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Soil contamination from heavy metals and persistent organic pollutants (PAH, PCB and HCB) in the coastal area of Västernorrland, Sweden

1. Introduction

1.1. Background

The assessment of soil contamination is a widely recognized environmental task in which one needs to evaluate the level of concentration of the dangerous particles in a soil mass. The detection of pollutants in soil includes measuring the concentrations of toxic elements and heavy metals (e.g. Cu, Zn, Ag, As, Pb, Z, V, Cd, Co, Cr, Hg, Mo), chemicals (Polycyclic Aromatic Hydrocarbons, PAH), organochlorine compounds, (Polychlorinated Biphenyls, PCB)

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and Hexachlorobenzene (HCB). Existing studies report heavy-metal pollution in various regions of Sweden (Johansson et al. 1995) and the Baltic Sea (Leivuori and Niemistö 1995; Renner et al. 1998; Vaalgamaa and Conley 2008; Vallius 2014; Zalewska et al. 2015) revealed on the basis of the chemical analysis of sediment samples.

Persistent organic pollutants (POPs), such as PCB, HCB and PAH, are toxic chemicals of various sources (Achten and Hofmann 2009; Gabryszewska and Gworek 2021) that negatively affect the environment. The main sources include industrial waste, power plants, transport emissions, municipal activities, agricultural waste and combustion (Lima et al. 2007). Due to the transportation cycle through the water/soil system, contaminants can be tracked in regions located far beyond the source of origin (Zaborska et al. 2011).

The concentrations, composition patterns and transport of chemicals and POPs in soil are highly complex (Bandowe et al. 2021), which makes them some of the most harmful sources of contamination and environmental pollution. For instance, PAH are resistant to environmental degradation due to their highly hydrophobic nature (Gan et al. 2009) and their widespread introduction to the environment through motor vehicle exhausts (Morillo et al. 2007). PCB adsorbs to soil as waste particles originated from industrial depositions, incinerators and biomass combustions (Kumar et al. 2014). Thus, the analysis of PAH and PCB concentrations in the bottom sediments is used as an indicator of ecosystem health (Grochowska et al. 2021).

Heavy metals, being chemical elements with high density that are toxic and difficult to decompose, easily accumulate in the marine biota. This can severely impair the environmental sustainability of coastal ecosystems (Siregar et al. 2020). The accumulation and distribution of heavy metals in marine sediments is strongly controlled by the origin of contaminants, specifically industrial activities, organic wastewater from urban discharge as the major sources of pollution (Chabukdhara and Nema 2012; Feng et al. 2012). Aside from this, it is affected by geochemical parameters, such as soil-grain size (fine grained/ /coarse grained) and the depth of the water column (White and Tittlebaum 1985), organic soil matter, soil acidity and altitude (Łyszczarz et al. 2020). Moreover, hydrological parameters affecting heavy-metal concentrations in sediments include the upstream location, river currents, depth, the strength of waves and reservoir shape (Smal et al. 2015).

1.2. Research objectives

The environmental assessment of contaminated soil prior to civil construction projects is based on soil sampling, methods of implementation, analysis and interpretation (Wong and Li 2003). Following such a research scheme, our study combined several tests for evaluating chemical and heavy-metal concentrations and leaching in soil stabilized by a different ratio of binders (120/150 kg per m^3) added to the dredged material. The trace analysis of heavy metals in the bottom sediments through leaching tests was performed using tools of the SGI and technical guidance of the Swedish Institute for Standards (SIS).

The motivation behind this study was to evaluate heavy-metal concentrations and leaching from soil samples stabilized using various binder/water ratios. The stabilization of soil and solid materials by binding agents is crucial for increasing compressive strength and improving the geotechnical properties (Lindh and Lemenkova 2021a; Bayraktar et al. 2015; Moghal et al. 2020; Lemenkov and Lemenkova 2021a, b). However, studying changes in the chemical contamination of soil treated by various binder ratios is omitted by most of the previous works (Lindh and Winter 2003; Ryden et al. 2006; Lindh and Lemenkova 2021b). Therefore, the analysis was carried out by plotting graphs in a way that visualizes variations of heavy-metal concentrations in soil samples stabilized by various binder combinations (cement/slag in various proportions and water content).

Practical objectives included geotechnical works undertaken by the Swedish Cellulose Company (Svenska Cellulosa Aktiebolaget, SCA) Biorefinery Östrand AB. Specifically, the purpose of soil tests was related to planned construction works which required the testing of soil to assess its geotechnical and environmental properties. The SCA received a permit in accordance with the Environmental Code for biorefinery construction from the Land and Environmental Court at the Östersund District Court by partial judgment on 2020.09.30, case M 757-19. The permit included Condition 2, which contains the requirements regarding the scaled-up stabilization/solidification (S/S) tests, as excerpted from the relevant part – before the full-scale S/S, the following properties of soil were tested: permeability (maximum 10^{-8} m/s), strength and leaching of heavy metals and contaminants. The S/S tests were conducted in the field at the planned business area on a scale larger than that in the previously performed laboratory tests using materials representative for the full-scale treatment of soil. To this end, a series of soil tests was performed in the laboratory of SGI on behalf of the SCA.

The aim of the study is to evaluate soil contamination and to assess the leaching of heavy metals, toxic elements and POPs. Soil samples were stabilized using existing methods (Lindh and Åberg 2017). The study objectives were to analyze the effects of binder ratios on leaching (best and worst cases). The tests included measuring the concentrations of heavy metals through leaching after the S/S of soil by different ratios of binders. The stabilizing agents included Portland cement and GGBFS. The experiment included sampling and the analysis of the excess water to soil, aimed at evaluating the effects of water content and soil surface area on the rheological properties of stabilized soil. The tests were performed on a larger scale at the site of the Timrå biorefinery to verify the tests on the leaching of heavy metals performed in a laboratory (Figure 1). The two sites were selected for sampling (A4 and B4). The scaled-up experiments were conducted on 24.06.2020 and followed the program developed in consultation with the County Administrative Board of Västernorrland, "Workflow for scaled-up stabilization tests of contaminated sediments, Biorefinery Östrand, SCA", 2020.05.12.

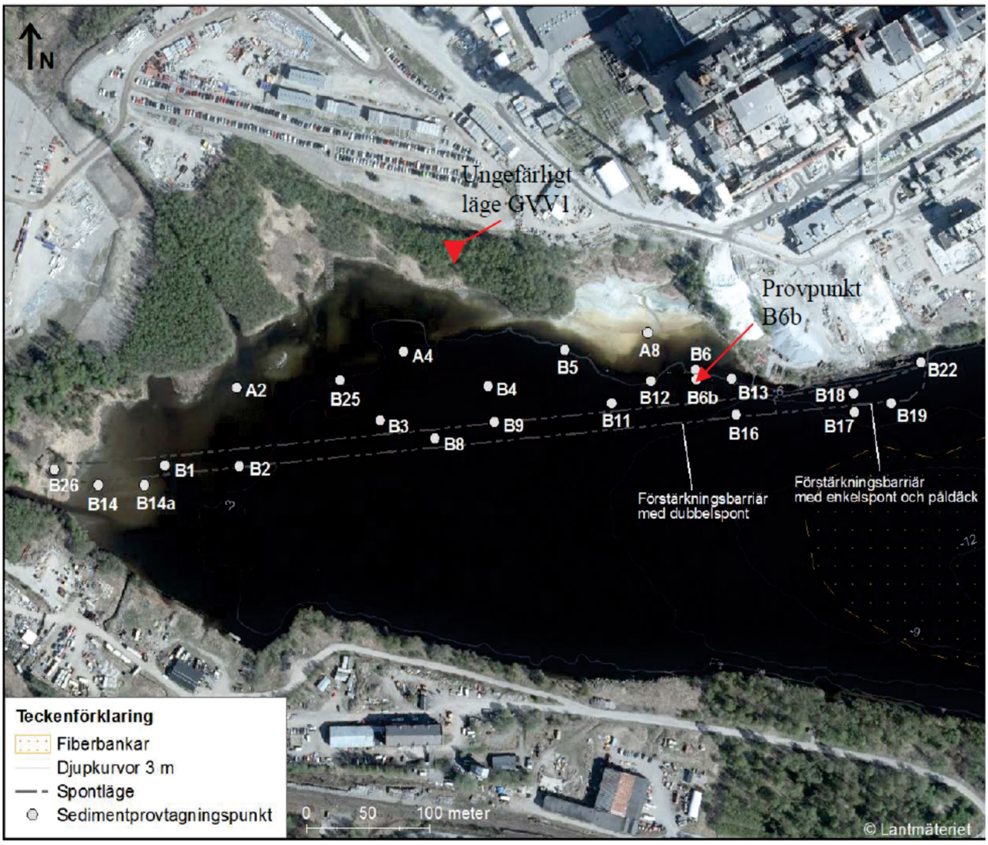

Fig. 1. Locations of sample sites in the Östrand region, Sweden, where samples were dredged: positions A4 and B4. The previous test point B6b used in the laboratory study is also marked

Rys. 1. Lokalizacje stanowisk pobierania próbek w regionie Östrand w Szwecji, gdzie pobrano próbki: pozycje A4 i B4. Poprzedni punkt testowy B6b użyty w badaniu laboratoryjnym jest również zaznaczony

2. Materials and methods

2.1. Sediment sampling in the Östrand region

The test experiments aimed at evaluating the geotechnical performance and environmental parameters of soil. The methodology is adapted to adjust binder ratios to achieve both the geotechnical and environmental technical quality requirements of treated soil. The soil samples, polluted by heavy metals and POPs: PAH, PCB and HCB, were dredged from the coastal area of Västernorrland, Sweden. The study adjusted the methodology of leaching with the aim of minimizing the number of trials while maintaining the quality of the analysis. We developed and applied a robust explanatory model that provides support in recommending final binder recipes where the leaching of heavy metals showed the best results. Laboratory geotechnical and environmental tests were performed in two different phases. These included tests on density and water ratio, particle size distribution, permeability, modified surface leaching and shake tests.

The quality requirements achieved in the field included permeability $<10^{-8}$ m/s, which enables the construction to be considered as a monolith with low water permeability. This requirement reduces the risk of the leaching of contaminants and shortens the service period caused by weathering or erosion. The experiments were performed with three different ratios of cement and slag as binding agents: 30/70, 50/50 and 70/30%. Three levels of binder quantity in kg/m^3 of dredged material were tested: 150 kg and 120 kg in phase 1 and 100 in phase 2. In addition, two levels of the water ratio were tested: 139 and 190%, that is, the excess of water. The test began with the uptake of the dredged material by digging with an excavator from a raft. Sampling took place in the test points A4 and B4 (Figure 1), which were selected in agreement with the County Administrative Board of Västernorrland. The sub-materials from sampling were sent to the SGI for the determination of grain distribution, density, water ratio and the chemical analysis of toxic elements and heavy metals. The density and water ratio were needed to determine the amount of binder to be used for S/S treatment in the field.

2.2. Workflow and general project description

The overall project description according to the test program was developed in accordance with the County Administrative Board of Västernorrland.

1. The bottom sediments were taken up using the bucket (volume approx. 1 per m^3). Samples were taken in the two test points, approx. one scoop per point. Samples from each point were placed into separate containers. The sediments were stored below the water level.
2. The representative samples were taken from each container and sent to the SGI for determination of the water ratio, density and grain-size distribution. The presence of the contaminants in sediments from each container was also determined.
3. The amount of binder was defined based on the density (selected recipe at 120 kg/m^3, and a comparative recipe at 150 kg/m^3) using the existing reports (Lindh 2001).
4. The soil masses in each container were homogenized. For this, a certain amount of sediments was taken out and placed in a forced mixer. Then, the correct amount of binder was added to the soil and mixed. Water corresponding to the weight of the binder was added to simulate the wetting method. Mixing was performed for around five minutes to achieve a homogeneous mixture. After mixing, the stabilized sediments were filled into plastic drums.

5. Temperature meters were installed in the plastic drums.
6. For each recipe, the specimens were made by filling fifteen piston sampling sleeves with the stabilized sediments and putting on lids.
7. The specimens were transported to the SGI where they were handled for environmental tests, such as storage in a heating cabinet, heat curing, evaluating strength and permeability as well as the chemical leaching of heavy metals through the reduced surface leaching tests (nine days) and shake tests.
8. Water samples were taken from the excess water (so-called clear phase) for chemical determination.
9. The evaluation of the results and a comparison of the contamination levels was made in the laboratory. Leaching tests from the scaled-up experiments were compared with those from the laboratory and against the area's groundwater and surface-water levels.

2.3. Temperature measurements T°C

Temperature measurements (Figure 2) were performed to estimate the maximum temperature during curing. After mixing in the binder, plastic barrels with a capacity amounting to 120 liters were filled with a stabilizer. Centrally positioned in the barrel was a tube with thermocouples attached on three levels. The temperature measurements were performed with a Testo 176 T4 using the 'T' type thermocouple technology. This type of logger has four channels, three of which were used to measure the stability and one channel was used to measure the ambient temperature. The measuring interval was set to 10 min.

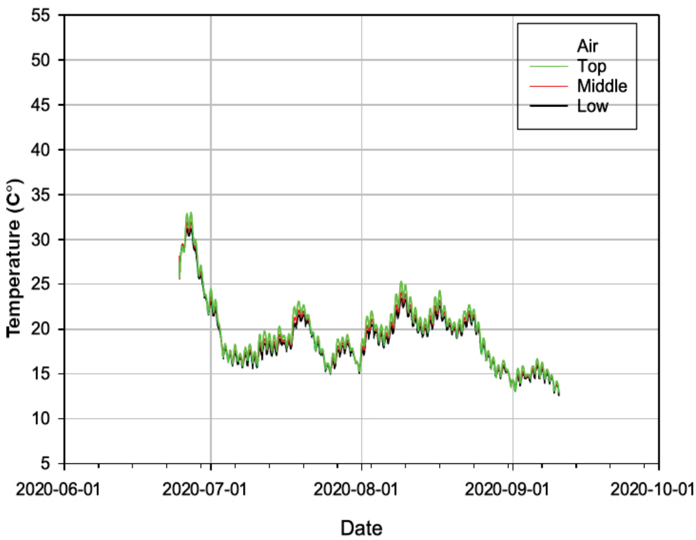

Fig. 2. Temperature of samples stabilized by 150 kg of binder per m^3 of the dredged soil collected from site B4

Rys. 2. Temperatura próbek stabilizowanych 150 kg lepiszcza na m^3 urobku pobranego z terenu B4

Large fluctuations in the air T°C were caused by the thermometer location – it was placed under the tarpaulin. Furthermore, the diurnal variations also have an effect on the stability. The highest T°C was ca. 33°C in the stable condition. The high air temperatures were due to the barrel being placed under a tarpaulin for protection against weather and wind. The graph shows that the stability is affected by the diurnal variations in the air temperature where the top sensor has the highest temperature. When a stabilized sample was covered by a water mirror, variations in the temperature decreased. The choice of mixture with materials from site B4 and 150 kg of binder is selected because this ratio had the lowest VBT and the highest heat development.

3. Results

The results of geotechnical and environmental tests showed positive effects of adding a large proportion of slag to the mixture. The ratio between the stabilizing agents (cement/slag) were more important for leaching than the amount of binder, as demonstrated in surface-leaching experiments. The results of the tests performed in the first phase showed that the most suitable mixture in the binder is 30% cement and 70% slag. The most optimal amount of binder is 120 kg per m^3 of dredged material, as defined in the course of Phase 2. This is robust enough to affect a change in the water ratio of soil material. However, the experiments also showed that the quality requirement for permeability is a better indicator of low leaching than leaching results themselves. The tested dredged soil samples were well suited to obtain a homogeneous material for stabilization regardless of the content of mesa and fiber.

3.1. Determination of density and water ratio

The determination of density and water ratio was performed on selected samples as a comparison between the natural water ratio and with an increased water ratio on the same material. The increase in water ratio was made using tap water. The results of the determination of density and water ratio are shown in Table 1.

Table 1. Density and water ratio of the untreated dredged materials

Tabela 1. Gęstość i wskaźnik wody w nieoczyszczonych pobranych próbkach

Test designation	Bulk density (tons/m^3)	Water ratio (%)
B6b_Lw (w_N)	1,378	139
B6b_Hw	1,282	190

The water ratio of the soil sample was determined by ten water ratio tests. The results of the tests are presented in Figure 3.

In order to evaluate the effects of a higher water ratio, tap water was added to the soil samples collected in B6b. The masses were then homogenized again with a new water ratio.

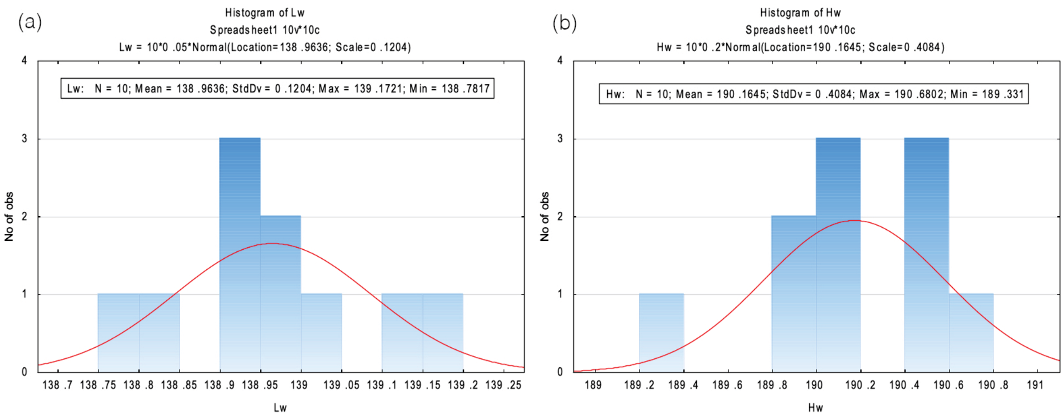

Fig. 3. Histogram of the water ratio in dredged soil samples collected on site B6b.
Lower water ratio in soil (a). Higher water ratio in soil (b)

Rys. 3. Histogram uwodnienia w próbkach gruntu pobranych na stanowisku B6b.
Niższy wskaźnik wody w glebie (a). Wyższy wskaźnik wody w glebie (b)

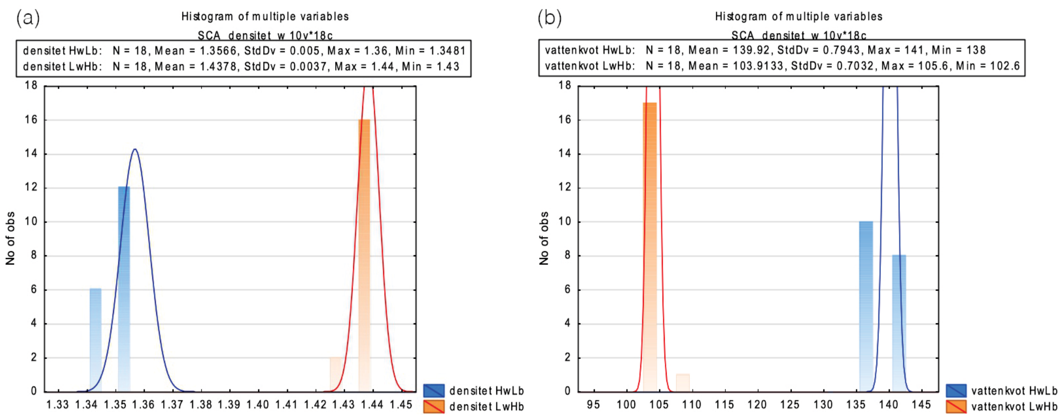

Fig. 4. Histogram of the density of stabilized specimens with a high water ratio and low binder (H_WL_B)
and a low water ratio and high binder content (L_WH_B) (left).
Histogram of the water ratio of the stabilized material for the two combinations (H_WL_B and L_WH_B).
The expected value is approx. 140% for H_WL_B and approx. 104% for L_WH_B (right)

Rys. 4. Histogram gęstości stabilizowanych próbek o wysokim wskaźniku uwodnienia i niskim spoiwie (H_WL_B)
oraz niskim wskaźniku uwodnienia i wysokiej zawartości spoiwa (L_WH_B) (po lewej).
Histogram wskaźnika wody stabilizowanego materiału dla dwóch kombinacji (H_WL_B i L_WH_B).
Oczekiwana wartość to około 140% dla H_WL_B i około 104% dla L_WH_B (po prawej)

The water ratio determinations from the new tests with increased water ratio are shown in Figure 3 (b). Here, the water ratio increased from around 139% to around 190% (exceed water). The standard deviation slightly increased for the new mixture with a higher water ratio. The density of the untreated dredged samples was evaluated to determine the optimal binder content that should be added into the samples (i.e. high and low levels). After stabilization, the density of the stabilized samples was measured again. The results of the density variations are shown in Figure 4. The expected value is 1.36 ton/m^3 for $H_W L_B$ and 1.44 ton/m^3 for $L_W H_B$. After stabilization, the water ratio of the stabilized soil was also determined.

3.2. Contamination of sediment samples

The content of contaminants (heavy metals, PAH, PCB, PHB, aliphatic compounds) in soil used in a scale-up experiment was evaluated and is summarized in Table 2.

The colors in Table 2 show samples containing the lowest and highest contaminant content in the series. Overall, the samples have an equivalent pollution content, although there are some differences. The sediments collected from the A4 test sites are considered to be more polluted than those from the B4 site. The highest mercury (Hg) content was detected in the group of sediments collected from the A4 site.

3.3. Contamination content in the excess water from admixture

In connection with the admixture of binders in the dredged masses, calcium (Ca) acts as a flocculant and can then lead to the formation of excess water on top of the stability. In the process of filling plastic barrels, water was sampled to check the possibilities of processing this water during the implementation of the planned agricultural construction. Sufficient excess water was formed in three of the four mixtures. No water sample could be taken from the mixture from sample point B4 with binder mixture 150 kg/m^3. Table 3 shows the pollution content in the excess water, which is compared with the reference value for industrial stormwater according to the Storm Tac.

The pollution content of the analyzed excess water is higher than the pollution levels stated as reference values for treated building stormwater, according to Condition 9 in a partial judgment from the Land and Environment Court at Östersund District Court on 30 September 2020, case M 757-19 for Hg and in some samples for Cr and Ni.

3.4. Surface leaching

The trial series consisted of five surface-leaching tests on concentrations of heavy metals, DOC and POPs measured on the ninth day in order to analyze and compare the worst

Table 2. Contamination content in a scale-up experiment on sediments collected from test sites A4 and B6 which were homogenized in the SGI laboratory (mg/kg TS). The results are compared with the sediment samples from site B6 which were homogenized in the SGI laboratory during the pilot study. Blue refers to the lowest content of the pollutants detected in sample series, while orange refers to the highest content, respectively

Tabela 2. Zawartość zanieczyszczeń w eksperymentach na większą skalę na osadach pobranych z poligonów badawczych A4 i B6, homogenizowanych w laboratorium SGI (mg/kg TS). Wyniki są porównywane z próbkami osadów ze stanowiska B6, które zostały zhomogenizowane w laboratorium SGI podczas badania pilotażowego. Podświetlenie na niebiesko oznacza najniższą zawartość zanieczyszczeń wykrytych w serii próbek, natomiast kolor pomarańczowy oznacza odpowiednio najwyższą zawartość

Element	Sediments from site A4	Sediments from site B4	B6 Pilot study
As	3.99	5.23	1.48
Cd	0.322	0.138	1.33
Co	3.45	9.43	2.61
Cr	56.1	41.6	26.5
Cu	26.5	20.4	28.9
Ni	11.2	21.6	25.1
Pb	15.2	12.4	26.8
V	14.1	37.1	12.4
Zn	68.3	75.4	243
Hg	5.41	1.7	5.07
Hg-methyl (ng/g TS)	6.79	1.98	1.03
PAH* L	<0.15	<0.15	<0.15
PAH M	0.22	<0.25	0.25
PAH H	<0.23	<0.23	0.073
PCB7**	0.013	0.0046	0.03
HCB***	0.0591	0.0102	<0.0050
aliphatic >C12–C16	102	<20	42
aliphatic >C5–C16	130	<24	64
aliphatic >C16–C35	201	48	366

and best cases. The leachate was analyzed on the DOC and heavy metals as a diffusion-controlled leaching of heavier POPs (PAHs, heavy aliphatic compounds). The analysis of these substances, which were then included in the shake tests, required some modification of the laboratory workflow. The pH values in surface leaching ranged from 11.4 to 12. The same pH range was measured in shake tests with both chemically pure water and groundwa-

Table 3. The contamination content in the excess water after the addition of a binder into the plastic barrels in the upscaling tests, minimum and maximum content (total of 3 samples)

Tabela 3. Zawartość zanieczyszczeń w nadmiarze wody po dodaniu spoiwa do beczek plastikowych w próbach upscalingu (zwiększania skali), zawartość minimalna i maksymalna (łącznie 3 próbki)

Element	Lowest content (µg/l)	Highest content (µg/l)	Reference value industrial stormwater Storm Tac (µg/l)
As	2.5	4.7	
Ba	112	361	
Pb	<0.2	0.3	30
Cd	0.1	0.3	1.5
Co	1.2	1.8	
Cu	6.4	11.5	45
Cr	1.0	45.5	14
Hg	4.5	11.5	0.07
Mn	0.4	0.5	
Mo	112	288	
Ni	13	24	16
S (mg/l)	249	360	
V	1.9	3.1	
Zn	<2	<2	270
hydrocarbon index, C10–C40	<100	<125	
hydrocarbon index, C10–C12	<10	<12.5	
hydrocarbon index, C12–C16	<10	<12.5	
hydrocarbon index, C16–C35	<60	<75	
hydrocarbon index, C35–C40	<20	<25	

ter collected from the study area. The pH level in the tests is assumed to be similar to that which can be reached if the groundwater is equilibrated with the stabilized soil in the field.

Figure 5 shows the results of leaching for substances that demonstrated a notable decrease with the increased slag content in the mixture, thus showing that the slag content affects leaching in an inverse way. Zinc is included when the three samples gave amounts that were below the detected limit of the analysis, shown as even bars. The decrease of the amount of DOC is interpreted as favorable, as DOC can be a carrier of pollutants of the functional groups on DOC (Pb). Similar trend for Pb, Ni and DOC can be an indicator

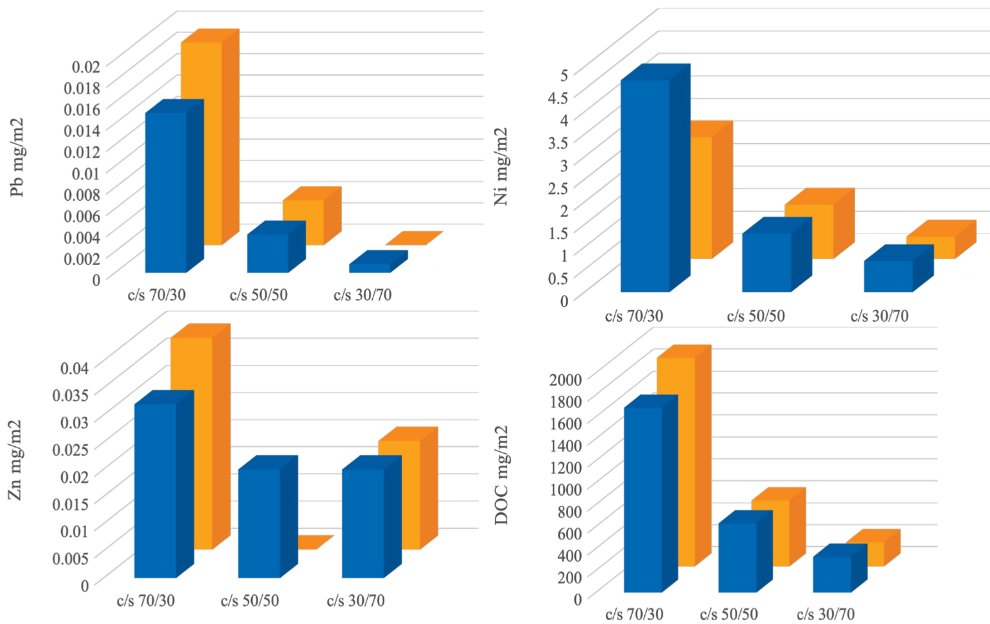

Fig. 5. Summary of the leached heavy metals on the ninth day per unit area (mg/m^2)
for substance elements that show a systemically decreasing trend with the increased slag content.
Orange shows the "best case" scenario, blue – the "worst case" scenario

Rys. 5. Zestawienie wyługowanych metali ciężkich w 9. dniu na jednostkę powierzchni (mg/m^2)
dla pierwiastków substancji wykazujących systematyczną tendencję spadkową wraz ze wzrostem zawartości żużla.
Kolor pomarańczowy oznacza „najlepszy przypadek", kolor niebieski – „najgorszy przypadek"

that Pb/Ni is released along with DOC. It can also be an indicator that all these substances are more sensitive to the differences in the permeability of material, since soil with the lowest permeability (stabilized by binder 30% cement/70% slag) also gives the lowest surface leaching. There is no marked difference in the leached amounts of heavy metals when the experimental test with the worst case are compared to the tests with the best case of binder ratios, Figure 5. This means that the ratio between the binders is a more decisive factor for leaching than the water/binder ratio.

Figure 6 shows leaching for the substances that demonstrated a bowl-shaped or unclear trend along with the increased slag content in the recipes. Thus, it could result that a higher slag content would be beneficial in reducing the risk of leaching. However, a recipe with a higher slag content could give a slightly higher leaching of Cu compared to the recipe with a higher proportion of cement.

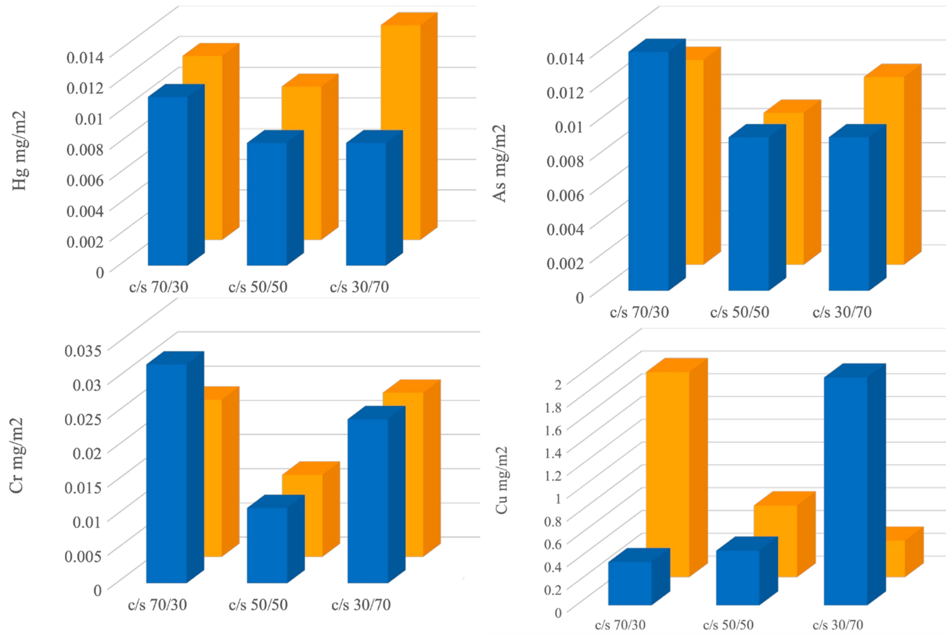

Fig. 6. Summary of the leached amount on the ninth day per unit area (mg/m^2) for substances that show a bowl-shaped or non-existent trend with the increased slag content.
Orange shows the "best case" scenario, blue – the "worst case" scenario

Rys. 6. Zestawienie ilości wypłukanej w 9. dniu na jednostkę powierzchni (mg/m^2) dla substancji wykazujących kształt czaszy lub braku tendencji ze zwiększoną zawartością żużla. Kolor pomarańczowy oznacza „najlepszy przypadek", kolor niebieski – „najgorszy przypadek"

3.5. Evaluation of a complete surface leaching test

The results are from the complete surface leaching experiment, which was carried out in full accordance with the standard. The experiment was performed on a sample with 50/50% cement/slag from the worst case series. The leaching in the reduced surface leaching experiments was analyzed on the ninth day. The reduced surface leaching experiments gave slightly higher mass flows. However, the differences were moderate and probably indicate a natural data variation. Table 4 shows the amount of leached heavy metals in the water samples.

Table 4. Leached amount at water sample on the ninth day (mg/m^2) in the upscaling experiments (total 12 samples). The comparison was made between the two corresponding recipes in a study (cement/slag at a ratio of 30/70% and high/low binder amount with low/high water ratio

Tabela 4. Ilość wypłukiwania w próbce wody w 9. dniu (mg/m^2) w eksperymentach zwiększania skali (łącznie 12 próbek). W badaniu dokonano porównania z dwoma odpowiednimi recepturami (cement/żużel w stosunku 30/70% oraz wysoka/niska ilość spoiwa z niskim/wysokim stosunkiem (wskaźnikiem) wody

Element (heavy metal)	Minimum amount per sample upscaling (mg/m^2)	Maximum amount per sample upscaling (mg/m^2)	Minimum amount per outlet (mg/m^2) pilot study	Maximum amount per outlet (mg/m^2) pilot study
As	0.01	0.04	0.009	0.011
Ba	2	8	2.6	3.8
Pb	<0.007	0.004	<0.001	0.001
Cd	<0.0003	<0.0003	<0.0003	<0.0003
Co	0.004	0.008	0.01	0.01
Cu	0.06	0.2	0.3	2
Cr	0.01	0.04	0.02	0.02
Hg	<0.007	0.04	<0.008	0.01
Mo	0.05	0.1	0.1	0.2
Ni	0.08	0.1	0.5	0.7
Zn	<0.01	0.3	<0.02	<0.02
DOC	<100	470	220	326

3.6. Shake tests for evaluating the equilibrium concentrations of metals L/S 10

3.6.1. Heavy metals: phase 1 and 2

The results of the equilibrium concentrations of the shake tests for metals are reported in Table 5. A comparison is made against the concentrations in groundwater that was used as leachate in one of the tests. The groundwater levels also represent the groundwater chemistry in the area and can be regarded as a reference for evaluating the equilibrium concentrations in the leaching experiments. The results indicate that the processed materials contribute to the equilibrium concentrations with elevated levels of DOC, Ba, Co and Ni compared to the area's natural groundwater chemistry. Ba and Ni also had the highest mass flows from the surfaces in the surface leaching tests, regardless of the ratio of binders. For the evaluation of the long-term environmental impact on the construction, it is mainly Ba and Ni that can be used as indicators of chemical impact. The results showed that leaching of Ba and Ni was

Table 5. Equilibrium concentrations during the shake tests at L/S 10 (μg/l)

Tabela 5. Stężenia równowagowe podczas testów wstrząsania przy L/S 10 (μg/l)

		50/50 C/S H_WL_B	50/50 C/S H_WL_B	30/70 C/S L_WH_B	30/70 C/S H_WL_B	30/70 C/S L_WL_B
	Groundwater	L/S 10 with MilliQ	L/S 10 with groundwater	L/S 10 with MilliQ	L/S 10 with MilliQ	L/S 10 with MilliQ
DOC (mg/l)	7.79	56	48	24	100	120
As	18.6	0.65	0.86	0.5	0.51	0.59
Ba	4.1	630	370	360	350	320
Cd	<0.50	0.004	0.008	<0.004	0.004	<0.004
Co	<0.50	5.9	6.8	4.8	1.7	2.1
Cr	<5.0	0.96	1.4	1.1	0.2	0.12
Cu	<1.0	<0.05	<0.068	64	3.4	3.5
Hg	<0.010	<0.1	<0.1	0.15	<0.1	0.13
Mo	12	15	24	19	19	22
Ni	3.1	280	290	210	140	190
Pb	<1.0	0.44	0.47	0.24	0.12	0.1
V	<5.0	15	28	29	30	33
Zn	<2.0	1.5	1.5	0.74	0.42	0.5

reduced when the binder contained a larger proportion of slag (Figure 5). This proves the fact that the optimal binder mixture should contain a larger ratio of slag.

3.6.2. Organic substances: phase 1 and 2

The levels of the POPs that have been detected in the shake test are summarized in Table 6. The leaching of organic substances occurred to a lesser extent from mixtures with a higher slag content, which results in the overall recommendation to add a higher proportion of slag when stabilizing soil. Since the shake test shows leaching mechanisms other than those applicable to the low-permeability materials (<10^{-8} m/s), the results are not representative enough for the final conclusions. However, the binder mixture has a beneficial effect on the leaching of POPs that are presented in the contaminated sediments.

Furthermore, the content of the oil-related pollutants in the aggregate samples B6b was much higher than that in the study area where the samples were dredged (see Table 6–7). This especially concerns heavier hydrocarbon fractions with a large molecular size, as the shake test demonstrated lower levels for heavy fractions compared to the light fractions

Table 6. Detectable equilibrium concentrations in shake tests at L/S 10 (µg/l)

Tabela 6. Wykrywalne stężenia równowagowe w testach wstrząsania przy L/S 10 (µg/l)

	50/50 C/S H_WL_B	50/50 C/S H_WL_B	30/70 C/S L_WH_B	30/70 C/S H_WL_B	30/70 C/S L_WL_B
	L/S 10 with MilliQ	L/S 10 with groundwaters	L/S 10 with MilliQ	L/S 10 with MilliQ	L/S 10 with MilliQ
PAH L	0.21	0.23	0.13	0.18	0.18
PAH M	0.07	0.076	0.049	0.037	0.082
oil index, sum	12300	1960	343	552	405
fraction C10–C12	406	250	269	241	312
fraction C12–C16	340	51	10.9	28	12.8
fraction C16–C35	8540	1300	58.4	255	72.3
fraction C35–C40	3030	360	<10.0	28.7	<10.0

Table 7. Leached content (µg/l) at L/S 10 in the upscaling experiments (4 samples in total). A comparison is made against the three corresponding recipes in the feasibility study (cement/slag at a ratio of 30/70% and high/low amount of binder with low/high water ratio, respectively)

Tabela 7. Zawartość wymywania (µg/l) przy L/S 10 w eksperymentach zwiększania skali (w sumie 4 próbki). Porównanie jest dokonywane z trzema odpowiednimi recepturami w studium wykonalności (odpowiednio cement/żużel w stosunku 30/70% i duża/niska ilość spoiwa przy niskim/wysokim stosunku wody)

Element	Lowest leaching, upscaling at 10 (µg/l)	Highest leaching, upscaling at 10 (µg/l)	Lowest leaching pilot study at L/S 10 (µg/l)	Highest leaching pilot study at L/S 10 (µg/l)
PAH L	0.6	2	0.13	0.18
PAH M	0.03	0.07	0.037	0.082
PCB 7 (amount)	0.003	<0.005	–	–
oil index, C10–C40	640	3380	343	552
oil index, C10–C12	<5	13	241	312
oil index, C12–C16	5.8	98	10.9	28
oil index, C16–C35	589	3030	58.4	255
oil index, C35–C40	43	237	<10.0	28.7

(compare PAH M with PAH L and the fraction C35–C40 with other fractions). The oil contamination in the soil mainly contains heavy aliphatic compounds of hydrocarbon fractions (Table 6). Shake tests on the equilibrium concentration regarding the oil index with 70% slag are also in line with the proposed guideline value of 400 µg/l for stormwater discharges to smaller lakes. Since shake test shows the equilibrium concentration that is not expected to be achieved in the field other than in the soil's inner pore water, treatment gives low emissions of POP substances as long as the permeability is lower than 10^{-8} m/s.

The results from the scaled-up experiments show good agreement with the results obtained by the feasibility study, except for the oil index, C16–C35. The results from the scaled-up experiments show higher levels there than can be explained. Based on the results of this study, a blend with binders of 30% Portland cement CEM II/A-V and 70% of slag type GGBFS demonstrated the best result in leaching.

4. Discussion

The excess water that formed in plastic barrels when binder was mixed into the contaminated sediments has been analyzed. The pollution content of the analyzed excess water is higher than the pollution levels stated as reference values for treated building stormwater according to Condition 9 in a partial judgment from the Land and Environment Court at Östersund District Court on 30.09.2020, case M 757-19 for Hg and in some samples for Cr and Ni. The temperature measurements show that the highest temperature is <35°T.

However, the analysis results of the surplus water are not considered to mean that any additional protection measures will be needed compared with what was previously described as a plan for controlling and managing water that arises during the construction phase of the project. It is estimated that there will be relatively limited amounts of the contaminated sediments that need to be stabilized/solidified. Therefore, the amount of the excess water that could be formed from this treatment is relatively limited. The extent of stabilization//solidification of the contaminated sediments is determined after the extended sampling of the sediments. A large part of the general excess water within the reinforcement line is estimated to be bound in the landfill.

In order to maintain the geotechnical and environmental technical quality during field construction, the amount of binder should amount to 120 kg per m^3 of the dredged material. From a geotechnical and environmental technical perspective, a binder mixture of 30% cement and 70% slag demonstrated the best result in terms of strength, permeability and leaching. A binder mixture with 30% cement of type CEM II / A-V or an equivalent product and 70% of GGBFS according to the SS EN 15167-1 is recommended for improving the soil properties for the best geotechnical and environmental performance.

An alternative mixture of soil with less slag (50%) and more cement (50%) can give a satisfactory effect in terms of strength and permeability (phase 1). When carrying out a large-scale project, scaling uncertainties in the form of uneven mixing technology should be taken into account for permeability requirements. Uneven mixing can result in a stabilized material showing varying environmental and geotechnical properties in the construction. When following up any long-term environmental impact from the finished construction, Ba and Ni serve as good indicators of environmental impact because they are released to the highest extent compared with the area's groundwater chemistry.

Conclusion

This paper has described a geochemical testing approach to evaluate the level of soil contamination caused by heavy metals and POPs (PAH, PCB and HCB) in the coastal area of the northern Baltic Sea in the Västernorrland area. The present work can be perceived as a continuation to soil testing aimed at environmental, geotechnical and geochemical assessment. From a geochemical point of view, the present approach makes more complete trials of binder ration allowing the analysis of their impact on the leaching process.

Several important contributions from this study can be explicitly recounted. They have shown the assessed leaching of heavy metals and POPs (PAH, PCB and HCB) from soil stabilized by cement/slag binders in various ratios.

Firstly, since adding binders (Portland cement and GGBFS) inherently involves variations in soil leaching due to changes in the structure of the stabilized specimens, the resulting disparity in leaching naturally estimates exhibit coherence between the binder ratio and its effects on the removal of heavy metals from soil samples. Thus, the comparison was made against the two mixes (cement/slag in 30/70% and high/low binder with low/high water ratio). The results showed that 70% slag decreases the leaching of heavy metals and POPs.

Secondly, the evaluation of min/max content of pollutants (µg/l) for heavy metals (As, Ba, Pb, Cd, Co, Cu, Cr, Hg, Mn, Mo, Ni, S, V, Zn) as well as the hydrocarbon fraction index in excess water that are ambiguous when considering only the chemical content are resolved through the inclusion of information on the binder ratio in soil samples.

Thirdly, a novel approach to the estimation of different blends of binding agents (30/70, 50/50, 70/30) and binder amounts (120 and 150 kg/m^3) is developed based on testing stabilized soil samples of the tested specimens using modified surface leaching and shake tests. The soil tests were performed using technical standards of SGI aimed at analyzing the effects of binder combinations on the leaching of toxic chemicals and heavy metals.

Next, a method for the direct testing of soil properties adapted from SIS standards and modified according to real-time soil conditions is applied in the workflow by the existing tools. In support of the existing similar studies using technical standard approaches of SIS, the results from this study have been documented qualitatively and quantitatively as a series of graphs using soil samples dredged from the study area.

Lastly, a method for measuring the equilibrium concentrations of DOC and heavy metals at L/S 10 (µg/l) has been designed and implemented using the SIS standards of shake experiments to compare the level of DOC in groundwater used as a leachate. The results of leached content were measured and estimated at L/S 10 in the upscaling experiments using 4 samples for PAH, PCB and various fractions of hydrocarbons: C10–C40, C10–C12, C12–C16 and C35–C40. The shake tests have shown the decrease in the leaching of heavy metals and POP substances from soil samples stabilized by a higher amount of slag added as a binder. A binder blend with 30% cement and 70% of GGBFS showed the best performance.

Importantly, the empirical advantage of testing the amount and ratio of stabilizing agents in soil samples has been shown to have practical value both in the construction industry as well as in environmental assessment. The latter especially concerns the comparative analysis of the lowest and highest contamination content in the excess water (heavy metals, PAH, PCB, PHB, aliphatic compounds) after the addition of binder for soil stabilization. Similarly, the proposed use of binders in various ratios also has the potential to support effective soil stabilization in case of weak soil, e.g. silt or clay. Waste products, especially of metallurgical origin, which are safe under given conditions, may become a source of the environmental pollution, e.g. when the pH is changing. This highlights the actuality of the environmental assessment of soil pollution, as presented in this study. More generally, the proposed approach of changing stabilizing agents contributes to the practical development of geotechnical methods of improving soil properties and structure, as needed for the construction industry.

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SOIL CONTAMINATION FROM HEAVY METALS AND PERSISTENT ORGANIC POLLUTANTS (PAH, PCB AND HCB) IN THE COASTAL AREA OF VÄSTERNORRLAND, SWEDEN

Keywords

marine pollution, soil, civil engineering, geotechnical engineering, Baltic Sea

Abstract

This paper presents an experimental study on the leaching of heavy metals, toxic chemicals and persistent organic pollutants (POPs) – PAH, PCB and HCB – from soil dredged from the coastal area of Västernorrland in northern Sweden. The soil was stabilized with cement/slag. Samples were subjected to modified surface leaching and shake tests using technical standards of the Swedish Geotechnical Institute (SGI). The experiments were performed using different blends of binding agents (30/70, 50/50, 70/30) and binder quantities (120 and 150 kg/m^3) to analyze their effects on leaching. Soil properties, tools, and workflow are described. Binders included Portland cement and ground granulated blast furnace slag (GGBFS). Samples were tested to evaluate the min/max contents of pollutants (µg/l) for heavy metals (As, Ba, Pb, Cd, Co, Cu, Cr, Hg, Mn, Mo, Ni, S, V, Zn) and the hydrocarbon fraction

index in the excess water. The leaching of heavy metals and POPs was assessed in sediments after the addition of the binder. The comparison was made against the two mixes (cement/slag in 30/70% and high/low binder with low/high water ratio). The results showed that 70% slag decreases the leaching of heavy metals and POPs. The equilibrium concentrations of DOC and heavy metals at L/S 10 (µg/l) were measured during the shake experiments to compare their levels in the groundwater that was used as a leachate. The leached content was assessed at L/S 10 in the upscaling experiments using four samples for PAH, PCB and various fractions of hydrocarbons: C10–C40, C10–C12, C12–C16 and C35–C40. The shake test showed a decrease in the leaching of heavy metals and POP substances from the soil subjected to stabilization by a higher amount of slag added as a binder. A binder blend with 30% cement and 70% of GGBFS showed the best performance.

ZANIECZYSZCZENIE GLEBY METALAMI CIĘŻKIMI I TRWAŁYMI ZANIECZYSZCZENIAMI ORGANICZNYMI (WWA, PCB I HCB) NA OBSZARZE PRZYBRZEŻNYM VÄSTERNORRLAND, SZWECJA

Słowa kluczowe

gleba, Morze Bałtyckie, zanieczyszczenia morskie, inżynieria lądowa, geotechnika

Streszczenie

Niniejszy artykuł przedstawia eksperymentalne badania dotyczące wymywania metali ciężkich, toksycznych chemikaliów i trwałych zanieczyszczeń organicznych (TZO): WWA, PCB i HCB z pobranej gleby na obszarze przybrzeżnym Västernorrland w północnej Szwecji. Gleba była stabilizowana cementem/żużlem. Próbki poddano zmodyfikowanym próbom wypłukiwania powierzchniowego i wstrząsom z zastosowaniem standardów technicznych Szwedzkiego Instytutu Geotechnicznego (SGI). Eksperymenty przeprowadzono przy użyciu różnych mieszanek środków wiążących (30/70, 50/50, 70/30) i ilości środka wiążącego (120 i 150 kg/m^3) w celu przeanalizowania ich wpływu na ługowanie. Opisano właściwości gleby, narzędzia i przebieg pracy. Spoiwa obejmowały cement portlandzki i mielony granulowany żużel wielkopiecowy (GGBFS). Próbki zostały przetestowane w celu określenia min/max zawartości zanieczyszczeń (µg/l) dla metali ciężkich (As, Ba, Pb, Cd, Co, Cu, Cr, Hg, Mn, Mo, Ni, S, V, Zn) i wskaźnika frakcji węglowodorowej w nadmiarze wody. Wymywanie metali ciężkich i TZO oceniano w osadach po dodaniu lepiszcza. Porównania dokonano dla dwóch mieszanek (cement/żużel w 30/70% i spoiwo o wysokiej/niskiej zawartości z niskim/wysokim stosunkiem wody). Wyniki wykazały, że 70% żużel zmniejsza wymywanie metali ciężkich i TZO. Stężenia równowagowe DOC i metali ciężkich przy L/S 10 (µg/l) mierzono podczas eksperymentów z wytrząsaniem w celu porównania ich poziomów w wodzie gruntowej stosowanej jako odciek. Zawartość wyługowaną oszacowano na poziomie L/S 10 w eksperymencie upscalingu (zwiększenia skali) przy użyciu 4 próbek WWA, PCB i różnych frakcji węglowodorów: C10–C40, C10–C12, C12–C16 i C35–C40. Próba wstrząsowa wykazała zmniejszenie wymywania metali ciężkich i substancji TZO z gleby poddanej stabilizacji większą ilością żużla dodawanego jako spoiwo. Najlepszą wydajność wykazała mieszanka spoiwowa zawierająca 30% cementu i 70% GGBFS.