



Trace element mobility in a polluted marine sediment after stabilisation with hydraulic binders



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ABSTRACT

The management of dredged marine sediment is an issue for many harbours, particularly when contaminant concentrations prevent disposal at sea. The stabilisation/solidification of the sediments with hydraulic binders for a use in road subgrade layer is a potential alternative solution. However, the environmental acceptability is not yet fully established. This paper presents the results of a case study to comprehensively determine the stabilisation of As, Cd, Cr, Cu, Ni, Pb and Zn. After demonstrating that stabilisation/solidification affects the microstructure of the sediment, a mobility study is realised (single, sequential and kinetic extractions). According to the regulatory/technical guidelines the studied elements do not pose a risk to the environment. However, results from more complex mobility studies reveal that stabilisation in the short term is only effective for Ni while other elements are mobilised after treatment by at least one type of extraction. Stabilisation in the long term is not universally effective.

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1. Introduction

In Europe, the OSPAR (Oslo-Paris) Commission regulates dredging activities (OSPAR Commission, 2008). Signatory countries to the OSPAR Convention are required to establish national action values for contaminants in dredged material. In France, this was achieved by the GEODE (Groupe d'Etudes et d'Observations sur les Dragages et l'Environnement/Group of Studies and Observations on Dredgings and Environment) which established reference thresholds for disposal at sea. The OSPAR convention recommends that other management options be considered where the characteristics of the dredged material are such that sea disposal is not permitted. In France, this is connected to the waste legislation and must comply with a methodological guide (SETRA, 2011) (Service d'Etudes sur les Transports, les Routes et leurs

Aménagements/a French government technical agency that oversees road, transportation and infrastructure) for the use of alternative materials in roads. Both require analysis of certain substances in the water soluble fraction of waste materials.

The mineralogical and/or chemical speciation of chemical elements within the sediment and the potential remobilisation of those elements from the solid matrix are affected by the change in environmental conditions following dredging and subsequent treatment. Single, sequential and kinetic chemical extractions are widely used as tools to interpret the element mobility in sediments. One important environmental variable is pH. A decrease in pH could be caused by acid rain, occurring either while the sediments undergo dehydration and storage prior to road construction, or in situ following road construction. A rise in pH will occur following the addition of hydraulic binders and/or lime during road construction. Single reagent extractions using e.g. $1 \text{ mol} \cdot \text{L}^{-1}$ HCl are recommended to predict element mobility in marine sediments under acid conditions (Doherty et al., 2000; Scouller et al., 2006; Leleyter et al., 2012), while NaOH can be used to predict element mobility under alkaline conditions (Hamdoun, 2013). Sequential extractions determine the geochemical distribution of the chemical elements, providing information on their affinity for different mineral phases (Ure et al., 1995; Leleyter and Probst, 1999). Kinetic extractions are used to

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investigate longer term element mobility. For example, the equilibrium time for all elements is reached in 24 h when EDTA is used as the extractant (Fanguiero et al., 2002; Cornu et al., 2004; Abi-Ghanem, 2008).

The dilution of contaminants in the sediment is not an acceptable treatment solution (SETRA, 2011). However, the addition of hydraulic binders during the stabilisation/solidification treatment is generally considered to reduce the mobility of elements (Bone et al., 2004; St Laurent et al., 2012). If this approach is taken, a water leach is required to establish the efficacy of contaminant immobilisation and highlight any remaining environmental risk of the material. Chatain et al. (2013) studied the mobility of Cu, Pb and Zn in treated dredged sediment by performing various leaching tests. She reported that only Zn mobility was increased by a pH variation. However, she explained that the treatment could immobilise this element as (oxi)hydroxides and reduced the association to the reducible fraction.

This paper presents the results of a detailed and multidisciplinary case study that follows the treatment and stabilisation of a contaminated marine dredged sediment. The physical, chemical, microstructural and mineralogical characterisations of the raw sediment are first established along with the mobility of As, Cd, Cr, Cu, Ni, Pb and Zn in the raw sediment. These elements are often present in dredged sediment and are targeted by GEODE thresholds and the SETRA methodological guide as they are of environmental concern. The acceptability of the treatment in terms of geotechnical and environmental parameters is then established.

The approach is therefore innovative with regard to the existing studies on reuse of sediment in civil engineering which focus only on the environmental or geotechnical aspects (Marot, 1994; Boutouil, 1998; Rey, 1999; Colin, 2003; Lemée, 2006; Agostini et al., 2007; Rezik, 2007; Lafhaj et al., 2008; Nguyen, 2008; Duan, 2008; Scordia et al., 2008; Tran, 2009; Brakni et al., 2009; Zentar et al., 2009; Agostini et al., 2010).

2. Material and methods

The sediment was dredged in June 2010 from the military port of Cherbourg, France. Approximately 600 L of dredged material was stored in several 70 L sealed plastic barrels.

2.1. Sediment preparation and treatment

2.1.1. Preparation of the raw sediment

The sediment initially contained a large amount of water (approximately 100% of the dry matter weight). Sediments were dehydrated in an oven at 40 °C. Clumps formed during drying were removed by crushing and sieving to 2 mm producing a test material of 0/2 dry sediment. Representative sub-samples were taken from this material for geochemical, mineralogical and geotechnical characterisation. The raw sediment was also characterised by loss of mass on ignition (LOI) (organic matter content measurement), particle size distribution and Atterberg limits. These parameters allow a classification according to the Unified Soil Classification System (USCS).

2.1.2. Stabilisation and solidification treatment

The raw sediment was treated with 3% of quicklime and 6% of cement CEMII/B 32.5R with respect to the dry mass of the mixture. After treatment, geochemical and microstructural analyses were undertaken on fragments of cylindrical samples of 50 mm in diameter by 50 mm in length, compacted at Optimal Moisture Content (OMC) and 96% of the Maximal Dry Density (MDD). These samples are initially used to determine mechanical strengths evolution of the treated sediment. Indirect tensile strength (ITS, application of a stress until the splitting failure) of the treated sediment is measured according to standard EN 13286-42, on a press with a 50 kN sensor. The standard Proctor optimum evolved from 19.5%; 1.62 t·m⁻³ before treatment to 20.4%; 1.57 t·m⁻³ after treatment. The MDD decrease is theoretically due to

the flocculation induced by the addition of the quicklime (LCPC-SETRA, 2000). Replicate samples were cured in a hermetically sealed mould at constant temperature (20 ± 2 °C) with an average relative humidity of 98%. Microstructural and mineralogical analyses were performed on treated sediment after 90 days of hydration and physico-chemical analysis after 180 days.

2.2. Mineralogical and microstructural analysis by automated mineralogy

Mineral identification was achieved using the Quantitative Evaluation of Mineralogy by Scanning Electron Microscopy (QEMSCAN®) technique for automated mineralogy. The technique is based on the point by point acquisition of energy dispersive spectra across a sample. Each spectrum is allocated to a specific mineralogy and sample mineralogy is inferred from the chemical spectra acquired (Rollinson et al., 2011) and false colour images allow the visual comparison of samples (van Veen et al., 2013a, 2013b). A review of the QEMSCAN® applications is given by Pirrie et al. (2009) and further details can be found in Gottlieb et al. (2000); Pirrie et al. (2004) and Pirrie and Rollinson (2011).

Samples were analysed on the QEMSCAN® 4300 system at the University of Exeter using particle mineralogical analysis (PMA) and field scan (FS) operating modes. Data collection was computer controlled using iMeasure software. Data were processed to produce a simplified mineral/phase list using iDiscover software. PMA operating mode systematically maps the composition of each discrete particle within the prepared sample providing quantitative data on mineralogy, particle grain size and shape. This was used to analyse both the raw sediment and solid residues following extraction with HCl (see Section 2.3). FS operating mode was used to map the monoliths (blocs or fragments) produced following sediment treatment after 90 days of hydration.

2.3. Geochemical analysis and extraction of the raw and treated sediment

The geochemical analysis focused on As, Cd, Cr, Cu, Ni, Pb and Zn. Total concentrations of these elements are required for assessment according to the GEODE thresholds values and can also be used to calculate the percentage of elements leached by the chemical extractions, thus estimating their operational mobility (Eq. (1)).

$$\text{Mobilised X (\%)} = 100 \times [\text{X}]_{\text{leached}} / [\text{X}]_{\text{total}} \quad (1)$$

Total concentrations were determined by solubilising 0.2 g of dry sediment in aqua regia (3.33 mL of conc. HNO₃ (68%) and 6.66 mL of conc. HCl (35%)) using microwave digestion (Berghof speedwave MWS-2). The analytical quality of the data was controlled using certified reference material HR-1 (Canada Centre for Inland Waters National Laboratory for Environmental).

Single batch extractions were performed at room temperature with pure water, 1 mol·L⁻¹ HCl (pH = 0) and 0.1 mol·L⁻¹ NaOH (pH = 13). A liquid to solid (L:S) ratio of 10:1 was used in each extraction. Where pure water was used as the extractant, the mixture was shaken for 24 h following EN 12457-2 standard; for the extractions using HCl and NaOH, the mixtures were shaken only for 1 h (Leleyter et al., 2012; Hamdoun, 2013).

Sequential extraction reveals the geochemical partitioning of the elements. Sequential extraction method described in Leleyter and Probst (1999) was chosen among several procedures because it was checked for selectivity, reproducibility and repeatability of the different steps and it was commonly used in literature (Bur et al., 2009; Cecchi et al., 2008; N'guessan et al., 2009; Salvarredy-Aranguren et al., 2008). The procedure dissolves selectively and efficiently all the chemical constituents of the sediment which can be affected by changes in physicochemical conditions (Leleyter and Probst, 1999; Leleyter and Baraud, 2005). So the successive extractions allow defining chemical affinity of the elements in the following order: elements dissolved by water (F1) which

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