



Effects of mineral amendments on trace elements leaching from pre-treated marine sediment after simulated rainfall events[☆]



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ABSTRACT

Bauxite extraction by-products (red mud) were used to evaluate their potential ability to stabilize trace elements from dredged and aerated/humidified marine sediment. The investigated by-products were: bauxaline[®] (BX) that is a press-filtered red mud; bauxsol[™] (BS) that is a press-filtered red mud previously washed with excess of seawater, and gypsum neutralized bauxaline[®] (GBX). These materials were separately mixed to dredged composted sediment sample considering 5% and 20% sediment: stabilizer ratios. For pilot experiments, rainfall events were regularly simulated for 3 months. Concentrations of As, Mo, Cd, Cr, Zn, Cu, and Ni were analyzed in collected leachates as well as toxicity. Results showed that Cd, Mo, Zn, and Cu were efficiently stabilized in the solid matrix when 20% of BX, BS, and GBX was applied. Consequently, toxicity of leachates was lower than for the untreated sediment, meaning that contaminants mobility was reduced. A 5% GBX was also efficient for Mo, Zn and Cu stabilization. In all scenarios, As stabilization was not improved. Compared to all other monitored elements, Mo mobility seemed to depend upon temperature-humidity conditions during pilot experiments suggesting the need of further investigations.

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1. Introduction

In the mid-1960s, environmental concerns arose in most European countries and national governments started to be more active in their attempts to monitor and control environmental pollution. Major focus was devoted to develop and implement environmental quality guidelines in policies and regulations (Van Wezel, 1999). Special attention was dedicated to the disposal of sediment resulting from dredging activities of port channels and seaways in order to maintain maritime navigation. Yearly, in France approximately 50×10^6 m³ of sediment are dredged out (Alzieu, 1999; Duclay et al., 2010). Recent studies have shown that the Mediterranean coastal sediment can be heavily contaminated due to industrial, port and anthropogenic activities (Andral et al., 2004; Tessier et al., 2011; Pougnet et al., 2014). Sediment pollution is

associated with potential economic, social and environmental problems (Eggleton and Thomas, 2004; Förstner, 2006; Arizzi Novelli et al., 2006; Libralato et al., 2008; Lofrano et al., 2016a). The management of dredged sediment must comply with the World London Convention for the prevention of marine pollution as a result of waste dumping (Duncan, 1973), and the OSPAR convention for the protection of marine natural environments of the North-East Atlantic (OSPAR, 1992). Sediment monitoring can be carried out through physico-chemical analyses and toxicity testing that became a widespread regulatory requirement of potential environmental hazards helping in decision-making about contaminated sediment (Prato et al., 2015). In France, dredged materials are managed considering quality guidelines including two regulatory levels (N1 and N2) defined on the basis of contaminant concentrations (trace elements and PCBs) in sediment. When the concentration of pollutants is <N1, sediment is classified as lightly contaminated with no significant impacts on the environment. Sediment is considered as contaminated if pollutants are between N1 and N2 levels thus dredging impacts must be investigated specifically. Highly contaminated sediment are

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ranked > N2. Toxicity testing is necessary in the case of contaminated and highly contaminated sediment to assess their potential environmental impact. To facilitate sediment risk assessment, several toxicity tests have been proposed such as bivalve embryotoxicity and Microtox[®] assays (Alzieu and Quiniou, 2001; Libralato et al., 2008). When the pollutant concentrations found in sediment is >N2, direct landfill is prohibited, and contaminants must be immobilized before dumping on land (Alzieu, 2005). For this reason, stabilization/solidification techniques involving the use of mineral amendments were found to be cost effective and promising treatments. They amplify the rate of stabilization by enhancing adsorption, precipitation and complexation reactions onto soil or sediment components (Peng et al., 2009; Scanferla et al., 2009). The most studied mineral amendments are phosphate materials, alumino-silicates (clay and zeolites), alkaline materials and iron bearing compounds (zero valent iron, goethite, hematite, and ferrihydrite). Alumino-silicates and iron-based materials are able to immobilize the highest number of pollutants (Kumpiene et al., 2008; Komarek et al., 2013; Mamindy-Pajany et al., 2013).

In a previous work, sediment toxicity after stabilization with mineral amendments was evaluated using Microtox[®] solid phase test which was widely used to evaluate the toxicity of polluted sediments (Doherty, 2001; Onorati and Mecozzi, 2004; Gonzalez-Merchan et al., 2014). Dredged sediment samples were first aerated/humidified for 4 months to biologically reduce the organic contamination (i.e. pre-treatment), and then stabilized with three commercial mineral additives: i) hematite; ii) zerovalent iron; and iii) zeolite. Results showed that all mineral additives acted as stabilizing agents decreasing the levels of dissolved metal concentrations and sediment toxicity, but their employ could be expensive due to the use of commercial products (Mamindy-Pajany et al., 2012). Amongst the non-commercial potential additives, red mud (RM) could represent an interesting solution. RM is a very alkaline by-product obtained after bauxite ore extraction (according to Bayer process) for aluminum (Al) production. It is produced in huge amounts since 1 ton of extracted Al generates in 0.5–1 ton of RM (Genç et al., 2003). It is characterized by the presence of Fe- and Al-oxy-hydroxides making this by-product an interesting material to be reused for metals or organics removal, for hydrogenation, or as an additive for cement and brick industries (Singh et al., 1993; Llano et al., 1994; Alvarez et al., 1995; Peng et al., 2005). Considering the amount of the produced RM and its adsorption properties, research activities on its potential reuse as a cost effective and safe material for environmental purposes are of great interest for Al producers. Actually, less than 3% of bauxite residues produced annually is used in a productive way (Evans, 2016).

On the basis of Mamindy-Pajany et al. (2012), the aim of this research study was to investigate the use of RM as a low cost mineral amendment in *ex situ off site* stabilization of trace inorganics in pre-treated dredged sediment samples. Pilot scale experiments were carried out for 3 months considering various RM formulations (i) bauxaline[®] (BX); ii) bauxsol[™] (BS); iii) bauxaline[®] neutralized by the addition of gypsum (GBX). BX, BS and GBX were mixed with dredged sediment including two “additive: sediment” ratios: i) 5% and ii) 20%. Treated sediments were regularly exposed to simulated rainfall events and leachates were collected and measured in order to evaluate its feasibility to stabilize sediment inorganic pollutants to be landfilled.

2. Material and methods

2.1. Sample collection

Sediment samples resulting from dredging activity in the Toulon French Navy harbor (Mediterranean Sea) were considered. After

dredging, sediment was kept on land and regularly aerated/humidified for 4 months by port authority. This procedure consisted in i) sediment mixing once a week promoting microbial consortia growth and, thus, biological degradation of contaminants as well as lowering salt and organic matter content, and ii) sediment humidification with tap water. Two sub-samples of this sediment were collected to carry out laboratory scale experimental activity. Sediment was already fully characterized (Mamindy-Pajany, 2010). Data from X-ray diffraction (XRD) of dredged sediment indicated the presence of calcite, feldspar, quartz and illite minerals. Mineralogical analysis of the fraction <2 μm showed that sediment clay fraction contained illite (73%), kaolinite (19%) and smectite (8%) (Mamindy Pajany et al., 2010). According to Table 1, sediment is highly contaminated by inorganics and can be classified as >N2 level for inorganic pollutants requiring *ex situ off site* management (Taneez et al., 2015, 2016).

2.2. Industrial by-products used as mineral amendment

We used an industrial by-product from bauxite extraction as mineral amendment to stabilize trace elements in marine dredged sediment. Bauxite by-product, commonly known as RM, is generated during alumina production from bauxite ore. RM is highly alkaline (pH 10–13), reddish brown in color with a fine particle size distribution containing Al, Fe, Si, and Ti oxides and hydroxides (Nadaroglu et al., 2010).

ALTEO plant (Gardanne, Bouches-du-Rhône, France) that supplies specialty alumina since 1893 from bauxite, provided RM for this study. Press-filtered raw RM is commercially known as bauxaline[®] (BX) and its composition is given in Table 2. Despite its interesting mineral composition, BX use has been hindered because of its high alkalinity. To overcome this limitation, neutralization treatments have been investigated. One of these treatments consists in promoting the formation of hydrotalcite, calcite and brucite from RM by precipitation of alkaline species from seawater (i.e. soluble Ca²⁺ and Mg²⁺). Using excess seawater resulted in neutralization of RM to approximately pH 8.5 (Burke et al., 2013; Kirwan et al., 2013). In this study, the resulting neutralized RM was bauxsol[™] (BS) (Table 2). Another possible treatment consists in pH neutralization by gypsum addition (CaSO₄·2H₂O) that is a soluble Ca²⁺ source expected to buffer BX alkalinity similarly to seawater effect (Kirwan et al., 2013). The resulting neutralized BX was called “gypsum neutralized BX” (GBX) (Table 2). For this study, BX and BS were provided by ALTEO plant (Gardanne, France), and GBX by INERIS Méditerranée (France).

2.3. Experimental setup

Experiments lasted 3 months and were conducted in Nice

Table 1

Total content of elements in sediment sub-samples (ND: not defined) (dark grey cells: concentration > N2). Data for 1st sediment sub-sample are from (Taneez et al., 2015); and for 2nd sediment sub-sample from (Taneez et al., 2016). Regulatory levels are from (Alzieu and Quiniou, 2001) and (Alzieu, 2005).

Elements	1 st sediment sub-sample (mg/kg)	2 nd sediment sub-sample (mg/kg)	French regulatory levels (mg/kg)	
			N1	N2
As	150	201	25	50
Cd	4.7	4.8	1.2	2.4
Cu	1721	1881	45	90
Mo	9.1	7.4	ND	ND
Ni	27	21	37	74
Zn	1869	3069	276	552
Cr	59	43	90	180

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