and B6b_150 (ratio *LWHB*) are a "forced" spread to provide greater variation in data material. The differences in the results from scaled-up laboratory experiment are caused due to a forced increase in water ratio. The difference in density is based on the soil samples: those from point A4 are well within the limits of the experiment, while those from B4 are higher due to a large difference in water ratio.

Parameters of sediments collected in Ostrand, Sweden

After evaluating density and water ratio, the amount of binder was determined. The selected binder mix was the same as in the laboratory, i.e., 30 % of Portland cement Basement *CEM II/A-V* (SS EN 197-1) [35] and 70 % of *GGBFS* [36]. The amounts of binder were 120 and 150 kg/m^3 of soil. The ratio of water to binder (*w*/*b*) is the parameter that controls both strength and permeability. Table 3 shows parameters of various sub-samples.

Table 3

Summary of the constituent parameters of the specimens

Test point	Water quota [%]	Density [Mg /m^3]	Amount of binder [kg/m^3]	*w/b*
A4	135	1.36	120	7.51
A4	135	1.36	150	6.21
B4	69	1.6	120	6.44
B4	69	1.6	150	5.35

A certain amount of dredged masses was taken out from the homogenised masses and mixed with binder and water in various proportions. The water was added to simulate the

wetting method, i.e., binder is mixed wet with the dredged masses, in a similar way as was previously done in Arendal 2 project in Gothenburg.

The mixing took place with 350 L of the forced mixer. From this mixture, a 120 L plastic barrel was filled with the stabilised dredged soil. The temperature sensors in form of thermocouples were placed in the samples to measure variations of heat and air temperature on the three levels. Test specimens were cast into sampling plastic sleeves. In total, 60 specimens were made, including 15 of each variant of mix ratio. The specimens were marked and transported to the SGI for storage, processing, and testing.

Contamination level in the aggregate sample B6b

The aggregate sediment samples used for stabilisation included samples from the point B6b (up to 100 cm depth). Total levels of pollutants in the aggregates are reported in Table 4 with statistical data on the SCA's sampling in the area. The sample contained Hg which is frequently found in this area as well as a fraction of heavy aliphatic solvents, which caused a distinct oil odour when samples were homogenised in the SGI.

Table 4

Chemical content of contaminants in aggregated soil samples tested in the experiments (B6b).

Element/substance	Unit	SGI's lab tests	Samples in double pontoon, contaminated sediments (0 - ca. 2.5 m)			
		B6b	No. of samples	Average	Median	Max.
As	mg/kg d.m.	1.5	24	4.9	4.2	12
Cd	mg/kg d.m.	1.3	24	0.25	0.12	1.5
Co	mg/kg d.m.	2.6	24	8.8	9.3	15
Cr	mg/kg d.m.	27	24	43.1	36.4	187
Cu	mg/kg d.m.	29	24	48.4	20.6	351
Hg	mg/kg d.m.	5.1	24	13.6	0.5	246
Ni	mg/kg d.m.	25	24	21.6	22.7	34
Pb	mg/kg d.m.	27	24	20.4	14.3	91
V	mg/kg d.m.	12	24	35.7	38.1	46
Zn	mg/kg d.m.	243	24	110	76.7	357
Methyl Hg	ng/g d.m.	1	10	6.9	1.7	25
PAH 16	mg/kg d.m.	0.32	17	3.1	0.8	24
PAH 11	mg/kg d.m.	0.32	17	3.3	1.5	19
PCB sum 7	mg/kg d.m.	0.03	10	0.02	0.0014	0.11
Aliphatic C10-C12	mg/kg d.m.	22	10	20	20	20
Aliphatic C12-C16	mg/kg d.m.	42	10	20	20	20
Aliphatic C5-C16	mg/kg d.m.	64	10	20.8	24	24
Aliphatic C16-C35	mg/kg d.m.	366	10	56.3	20	165
d.m. 105 °C*	%	53	24	59.9	64.6	79.5
Remains after LOI **	% of d.m.	94	15	95.2	96	98.2
TOC	% of d.m.	6.2	15	1.5	1.6	3.9

* In Tables 4 and 5, the d.m. is a dry matter. Respectively, the 'd.m. 105 °C' indicates that it was determined at 105 °C in the oven

** LOI (Loss of Ignition) is a test used in this study for the analysis of chemical and mineral makeup of soil using applied methods of inorganic analytical chemistry (reported in Tables 4 and 5)

The aliphatic content was much higher in the aggregate sample B6b compared to average and median levels in the dredging area. The comparative analysis of chemical

content for aggregate samples in the tests in B6b with analysis of samples prepared for stabilisation and modified using table from Sweco 2018, is presented in Tables 4 and 5.

Table 5

Chemical content of contaminants in soil samples prepared for stabilisation

Element/Substance	Unit	SGI's lab tests	Samples between the double pontoon and shoreline, contaminated sediments (0 - ca. 2.5 m)			
		Point B6b	Antal prover	Average	Median	Max.
As	mg/kg d.m.	1.5	25	3.4	3.7	11
Cd	mg/kg d.m.	1.3	25	0.8	0.6	2.8
Co	mg/kg d.m.	2.6	25	5.5	3.6	14
Cr	mg/kg d.m.	27	25	68	36.8	377
Cu	mg/kg d.m.	29	25	34.1	27.9	107
Hg	mg/kg d.m.	5.1	25	13.5	3.2	155
Ni	mg/kg d.m.	25	25	23.4	24.3	41
Pb	mg/kg d.m.	27	25	28.7	16.9	149
V	mg/kg d.m.	12	25	23.3	17.9	49
Zn	mg/kg d.m.	243	25	171	151	537
Methyl Hg	ng/g d.m.	1	23	8.6	1.2	67
PAH 16	mg/kg d.m.	0.32	16	6.4	1.0	35
PAH 11	mg/kg d.m.	0.32	16	5.2	1.0	24
PCB sum 7	mg/kg d.m.	0.03	16	0.04	0.0049	0.15
Aliphatic C10-C12	mg/kg d.m.	22	16	20.5	20	22
Aliphatic C12-C16	mg/kg d.m.	42	16	25.5	20	42
Aliphatic C5-C16	mg/kg d.m.	64	16	38.3	20	64
Aliphatic C16-C35	mg/kg d.m.	366	16	117.5	42	366
d.m. at T = 105 °C	%	53	25	50.8	54.4	73.2
Remains after LOI	% of d.m.	94	16	94.7	94.7	98.1
TOC	% of d.m.	6.2	16	2.4	2.2	6.2

Surface leaching after 36 days: worst and best case

Water sampling was done at 9 and 36 days of surface leaching experiments. Table 7 compares the results from the upscaling experiment against the results for two corresponding recipes in the feasibility study. Figure 4 shows leaching of substances with a clearly reduced trend with a higher slag content after 36 days of leaching. No support is given for the fact that the worst case, *HWLB* ratio, gives a higher risk of leaching compared to the best case *HBLW* ratio.

There is no environmental benefit in advising recipes with excessively high amounts of binder, as long as the quality requirement of 10^{-8} m/s is satisfied. Figure 5 shows leaching for substances showing neutral or increasing trend with the increased content of slag.

In general, we noted that higher amount of binder increases leaching of heavy metals, which shown similar results in the upscaling experiment compared to the standard test. However, various effects were noted for different elements. For example, Ba is a possible indicator of leaching in the stabilised soil, while Ni cannot be considered for that. Besides, a higher slag content leads to the increased leaching of As and Cr, although 70 % of slag resulted in the lowest permeability. Different behaviour of As and Cr compared to other heavy metals may indicate that leaching is affected more by surface chemistry itself than by the permeability. Leaching of Hg was low, so that it was not detected in the leachate.

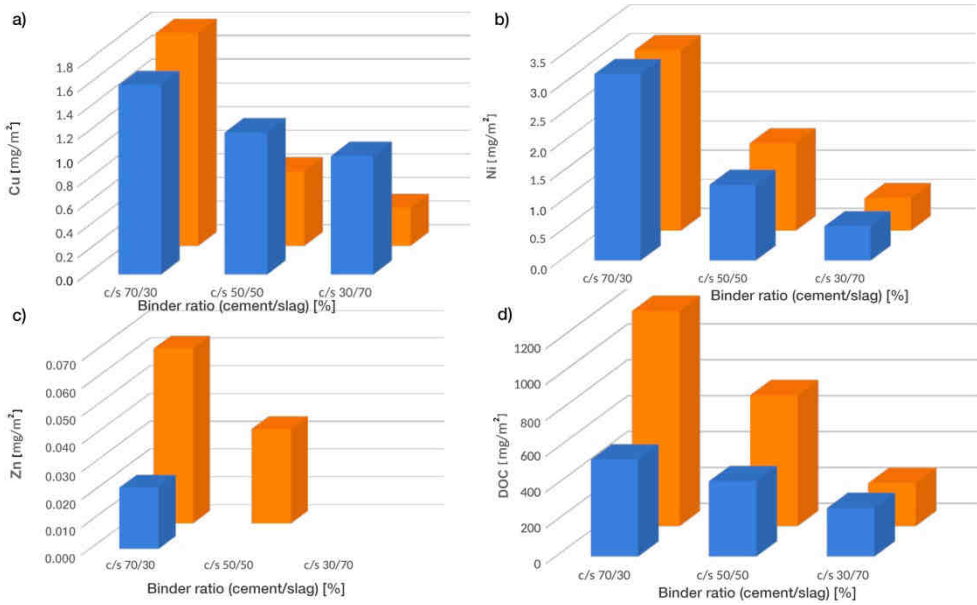

Fig. 4. Summary of the leached amount per unit area for: a) Cu, b) Ni, c) Zn and d) DOC on the day 36th for the best and worst cases. Orange colour - best case *HBLW* (High binder-Low water ratio in a stabilising mixture). Blue colour - worst case *HWLB* (High water-Low binder ratio in a stabilising mixture). Substances show a systematically decreasing trend with the increased slag content. Variations in 3 cases of bars show ratio cement/slag as 70/30 %, 50/50 % and 30/70 %

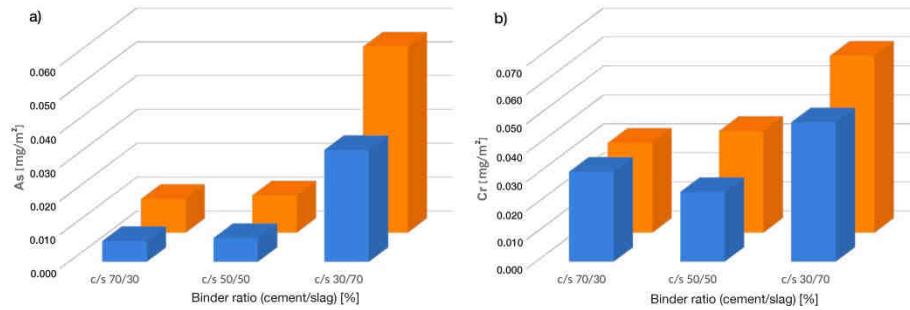

Fig. 5. Summary of leached amount for: a) As and b) Cr on day 36th for the best and worst cases. Orange colour - best case *HBLW* (High binder-Low water ratio in a stabilising mixture). Blue colour - worst case *HWLB* (High water-Low binder ratio in a stabilising mixture). Substances show a non-existent or increasing trend with increased slag content. Variations in the 3 cases of bars show proportions of cement/slag in binder ratio as 70/30 %, 50/50 % and 30/70 %

Table 6 shows the amount of leached elements at water outlet after day 36. Variation between the lowest and highest leached amount of heavy metals is relatively small, which is consistent with the results on soil permeability. Regardless of mixture of binder ratio and variations in natural content of tested samples, soil properties shown low leaching. The results from the scaled-up tests show agreement with those from the laboratory experiment. They show that it is possible to carry out stabilisation on a larger scale in a safe

and controlled way to achieve stabilised soil cleaned from contaminants. Tested soil samples shown strength above 140 kPa. The results of the permeability tests showed a clearly lower values than 10^{-8} m/s. The leaching of the scaled-up test correlated with the laboratory test.

Table 6

Leached amount at water outlet after 36 days [mg/m^2] in the upscaling experiments (a total of 12 samples). Comparison is made against the two corresponding mixes in the standard test: cement/slag in ratio 30/70 % and high and low amount of binder combined with the low and high water ratio

Element (heavy metal, DOC)	Min. amount per outlet upscaling [mg/m^2]	Max. amount per outlet, upscaling [mg/m^2]	Min. amount per outlet, pilot study [mg/m^2]	Max. amount per outlet, pilot study [mg/m^2]
As	0.01	0.05	0.03	0.06
Ba	1.5	7.7	0.8	1.1
Pb	< 0.0007	0.002	< 0.00081	< 0.00082
Cd	< 0.0003	< 0.0003	< 0.0003	< 0.0003
Co	0.01	0.01	0.02	0.02
Cu	0.2	0.3	0.5	1
Cr	0.02	0.04	0.05	0.06
Hg	< 0.007	0.02	< 0.008	< 0.008
Mo	0.06	0.2	0.1	0.1
Ni	0.09	0.2	0.6	0.6
Zn	0.3	0.4	< 0.02	< 0.02
DOC	160	610	240	270

Table 7 shows the leached content at L/S 10 in the upscaling tests (4 samples in total). Comparison is made against the 3 corresponding mixes in a standard test: cement/slag in ratio 30/70 % and high/low amount of binder combined with low/high water ratio.

Table 7

Leached content at L/S 10 in the upscaling experiments (4 samples in total). Comparison is made against the 3 corresponding mixes in a standard test: cement/slag in ratio 30/70 % and high/low amount of binder combined with low/high water ratio

Element (heavy metal, DOC)	Groundwater	Lowest leaching upscaling at L/S 10 [µg/L]	Highest leaching upscaling at L/S 10 [µg/L]	Lowest leaching feasibility study at L/S 10 [µg/L]	Highest leaching feasibility study at L/S 10 [µg/L]
As	18.6	0.7	1.1	0.5	0.59
Ba	4.1	160	520	320	360
Pb	< 1.0	0.15	0.28	0.1	0.24
Cd	< 0.5	< 0.004	< 0.004	< 0.004	0.004
Co	< 0.5	3.7	4.2	1.7	4.8
Cu	< 1.0	31	68	3.4	64
Cr	< 5.0	0.59	1.0	0.12	1.1
Hg	< 0.010	< 0.1	< 0.1	< 0.1	0.15
Mo	12	25	29	19	22
Ni	3.1	40	52	140	210
V	< 5.0	3.6	4.6	29	33
Zn	< 2.0	< 0.2	0.75	0.52	0.74
DOC	7 790	18 000	38 000	24 000	120 000

The pollution content in the analysed excess water was higher than those stated as reference values for purified construction stormwater for Hg and in some samples for Cr

and Ni according to condition 9 in a judgment of the Land and Environment Court at Ostersund District Court on 30.09.2020, case M757-19. However, the analysis of the surplus water shown that no additional protection measures are needed compared with those described in a plan for controlling and managing water that arises during the construction phase of the project. Hence, it is estimated that there will be relatively limited amounts of contaminated sediments that need to be stabilised as well as the amount of excess water that could be formed from this treatment. In general, a large part of the excess water within the reinforcement line is estimated to be bound in the landfill.

Discussion

In this study, the dynamics of leaching of heavy metals, PAHs and PCBs from contaminated soil was assessed with a case study of Sweden. The tests were performed using existing standards of SIS for assessment of leaching behaviour of contaminated soil. The summary of the leached amount per unit area [mg/m^2] for Cu, Ni, Zn and DOC and comparisons between the best (*HBLW*) and worst cases (*HWLB*) of different binder ratios are depicted in Figure 4, while leached amount per unit area [mg/m^2] for As and Cr - in Figure 5, respectively. The results are discussed below in terms of leaching of the contaminants affected by two factors: (i) the effects of changing the binder ratio (cement or slag) used for stabilising soil samples and (ii) the effects of the composition of the stabilising agent regarding water/binder ratio: best case (*HBLW*) and worst case (*HWLB*).

The analysis of the differences between the leaching cases of heavy metals and toxic substances, can be supported by the comparison of Table 6 and 7 which show that leaching content [µg/L] in samples is quite different for test showing variations in highest and lowest leaching. This indicates that changes in high/low amount of binder combined with the low/high water ratio play a key factor for the leaching of heavy metals from soil. This proves that the optimal ratio and content of binder is a major factor for treatment for contaminated soil, as also noted in previous existing studies [48-51].

We have observed from the inspection of the graph (Fig. 5), that in case of As and Cr leaching from the soil samples, leaching show a non-existent or increasing trend with increased slag content, so that these elements cannot be influenced by the added slag content which was assessed in cases of binder ratio as 70/30 %, 50/50 % and 30/70 % by gradual change of stabilising agent added to the mix. Though *GGBFS* slag Bremen affects leaching in the most cases, there are notable differences in leaching of As and Cr compared to the Cu, Ni, Zn and DOC that, on the contrary, demonstrated a systematically decreasing trend with the increased slag content. This is a consequence of the concentrations of anions, time of treatment, sorption properties and matrix minerals, as noted earlier [52-54].

Leaching of Cu may result of local corrosion and its differences against other heavy metals could be accounted for pH shifts from microbial metabolism, as noted earlier [55]. The source of DOC leaching can originate from asphalt released by rainfall wash and runoff, which results in potentially environment influence by transferring hazardous matters into aqueous and soil system [56]. However, the concentrations of metals may differ with depth, according to recent geochemical studies on elements behaviour [57]. In our experiments, leaching of DOC demonstrated variations in best and worst cases thus controlled by the binder/water ratio. Leaching of Zn may be affected by the different physicochemical properties of the soils in testing sites (test sample stations: A4, B4, B6b and B6B). Besides, the variations can be associated with the micronutrient source that may

influence Zn leaching and its distribution among soil fractions in the depth of the layers, which agrees to the previous works [58].

Conclusion

The research revealed a clear effect of increased slag content in stabilised soil mixture on the quantity and quality of leaching of the heavy metals, PAHs and PCBs accumulated in the contaminated soil. Thus, increase content of slag up to 70 % decreased leaching, which can reduce risk of negative environmental impacts from heavy metals. Besides, in the soil stabilised with cement and slag, in relation to the samples where stabilising agents were added in various ratios to the contaminated soil, an effect of the ratio between the binder components on leaching was found. At the same time, we did not find enough evidence if ratio between water and amount of binder, significantly affect leaching. The permeability of soil was characterised by a decrease along with the increased slag content.

At the same time, changes in water and binder ratio in a stabilised mixture affected soil permeability. Moreover, we noted that dredged sediments can be homogenised even if soil contains elements of mesa and fibre. Furthermore, it was found that the external inclusions did not affect the soil properties with regard to the geotechnical requirements of its quality. Amount of binder added as stabilising agents showed the effects on strength properties of soil which increased proportionally with the slag content. Accordingly, stabilising soil by a proper amount and ratio of binding agent (cement/slag) improves bearing capacity of soil which satisfies technical requirements and standards of construction works.

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