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Research article

Enhanced remediation of organotin compounds and metal(loid)s in polluted sediments: Chemical stabilization with mining-wastes and nZVI versus physical soil washing

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ABSTRACT

Here we describe two innovative approaches for remediating sediments contaminated with organotin compounds (OTCs, mainly TBT) and metal(loid)s. The first involves chemical stabilization through amendments with nanoscale zero-valent iron (nZVI), dunite mining waste, and coal tailings, materials that have not been previously studied for OTC remediation. The second focuses on physical soil washing, using grain-size separation and magnetic separation to isolate the most polluted fractions, thereby reducing the volume of contaminated material destined for landfills. The results for the first approach indicated that OTC degradation occurred mainly through nZVI application, with concurrent immobilization of As and mobilization of Cu. Furthermore, combining nZVI with coal tailings enhanced OTC degradation whereas dunite mining waste effectively immobilized Zn. In turn, in the second approach, grain-size separation efficiently removed coarse material (>500 μm) with low pollutant concentrations. Subsequent magnetic separation selectively concentrated less than 5% of the initial volume of sediment in a magnetic fraction that showed the highest contaminant content. Therefore, 95% of material revealed lower contaminant concentrations than the feed material. These findings highlight the potential of combining physical soil washing, which significantly reduces the volume of contaminated sediments, with chemical stabilization, which can effectively stabilize the polluted fractions isolated in the physical treatment.

1. Introduction

More than 800 organotin compounds (OTCs) are known. Most of them are of anthropogenic origin, except methyltins, which can be also generated through biological methylation (Miguez and Tarazona, 2024). OTCs can be present as mono-, di-, tri-, and tetra-substituted analogues, increasing toxicity from mono- to tri- and decreasing through the addition of the fourth butyl group (Gomez et al., 2024). Tributyltin (TBT) is one of the most toxic OTCs (Chen et al., 2024a,b), exerting effects such as gastropod imposex or oyster malformation at concentrations as low as 1 ng/L of TBT in water (European Commission, 2005). TBT can be degraded through sequential debutylation by biological and physico-chemical processes. However, Viglino et al. (2004) estimated

that TBT has a significant half-life of 87 ± 17 years in sediments in anoxic conditions.

In coastal areas, sediment fine fractions can potentially host dissolved TBT in water, as it tends to get adsorbed or bound, although this is a reversible process and therefore sediments can be also a sink for renewed contamination (Bajt et al., 2024). In this context, in September 2008, the International Maritime Organization (IMO) brought in a ban on the use of TBT (International Maritime Organisation/AFS Convention, 2001). This prohibition was adopted by many countries that ratified the Convention on the Control of Harmful Anti-fouling Systems on Ships (AFS Convention, 2001). However, this compound is still used in the construction of large vessels in some developing countries and is even used to paint smaller vessels (de Oliveira et al., 2020). Marine water and

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sediment pollution by OTCs may be aggravated in harbour areas with the interaction of multiple sources of contamination such as heavy industry, navigation, waste disposal, etc., causing the release and accumulation of a variety of organic and inorganic contaminants (Baragaño et al., 2022; Kennish, 2021). In this context, heavy metal(loid)s such as As, Cr, Cu, Ni, Pb, and Zn are commonly found in sediments (Wu et al., 2024). Given the toxicity and potential bioaccumulation of these metal(loid)s, immobilization strategies to reduce their capacity to migrate are required (Baran et al., 2024).

Considerable efforts have been devoted to the remediation of TBT-polluted water (Ayanda et al., 2013a, 2013b; Ottosen et al., 2006; Yamashita et al., 2012); however, less attention has been given to TBT-contaminated sediments. In this regard, some authors reported >99% of TBT removal by thermal treatments at 1000 °C (Song et al., 2005), or at 230 °C under a pressure of 34.5 atm (Mostofizadeh, 2003). Also, different results were achieved by steam-stripping (Du et al., 2014a), bioremediation (Polrot et al., 2024; Sakultantimetha et al., 2011), phytoremediation (Xiong et al., 2024), chemical oxidation (Rajendran et al., 2022), and electrochemical oxidation (Norén et al., 2022). However, physical separation approaches (soil washing) have not been attempted.

Physical soil washing technologies aim to concentrate the contaminants into a small volume of soil or sediment by exploiting the physical properties of the polluted particles through mineral processing techniques (Baragaño et al., 2023). For example, grain-size separation and magnetic separation are based on the distribution of particle sizes and magnetic properties (ferro-, dia- or paramagnetic behaviour), respectively (Dermont et al., 2008; Sierra et al., 2013). According to grain-size distribution, fine particles tend to concentrate most contaminants due to a greater surface area and surface charge (Gyabaah et al., 2023). In turn, magnetic separation of the contaminants can be achieved through binding to ferromagnetic materials such as metallic minerals, whereas geogenic non-metallic material present in sediments is typically bound to diamagnetic minerals such as quartz or others (Baragaño et al., 2023).

Unlike physical soil washing, chemical stabilization is a green remediation technology that seeks to reduce pollutant availability and it has not been tested in TBT-polluted sediments. This approach involves the application of amendments to immobilize contaminants through chemical mechanisms such as sorption, oxidation-reduction, etc. (Palansooriya et al., 2020). Given that the efficiency of this technique varies depending on the pollutants and physicochemical properties of the soil or sediments to be treated, it is usually necessary to explore a range of amendments. Among these, non-hazardous sub-products from mining activities have some favourable properties for the treatment of polluted soils (Díaz et al., 2023) or sediments (Deng et al., 2021) and their use is also beneficial because it complies with the principles of the circular economy (Baragaño et al., 2019; Breure et al., 2018). In this context, modified mineral products have been used for TBT sorption from wastewater (Du et al., 2014, 2017), although their effects on TBT-contaminated sediments were not reported. Another option within the amendment alternatives for chemical stabilization is the use of zero-valent iron nanoparticles (nZVI), which have shown particularly high immobilization rates for metal(loid)s such as As, Cd, Cr, Hg, Pb or Zn (Baragaño et al., 2020a; Vítková et al., 2018) in nanoremediation treatments. Of note, the application of nZVI to remove TBT from landfill leachates revealed adsorption capabilities at alkaline pH and enhanced TBT degradation at acidic pH (Peeters et al., 2015). In a different context of nanoremediation, nanocomposites made from ZnO nanoparticles supported on activated carbon achieved TBT adsorption efficiencies of up to 99.8% from seawater (Ayanda et al., 2013a).

Given the preceding considerations, we evaluated the capacity of chemical stabilization and physical soil washing to remediate sediments contaminated with OTCs (mainly TBT) and multiple metal(loid)s. This was achieved through an incubation experiment to assess the performance of different types of mining waste, used as amendments, either alone or in combination with nZVI, for the degradation or

immobilization of pollutants. Additionally, grain-size separation and magnetic separation were tested to determine the feasibility of physical soil washing in reducing the volume of polluted sediments. The novelty of this work lies in the use of mining waste amendments and nanomaterials that have not been previously studied for the treatment of OTCs in sediments, as well as the evaluation of physical separation techniques, which have also not been applied to TBT remediation. Furthermore, we examined these strategies independently, providing insights into their individual effectiveness as well as their synergistic potential for remediating sediments contaminated with OTCs and metal(loid)s. The advantages, limitations, and feasibility of integrating these two approaches are thoroughly discussed.

2. Materials and methodology

2.1. Samples

Polluted sediment was collected from the Port of Gothenburg, Göta älv river mouth, located on the west coast of Sweden. This port is the largest in Scandinavia and one of the most important in Europe. Dredging activities are required not only to allow current shipping activities but also to expand the port by building a new terminal. The decontamination of the sediments collected by dredging could allow their use, when mixed with other binders, as a construction material for the new terminal (Norén et al., 2024; Svensson et al., 2022).

2.2. Sediment and amendment characterization

A bulk sample of 5 kg of sediment polluted mainly with Zn and OTCs (labelled S) was used for the experiments. Before laboratory analysis and experiments, the sample was air-dried and sieved through a 2-mm mesh to confirm that all particles were below this grain-size. One organic and two inorganic amendments were tested.

The first inorganic amendment was dunite waste derived from mining activities (labelled D), provided by Pasek Minerals, which was taken from David Mine, in Landoi (northern Spain). A full mineralogical characterization of this material was described in previous work (Baragaño et al., 2019; Díaz et al., 2023), revealing a high Mg and Ca content and alkaline pH as favourable properties for chemical stabilization. The dunite used in this study was taken from the mineral processing plant and it comprised mainly fine particles from 0 to 3 mm in diameter.

The second inorganic amendment was nZVI. This material (labelled N) was the commercial product named NANO FER 25S, a nanoparticle water suspension commercialized by NANO IRON s.r.o. (Rajhrad, Czech Republic). According to the commercial specification, the Fe(0) content is 14–18% and iron oxides account for 2–6%. The particles have an average diameter of around 60 nm, the suspension is strongly alkaline (pH 11–12), and the active surface area is 20 m²/g (Baragaño et al., 2020b).

Finally, the organic amendment was coal tailings, a waste product from coal mining (labelled C). This fine-grained material taken from a coal processing plant, Lavadero Batán in Mieres (Asturias, Spain), was used in combination with the inorganic amendments mentioned above. To the best of our knowledge, this material has not been used as a soil or sediment amendment before, but the high content of C should be beneficial, as demonstrated with other organic amendments such as biochar (Wang et al., 2018) and coal foams (Janeiro-Tato et al., 2022).

The sediment and amendments were characterized before the experiments. The pH was determined using a pH electrode (Mettler Toledo S400) in a 1:2.5 water-to-sediment extract (Porta Casanellas et al., 1986). Total carbon (TC) and total nitrogen (TN) contents were analysed in a LECO CN-2000 module. Representative subsamples of 0.25 g were used to determine the total metal(loid) content. The samples were first digested by Aqua regia in a microwave oven (Milestone ETHOS 1, Italy), then passed through a 0.45 µm filter and diluted for the quantitative

determination of the elements by Inductively Coupled Plasma Mass Spectrometry (ICP-MS 7700, Agilent Technologies, California, USA) using IDA (Isotopic Dilution Analysis). The detection limit for all elements was 20 µg/L. Metal(oid) availability was determined using the TCLP test (Toxicity Characteristic Leaching Procedure) following USEPA Method 1311 (1992). The OTCs TBT, dibutyltin (DBT), and monobutyltin (MBT) were analysed at an external accredited laboratory using SS-EN ISO 23161:2011 (International Organization for Standardization, 2011) by gas chromatography-mass spectrometry (GC-MS).

2.3. Chemical stabilization experiment

The 30-day experiment was conducted in pots maintained at an average temperature of 13 ± 4 °C and 15/9 h (day/night cycle) with a total weight of 150 g. The amendments were applied to the sediment by mechanical mixing. The sediment (S) was combined with the coal tailings (C) and one of the two inorganic amendments, either dunite (D) or nZVI (N). Each inorganic amendment was also tested alone. A control test without any amendment was also included. Each treatment was tested in triplicate. The proportions of sediment and amendments are shown in Table 1.

The pots were watered three days per week due to evaporation, maintaining 60% of water-holding capacity (w/w) throughout the experiment. After 30 days, the sediments were removed from the pots and subjected to the following analyses (methods previously described in section 2.2): pH, total metal(oid) contents, metal(oid) concentrations after TCLP, and TBT, DBT, MBT contents.

2.4. Physical soil washing experiments

2.4.1. Grain-size and pollutant distribution

A representative 1 kg sample was wet-sieved in 100 g batches using a standardized series of RETSCH screens, following norm ASTM D-422-63. Different fractions (<63 µm, 63–125 µm, 125–250 µm, 250–500 µm, 500–1000 µm, and >1000 µm) were obtained and their metal(oid) and OTC contents were analysed by ICP-MS and GC-MS respectively, as in sediment and amendment characterization.

2.4.2. Magnetic separation approach

The separation was carried out with an OUTOTEC Laboratory Wet High Intensity Magnetic Separator (WHIMS) 3×4L apparatus. This device can separate paramagnetic (weakly magnetic) from non-magnetic materials and its operation under wet conditions makes it suitable for fine particles (<500 µm) (Baragaño et al., 2021). The feed was a mixture of 50 g of dry sediments (S) and 200 g of water. This mixture passed through a matrix canister (separating chamber) filled with magnetized steel spheres measuring 6.5 mm in diameter where the separation was performed. The magnetic fraction (MF) was retained by the spheres, while the non-magnetic fraction (NMF) passed through the canister with the water and was collected in a tray. After recovering the entire NMF, the magnetic field was turned off to recover the MF, both were air-dried and analysed. The maximum output voltage was adjusted to 25% for all tests through the variable magnetic field intensity of the equipment (Sierra et al., 2014). The tests were carried out on the fractions with higher OTC and metal(oid) concentrations (i.e. 500–250 and 250–125 µm, see results). After each test, all samples were air-dried and then

Table 1
Proportion (%) of sediment and amendments used in the chemical stabilization experiment.

Treatments	S	SC	SD	SN	SCD	SCN
Amendments						
Sediment	100	80	80	98	80	80
Coal tailings		20			10	18
Dunite waste			20		10	
nZVI				2		2

weighed to obtain the weight recovery (WR), and the percentage of material recovered in each fraction (MF and NMF). WR was calculated from the mass, in grams, obtained in each fraction using the following expression (1), in the case of the MF:

$$WR_{MF} = \frac{m_{MF}}{m_{MF} + m_{NMF}} \cdot 100 \quad (1)$$

The samples were then used to analyse the concentration of metal (oid)s and OTCs using ICP-MS and GC-MS respectively. From these data, element recovery (ER) was calculated for each pollutant studied by considering the amount of the element in each fraction with respect to the total amount of the same element in the feed material. The following formula (2) describes calculations for ER in the case of the MF:

$$ER_{MF} = \frac{WR_{MF} \cdot c_{MF}}{(WR_{MF} \cdot c_{MF}) + (WR_{NMF} \cdot c_{NMF})} \quad (2)$$

Finally, samples were also analysed by powder x-ray diffraction (PXRD) on a PANalytical X'Pert Pro MPD diffractometer with Cu Kα1 radiation (1.540598 Å). After determining the position of Bragg peaks over the range of $2\theta = 5-80^\circ$, the minerals were identified using databases of the International Centre for Diffraction Data, ICDD PDF-2 catalogue (ICDD, 2003).

2.5. Statistical analysis

All analytical determinations were performed in triplicate. The data obtained were treated using the SPSS program, version 27.0 for Windows. Analysis of variance (ANOVA) and test of homogeneity of variance were done. In the case of homogeneity, a post hoc least significant difference (LSD) test was conducted. If there was no homogeneity, Dunnett's T3 test was performed. A correlated bivariate analysis was also performed using Pearson correlation.

3. Results and discussion

3.1. Sediment and amendment characterization

The initial characterization of the sediment and the amendments used in the incubation experiment is summarized in Table 2. Sediment properties revealed a slightly acidic pH, and low TN and TC contents. To evaluate the content of potential pollutants, The Norwegian Guidelines for Risk Assessment of Contaminated Sediments (Direktoratsgruppen vanddirektivet, 2018) and the Canadian Sediment Quality Guidelines for the Protection of Aquatic Life (CCME, 2001) were considered as they are commonly used in Sweden due to the lack of a national guide in this country and the similarity of the geological environment (Bakke et al., 2007; INSURE, 2017; Norén et al., 2020). Based on the Norwegian Guidelines, for As, Cu, Pb and Zn, the sediment can be categorized as class I (background condition), although based on TBT concentrations it falls into class V (very poor quality; extensive toxic effect). However, following the Canadian Guidelines, which do not include threshold values for TBT, the sediment can have potential toxicological effects due to high concentrations of some metal(oid)s. In the case of Cu and Zn, the threshold for effect levels (TEL values) were exceeded (>18.7 mg/kg and >124 mg/kg, respectively).

The three amendments (dunite, coal tailings, and nZVI) used in the experiment showed a basic pH, although large differences in TN and TC contents were found between them (Table 2). Dunite is an inorganic mining waste that is poor in TN and TC, whereas the coal tailings revealed high concentrations of these elements. Furthermore, As and Cu concentrations in both amendments exceeded the TEL values from the Canadian Guidelines (>7.2 mg/kg and >18.7 mg/kg, respectively) (CCME, 2001).

Table 2
Initial characterization of the sediment (S), dunite waste (D), and coal tailings (C) used in the experiment.

Parameters	Units	Samples			
		S	D	C	
	pH	6.3	9.2	9.4	
	TN	mg·kg ⁻¹	1500	<DL	4316
	TC		23,400	2.2	318,716
Total concentration	As	mg·kg ⁻¹	6	16	17
	Cu		40	29	28
	Pb		28	18	26
	Zn		140	38	67
	As	mg·kg ⁻¹	0.18	<DL	<DL
Available concentration (TCLP)	Cu		0.70	<DL	0.39
	Pb		0.18	0.06	0.13
	Zn		20.52	<DL	5.13
	MBT	μg·kg ⁻¹	120	–	–
OTC	DBT		51	–	–
	TBT		58	–	–

< DL, below detection limit.

3.2. Chemical stabilization

At the end of the experiment, all the treatments showed significantly higher pH values compared to the control sediment (Table 3). This increase in pH was due to the alkaline pH of the different amendments applied, especially in the case of the two mining wastes (D and C application). The combination of organic and inorganic amendments led to a similar increase in pH, except when nZVI was applied (treatments SN and SCN). The application of nZVI alone did not affect pH as much as the coal tailings, although simultaneous application did increase the pH.

Regarding the total contents of the target elements (Table 3), the coal tailing amendment slightly increased the total content of As as expected, due to its As content. The presence of As in this type of material is related to its source (Ketris and Yudovich, 2009). Moreover, the application of nZVI increased the Cu concentration in the sediment considerably, even when applied in combination with the coal tailings (SN and SCN). The presence of Cu in nZVI was described in previous work (Baragaño et al., 2020c). The total content of Pb and Zn in the sediment was not modified after the application of the treatments, indicating that there was no significant contribution of these metals from the amendments but rather a dilution effect for some of them.

Fig. 1 shows the availability, as shown by TCLP, of all the metal(loid)s at the end of the experiment. The availability of As in the sediment increased significantly after the application of the dunite waste (SD); the same occurred, to a lesser extent, with the addition of the coal tailings (SC), and even when these two amendments (SCD) were combined. The observed increase in As availability after the application of dunite waste was unexpected as previous studies showed the opposite effect in polluted soils (Baragaño et al., 2019; Díaz et al., 2023). Our observation

could be attributed to the higher concentration of As in the dunite (16 mg/kg As) compared to the sediment (6 mg/kg As). In contrast to the previous studies reporting contradictory results, the As concentration in the polluted soil was higher than that of the dunite. Of note, the concentration of As (available and total) was very low in the sediments. In contrast to dunite waste, the use of nZVI successfully immobilized As, even in the presence of the coal tailings (SN vs. SCN). The As immobilization after nZVI application is due mainly to adsorption onto iron oxides originated in the solid phase through inner-sphere surface complexation (Baragaño et al., 2021). The application of coal tailings and dunite waste decreased Cu availability as a result of the increase in pH, which may cause precipitation of the metal. However, the interaction between Cu and organic matter cannot be overlooked when applying coal tailings to the sediment. In turn, the use of nZVI (SN) increased the availability of Cu in the sediment. Based on the total contents (Table 3), the increase in Cu availability observed in this treatment may be explained by the presence of this metal in the nanomaterial. Note that Cu is readily mobile and susceptible to leaching. Nevertheless, the addition of the coal tailings amendment (SCN) significantly reduced this effect, suggesting that the Cu from nZVI reacts with organic matter from coal thus being immobilized. This interaction between nZVI and organic amendments has been demonstrated through the modification of biochar via impregnation with this nanomaterial (Su et al., 2024), an emerging technology that has yielded excellent results, comparable to those obtained in this study. In this context, the modification of coal wastes through nZVI impregnation emerges as a promising application pathway.

All amendments decreased the availability of Pb, especially in the case of the nZVI treatment (SN and SCN). Finally, the treatments showed high differences in Zn availability (Fig. 1). The mobility of this metal decreased by 31%, 36% and 66% after the application of nZVI, coal tailings, and dunite waste, respectively. Therefore, the latter emerged as the most effective amendment for Zn immobilization among the three materials tested. Consequently, the combination of coal tailings with dunite waste (SCD) achieved a greater degree of Zn immobilization than the organic amendment alone. Surprisingly, the combination of coal tailings and nZVI (SCN) revealed better immobilization than applying each amendment alone, probably due to a double effect immobilization mechanism, i.e. precipitation caused by the increase in pH plus sorption interaction between Zn and iron oxides created after nZVI oxidation in the sediment.

These results for metal immobilization are highly promising, as they achieve high efficiency ratios at significantly lower doses compared to those reported in the literature. For instance, Baran et al. (2024) demonstrated that cellulose waste, biochar, and dolomite were tested to reduce the mobility of Cd, Cu, Pb, and Zn in marine sediments, with cellulose waste showing the best results: a 71% reduction in Zn mobility at a dose of 45%. In our case, a 66% reduction was achieved using only 20% dunite, highlighting this additive's competitive advantage over

Table 3
Total metal(loid)s contents and pH of the sediment at the end of the experiment. The threshold levels of the Canadian and Norwegian Guidelines (for the different classes) are also shown in the table.

		pH	As	Cu	Pb	Zn
			mg · kg ⁻¹			
Canadian Guidelines	TEL		7.2	18.7	30.2	124
Norwegian Guidelines	I		0–15	0–20	0–25	0–90
	II		15–18	20–84	25–150	90–139
	IV		71–580	84–147	1480–2000	750–6690
	Total concentration	S	4.8 ± 0.1d	6.1 ± 0.1d	32.5 ± 0.3cd	25.03 ± 0.2a
	SC	6.5 ± 0.1b	8.6 ± 0.2b	30.75 ± 0.6d	24.76 ± 0.5 ab	119.8 ± 1.2b
	SD	6.7 ± 0.1a	5.0 ± 0.2e	34.55 ± 0.1c	20.94 ± 0.2b	112.3 ± 1.7c
	SN	5.7 ± 0.1c	6.8 ± 0.1d	71.67 ± 0.7b	25.18 ± 0.4a	133.7 ± 1.8a
	SCD	6.5 ± 0.1b	7.4 ± 0.3c	31.98 ± 0.1c	22.69 ± 0.4 ab	116.4 ± 1.4bc
	SCN	6.6 ± 0.1 ab	9.2 ± 0.1a	77.88 ± 2.0a	25.70 ± 0.5a	127.5 ± 0.7a

For each row, different letters in different samples indicate significant differences (n = 3, ANOVA; p < 0.05). Typical deviation is represented by ±.

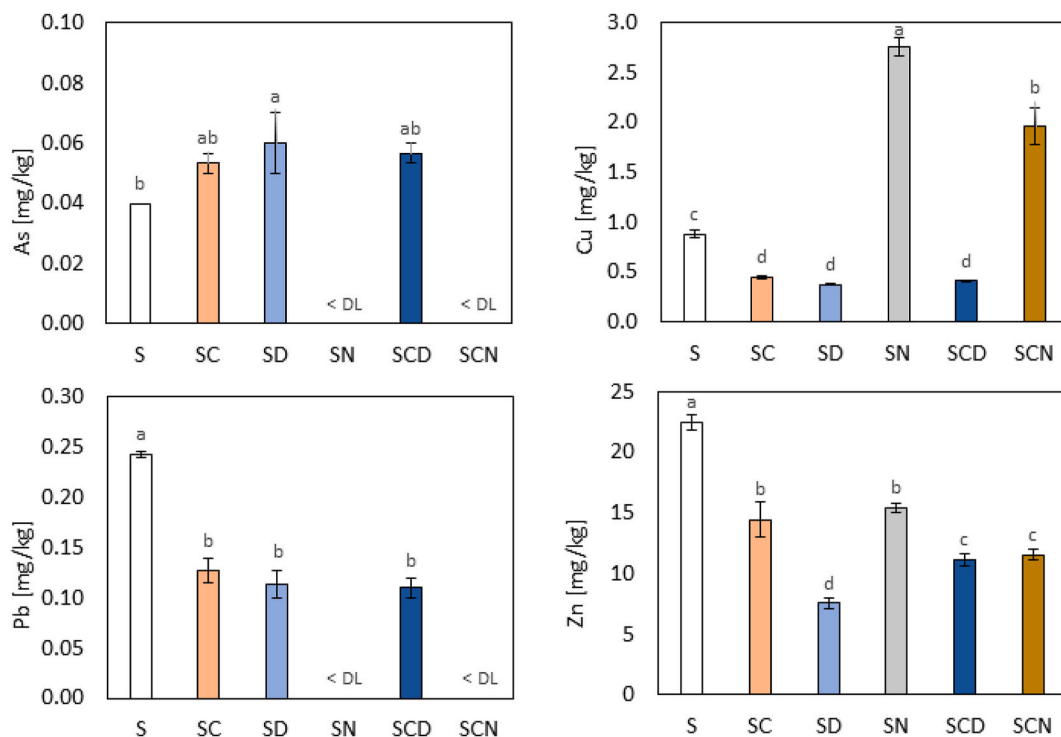


Fig. 1. Concentrations of available As, Cu, Pb and Zn (after 30 days using TCLP test) in the control sediment (S) and treatment with coal tailings (SC), dunite waste (SD), nZVI (SN), coal tailings-dunite waste (SCD), and coal tailings-nZVI (SCN), as analysed by TCLP extraction (<DL: under the detection limit).

other amendments. Furthermore, compared to dolomite—an amendment more widely used and better known than dunite—our results show a 66% reduction in Zn mobility at lower doses, whereas dolomite achieved only a 30% reduction at a higher dose (45% by weight), according to Baran et al. (2024).

Regarding OCTs, Fig. 2 summarizes the main results obtained. Of note, the DBT content (not graphed) was under the limit of detection in all treatments after 30 days. This observation indicates that the 51 $\mu\text{g}/\text{kg}$ of DBT in the initial sediment sample (Table 2) was degraded in all the treatments after 30 days. Also, a similar reduction in the concentration of MBT was found in all the treatments compared to the control, apart from SN and SCN, where the MBT content showed a greater reduction, especially in SCN. In the case of TBT, all the treatments showed a decrease in the content of this compound compared to the initial values of the sediment (58 $\mu\text{g}/\text{kg}$) (Table 2). However, the differences between the treatments were not significant after 30 days (Fig. 2).

These results (Fig. 2) should be interpreted taking into account not only the effects of the degradation of OCTs but also other factors, including photo-degradation by UV radiation produced during sample manipulation and the experimental time. In fact, hydroxyl radicals are

generated and react with the tin-carbon bond, leading to the removal of a butyl group (Fletcher and Lewis, 1999); for instance, MBT degradation proved to be effective through oxidation by hydroxyl radicals (Brosillon et al., 2014), which could be related to the increase in pH caused by the addition of the amendments in our experiment (Table 3). In addition, UV radiation degrades DBT and MBT twice as fast as TBT (Finnegan et al., 2018). This observation may explain the disappearance of DBT from the sediment; however, MBT could have persisted as a product of the degradation of the other two compounds. An additional effect that may also have occurred in all the treatments, namely the exposure of the sediment to aerobic conditions, resulting in a three times faster degradation than under anaerobic conditions (Brandschl et al., 2001). In particular, the SCN treatment revealed MBT levels under the detection limit after 30 days, thus revealing a synergistic effect between nZVI and coal tailings. These excellent results could be due to the beneficial effects of the coal to improve the degradation of MBT caused by nZVI by adding biostimulation. In fact, adverse, although temporary, effects of nZVI on sediment microbiology (see Castaño et al., 2021 and references therein) could be expected; however, in our case, this could have been corrected by the introduction of an additional carbon source (coal), which might

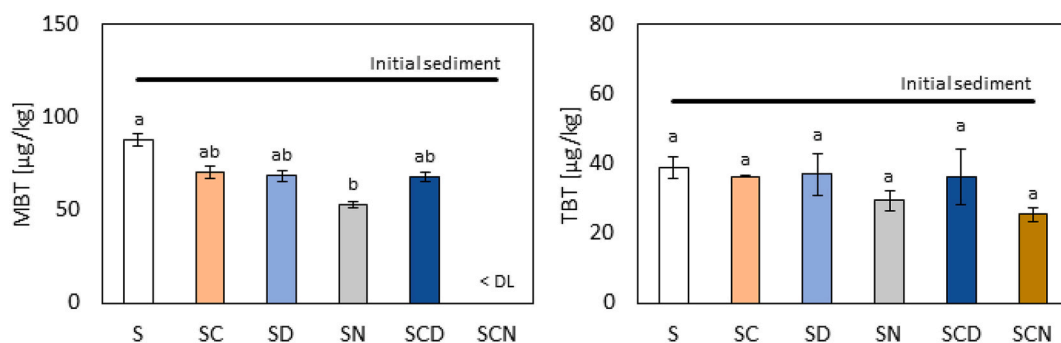


Fig. 2. Concentrations of MBT and TBT after 30 days in the control sediment (S) and treatments with coal tailings (SC), dunite waste (SD), nZVI (SN), coal tailings-nZVI (SCN) and coal tailings-dunite waste (SCD). (<DL: below detection limit, 10 $\mu\text{g kg}^{-1}$).

stimulate certain microbial populations, particularly those involved in MBT degradation. Nevertheless, further research focused on microbial characterization is required to fully elucidate this mechanism. In contrast to metal treatment, the synergy between carbon materials, such as biochar modified with nZVI, has not been widely explored for the treatment of TBT-contaminated soils or sediments. While this approach has been applied to organic pollutants, it has not yet been extended to OTCs (Chen et al., 2024b). However, based on our results, this could represent a promising technique for future research.

3.3. Second remediation strategy: physical soil washing

3.3.1. Grain-size analyses

The samples obtained after grain-size separation were analysed, and the results are presented in Fig. 3 as a percentage of distribution across grain-size fractions. In this regard, all the metal(oid)s studied showed similar distribution patterns across the grain-size fractions (Fig. 3a). Concretely, more than 50% of the metal(oid)s were in the 500–125 μm fraction, with Pb reaching 61% in this range. Comparison of weight recovery with metal recovery revealed similar values for most grain-size fractions, except for the 500–250 μm fraction, in which metal(loid) recovery exceeded weight recovery, and the 125–63 μm fraction, which revealed the opposite trend. A joint analysis showed that the weight recovery above 500 μm was low and the amount of metal(oid)s accumulated was not significant. However, the intermediate particle sizes (between 500 and 125 μm) were the most enriched and, the coarser sizes (500–250 μm) revealed higher contamination than the finer ones (250–125 μm).

Surprisingly, OTCs showed a different behaviour (Fig. 3b) compared to the metals. In this regard, higher concentrations of these contaminants were detected within the coarser grain-size fractions (>500 μm). While variations in the concentrations of different OTCs were found,

with DBT and TBT surpassing MBT, a consistent distribution pattern was observed across the grain-size spectrum for all three types of OTC. Of note, TBT was enriched in the 1000–500 μm grain-size fraction.

However, it is pertinent to consider not only the elevated concentrations within a designated range, but also the relative weight of such fractions, whether substantial or minimal. In this regard, examination of the amount of these compounds within individual grain-size fractions (Fig. 3b) in relation to the weight percentage of the material revealed the predominance of OTCs within the 500–125 μm range, accounting for approximately 60% of the total contaminants. In contrast, the finer ranges (125–63 μm and <63 μm) accounted for around 30% of the contaminants. Finally, less than 5% of the contaminants were found in the coarse fractions exceeding 500 μm . This observation is consistent with the material percentage in each fraction, as less than 5% of the material was present in the coarse fractions, while the higher values were found in medium-size particle size ranges.

Based on these findings, it is evident that employing grain-size separation is effective in targeting and eliminating the most enriched grain-size fractions, thereby reducing the overall concentration of pollutants. However, it is noteworthy that none of the fractions showed concentrations below the limits specified for TBT, as outlined in the Norwegian Guidelines (Direktoratgruppen vanndirektivet, 2018).

3.3.2. Magnetic separation

Consistent with the distribution of contaminants based on grain-size analysis, the fractions ranging from 125 to 500 μm and those <125 μm were chosen for magnetic separation. The results obtained are shown in Table 4.

For the <125 μm fraction, 5% of the initial material was captured in the magnetic fraction (MF) showing higher concentrations of Fe, Cu, Pb, and Zn compared to the feed material. However, the most significant outcomes were observed in the non-magnetic fraction (NMF), which corresponded to 95% of the initial material, thereby highlighting concentrations lower than the initial levels for all elements. Comparison of the concentrations of Cu, Pb, and Zn with the thresholds (Canadian TEL and Norway guidelines) revealed that the former fell below the permissible limits. In the case of OTCs, the analyses were performed on the feed material and the NMF due to the amount of material required, as the mass for the MF was very low. The concentrations of MBT and TBT in the NMF were below those in the initial sediment. Surprisingly, the concentration of DBT in the NMF was below the limit detection of the equipment (0.03 mg kg^{-1}). In conclusion, the remediation technique decontaminated 95% of the material.

For the 500–125 μm fraction, the results were less promising, as concentrations in 97% of the material transferred to the NMF showed similar levels to those present in the feed material. However, higher concentrations of Cu and Zn were evident in the MF. Of note, for both size fractions subjected to magnetic separation, the concentration of Fe increased in the MF compared to the NMF, thereby indicating that the effectiveness of the separation is attributable to the concentration of ferromagnetic materials transferred to the MF. However, similar results were obtained in the case of OTCs, thus the concentrations in the initial sediment were higher than in the NMF, as in the case of the finest particles (<125 μm).

Magnetic separation is a valuable technique for segregating magnetic particles into the MF, which typically binds pollutants. This method facilitates the concentration of pollutants within a smaller volume of material, consequently yielding a larger proportion of clean sediment with minimal or low concentrations of pollutants. Moreover, organo-metallic compounds generally have intermediate to high magnetic susceptibility (Choi et al., 2014) and therefore their concentration by magnetic separation methods is feasible, although the feasibility of magnetic separation has not been successfully demonstrated using WHIMS until now.

To further study the distribution of contaminants between fractions, PXRD analyses were performed, revealing significant crystalline

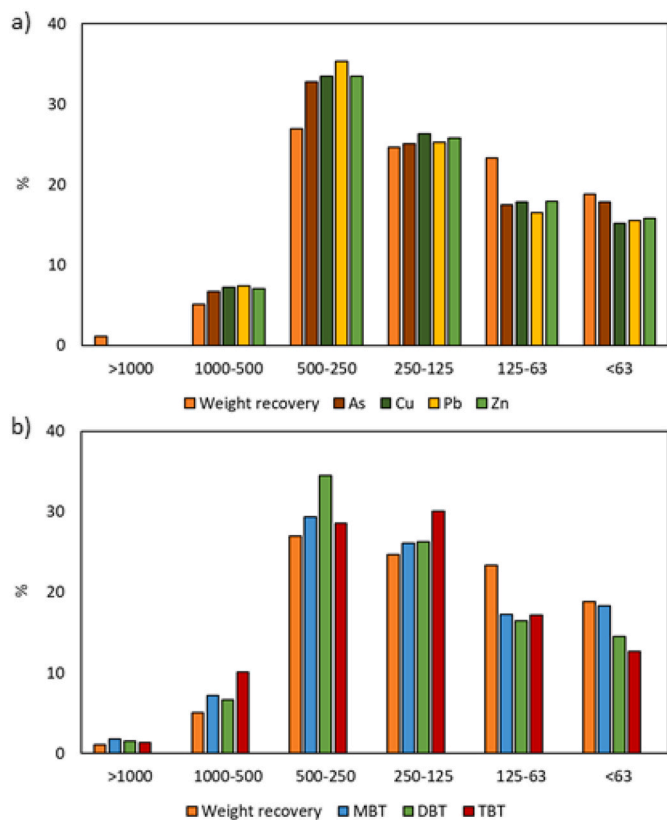


Fig. 3. Weight recovery (WR) and element recovery (ER) for metal(oid)s (top) and OTCs (bottom) in the different grain-size fractions obtained after grain-size separation.

Table 4

Weight recovery (WR) and metal(loids) concentrations in samples originated through the magnetic separation. The threshold levels of the Canadian and Norwegian Guidelines (for the different classes are also shown in the table).

	-	WR	Fe	MBT	TBT	As	Cu	Pb	Zn
		%		$\mu\text{g}\cdot\text{kg}^{-1}$				$\text{mg}\cdot\text{kg}^{-1}$	
TEL						7.2	19	30	124
Norway	I					0–15	0–20	0–25	0–90
Guidelines	II					15–18	20–84	25–150	90–139
Bulk sediment			2.4	120	58	6.4	40	28	140
<125			2.2	162	92	5.2	27	26	107
MF		5	7.0	-	-	5.8	42	33	178
NMF		95	1.9	120	28	3.8	16	20	94
500–125			2.6	241	79	6.5	34	31	125
MF		3	4.5	-	-	6.8	49	34	189
NMF		97	2.6	180	32	6.5	33	31	123

compositional variations between the MF and NMF of different grain sizes (Fig. 4). In the MF from the treatment of the finer grain size fraction (<125 μm), quartz and amphibole were the most abundant mineral phases, while albite, clinocllore, epidote, biotite, and microcline were minor constituents, with hematite potentially present. Conversely, in the non-magnetic fraction, quartz and albite emerged as major phases, accompanied by minor components such as microcline, epidote, biotite, and amphibole, with the possible presence of epidote. In the case of the MF from the coarser grain sizes (500–125 μm), a shift in the dominant phases was detected, with amphibole diminishing in importance while albite and microcline became prevalent, alongside quartz. In addition, the possibility of iron oxide derivatives resembling titanomagnetite increased, particularly in non-magnetic fractions. In these fractions, microcline emerged as a major phase, mirroring trends observed in the finer fractions. Therefore, the presence of hematite and titanomagnetite identified in the magnetic fractions indicates their role as the mineralogical phases responsible for hosting the metal(loids) studied, as these ferromagnetic particles were predominantly associated with the MF.

4. Conclusions

This study aimed to develop innovative approaches for remediating contaminated sediments, comparing the efficacy of chemical stabilization for immobilizing or degrading contaminants with that of physical soil washing, which reduces the volume of contaminated sediment by concentrating pollutants in smaller fractions.

Our findings highlight the high efficiency of nZVI combined with coal tailings for OTC degradation and As immobilization, while acknowledging its limitations for Cu, which requires further optimization. Additionally, dunite waste emerged as a highly cost-effective alternative for immobilizing metals such as Zn, offering a competitive advantage over more commonly used amendments like dolomite, particularly due to its lower required dosage and alignment with circular economy principles.

In the case of physical soil washing, the technique successfully removed coarser fractions (>500 μm) with lower contamination levels. Combined with magnetic separation, it concentrated the most polluted fractions (<125 μm) into only 5% of the initial sediment volume, leaving 95% of the material with significantly reduced contamination. However, the intermediate fraction (125–500 μm) posed challenges, likely due to

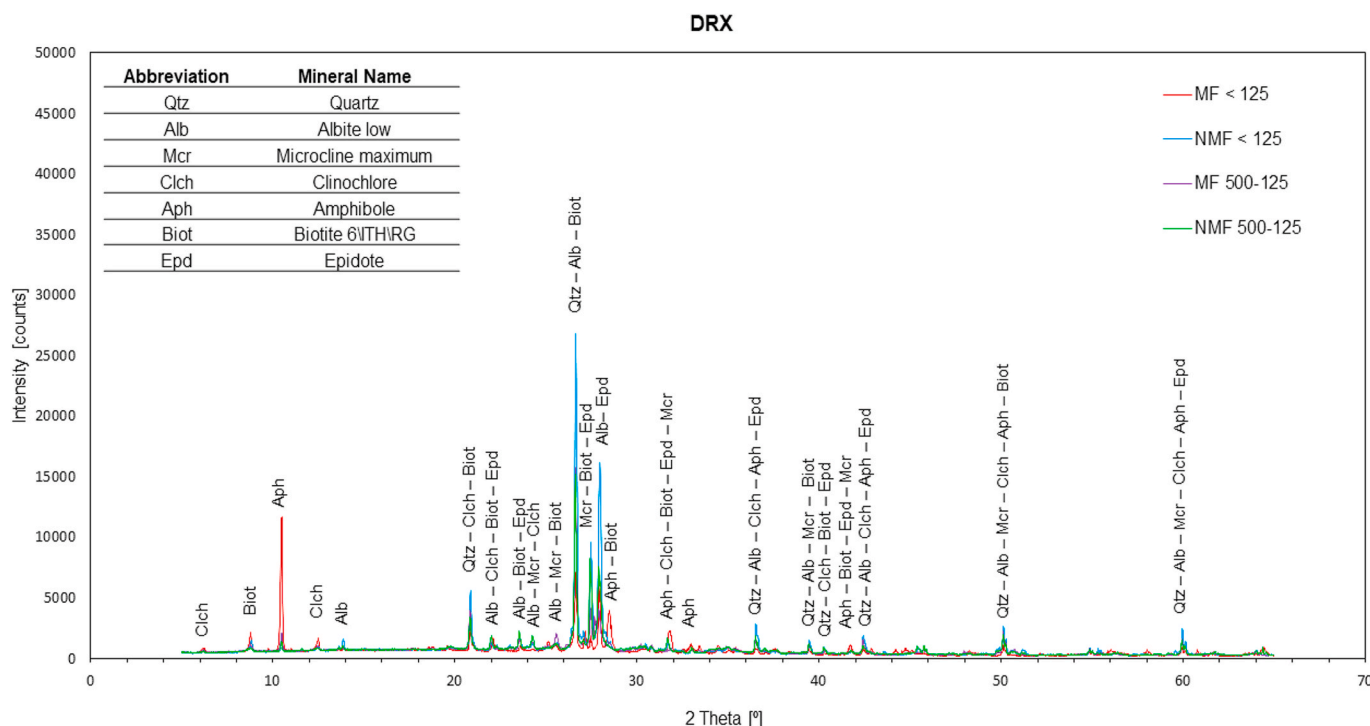


Fig. 4. X-ray diffraction pattern of the samples obtained after magnetic separation (magnetic and non-magnetic fractions).

the mineralogy of ferromagnetic phases, underscoring the importance of prior mineralogical characterization when applying this technique.

In conclusion, the two remediation strategies tested in this study not only demonstrated their individual potential but also their complementarity. Physical soil washing effectively reduces the volume of contaminated sediments, while chemical stabilization mitigates environmental risks by minimizing contaminant leaching. The integration of these approaches could represent a promising pathway for the sustainable management of contaminated sediments in real-world applications.

CRedit authorship contribution statement

A.M. Díaz: Writing – original draft, Investigation, Data curation. **D. Baragaño:** Writing – original draft, Supervision, Data curation, Conceptualization. **J.M. Menéndez-Aguado:** Supervision, Methodology. **A. Norén:** Writing – review & editing, Resources. **K. Karlfeldt Fedje:** Writing – review & editing, Resources. **E. Espín:** Investigation, Data curation. **J.R. Gallego:** Writing – review & editing, Funding acquisition, Conceptualization.

Declaration of competing interest

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

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Data availability

Data will be made available on request.

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