



Evaluation of phytotoxicity of seaport sediments aged artificially by rotary leaching in the framework of a quarry deposit scenario



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ABSTRACT

In the framework of an ecological risk assessment of seaport sediments for terrestrial ecosystems when deposited in quarries, we simulated the “ageing” of sediments exposed to rain. This experiment highlighted an inflection point at the solid/liquid ratio 1/25, after which the extraction of pollutants increases moderately. The raw sediments studied inhibited the germination of *Lolium perenne* and *Armeria maritima* (a halophytic species) seeds. Furthermore, they affected the early development of *L. perenne*. The same sediments, leached at a ratio of 1/25, presented a reduction of acute (germination) and chronic (growth) phytotoxicity. The bioconcentration factors of the metals studied decreased with the leached sediment, except for Cu which was still clearly identified in root parts. Thus rotary leaching tests and phytotoxicity bioassays can be used to provide an initial assessment of the ability of plants, particularly halophytes, to colonize deposits of dredged seaport sediments.

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1. Introduction

Seaport sediments are often highly contaminated by heavy metals (Andral et al., 2004; Schintu et al., 2009) and organic compounds such as organotins, PAHs and PCBs (Gomez-Gutiérrez et al., 2007; Mille et al., 2007; Cassi et al., 2008). In France, regulations for the management of dredged marine sediments are based on two levels of contamination (N1 and N2) set out in the French Decree of 14 June 2000 “relating to the reference levels to be taken into account for their management” (Alzieu and Quiniou, 2001). The conditions for using these thresholds are the following: (i) below level N1 the potential impact is deemed negligible, since the contents are considered to be comparable to environmental background contamination; (ii) between level N1 and level N2 additional investigation may be necessary depending on the project considered; (iii) above level N2, additional investigation is necessary since the significant indices recorded give rise to the assumption that the operation has a potentially negative impact. It is therefore necessary to perform a specific study focused on the sensitivity of the environment to the substances concerned, with in particular the assessment of the foreseeable impact of the latter. Dredged sediments above threshold N2 cannot therefore be discharged into the sea and must be treated before storage on

land. Sediments with levels between thresholds N1 and N2 can be considered as being in the same situation as a function of the project and the local context. In both cases, the potential impact of these sediments during the time they are managed on land must be assessed (Pejñeneburg et al., 2005). This assessment concerns in particular the evaluation of the ecotoxicological effects linked to sediments and/or the emissions from sediments (Gourmelon et al., 2003). Nonetheless, relevant legislation as yet fails to define an ecotoxicological test to assess the impact of sediments on terrestrial environments.

The main problem is to evaluate the effects of the initial sediment deposited, sediment ageing due to climate and the impact of rain on representative terrestrial organisms. To do this it is necessary to evaluate or mimic the ageing of sediment and its pollutant behavior, especially in the case of dredged sediment deposited on soil. Column leaching has been used to study the mobility of toxic elements on industrially contaminated land and to evaluate the predictive mobility of metals in run-off in urban soils and dredged sediments (Tack et al., 1999; Anderson et al., 2000). Several factors can influence the leaching of sediment deposits such as rainfall frequency and the alternation of humidity/dryness (Piou et al., 2009). Leaching experiments on contaminated marine sediments performed in a bath reactor were conducted as a function of contact time between sediment and ultra pure water according to the French standard AFNOR NF XP X31-210 (Mamindy-Pajany et al., 2008). They showed that arsenic

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and major elements are rapidly leached but in low concentrations ($15 \mu\text{g L}^{-1}$) and in an alkaline medium with an equilibrium pH equal to 8 (Mamindy-Pajany et al., 2008). Mamindy-Pajany et al. (2010) also used elutriates for the ecotoxicological evaluation of dredged Mediterranean seaport sediment, and their elutriates were prepared according to the AFNOR standard protocol with artificial sea water as leaching medium with a ratio of 1:10 [sediment:water] (dry mass weight of sediment). Previous works focusing on dredged marine sediment have shown that column leaching significantly decreases the germination inhibition of some species and that it can be used to assess the capacity of these sediments to allow colonization by plants, notably *Lolium perenne* (Bedell et al., 2013). However, mimicking rain contact in columns is still limited (in terms of annual water contact) and time consuming (from several days to several months percolation). Other possibilities, such as rotary leaching, can be less time-consuming for simulating several years of water contact. Such dynamic leaching tests provide insight into the dynamics of metal release under laboratory conditions, and such metal release in dredged sediment can be related to a timescale by increasing the cumulative liquid to solid ratio (L/S ratio) (Van der Sloot et al., 1996; Tack et al., 1999; Bedell et al., 2009).

The choice of organisms to be tested is also very important when evaluating the ecotoxicity of contaminated sediments. Studies on seed germination are considered representative of short term impacts and above all allow evaluating the effects of acute toxicity. These tests have demonstrated a significant reduction of the germination of certain species in soils and sediments contaminated by different metallic trace elements (Adam and Duncan, 2002; Chen et al., 2002). Plant germination and growth tests are bioassays often performed in the domain of ecotoxicology to identify acute toxicity. These bioassays assess in particular the potential inhibition of seed germination by polluted solid matrixes. In addition, different studies have shown the usefulness and efficiency of these phytotoxicity tests for evaluating the toxicity of environmental (soils, sediments) and anthropic (compost, wastewater treatment plant sludge, sewage sludge) matrixes (Fuentes et al., 2004; Bedell et al., 2006; Czerniawska-Kusza et al., 2006; Oleszczuk et al., 2012). Lastly, in addition to the contaminants present in marine sediments, residual salinity can harm plant life in the same way as sediment heavy metal content (Tiquia and Tam, 1998; Chen et al., 2002). Such phytotoxicity tests must be also performed with halophytic plants, such as *Armeria maritima*, that is to say plants adapted to salty media (Bedell et al., 2014).

Our study was carried out in the framework of the “SEDIGEST” (Sustainable management of sediments dredged in seaports) project.¹ This project, financed by the French program ANR PRECODD, is intended to formulate a method for assessing the potential risks of treated seaport sediments to aquatic and terrestrial ecosystems in a quarry fill scenario (Perrodin et al., 2012). In the framework of this scenario, our specific objective presented in this paper was to determine the level/time from which deposited sediment could be colonized by plants. Consequently, this entailed simulating the “ageing” of these sediments, particularly under the action of rain, until reaching optimum conditions at which plants can germinate and colonize the deposit. Firstly, different solid/liquid ratios were obtained by simulating the exposure of two sediments 1 and 2 to increasing amounts of water, i.e. volumes of rainfall, which were then carried over to the annual average in order to estimate the number of years corresponding to these volumes of water. This is why we simulated the leaching of sediments by water using a rotation leaching device, in order to obtain sediments “aged” by leaching from several months to several years. Then we characterized these

ratios chemically, especially for chlorides (desalination), and several other parameters (pH, DOC, absorbance, etc.), and pollutants (Zn, Cd and Cu concentrations). Accordingly, we chose a ratio that would permit us to monitor both germination (acute test) and early growth/development (root elongation) with *L. perenne* and *A. maritima*. This assessment of phytotoxicity was also performed for several weeks (chronic test) on initial sediment 2 and aged artificially at the ratio chosen, by carrying out germination and growth tests with *A. maritima* to determine the effect of certain elements on the plant (e.g. bioaccumulation).

2. Materials and methods

2.1. Sediments

2.1.1. Choice and characteristics

The sediments studied were chosen firstly for their representativeness of the treatment solutions considered for their management (SEDIMARD Program²), and, secondly, for their level of contamination (the requirement for a response in the framework of this methodological development program) and their physicochemical characteristics. Thus the sediment used here came from the SEDIMARD program in which these three treatments were performed after sieving to eliminate the coarse fraction (gravels, etc.). Sediment 1 (Toulon 0–20) came from the French port of Toulon (Var, 83), and was dredged in April 2008, dried and aired for 5 months (April to September 2008) and then screened at 0–20 mm (end of November 2008). Sediment 2 (Toulon fines) also came from the port of Toulon. It was taken on 31 March 2006, cleaned (beginning of April 2006), and dried and aired for 5 months (April to September 2006). The main characteristics of these sediments after these treatments, notably their contaminant contents, are presented in Table 1 which also includes the two levels of contamination (N1 and N2) of the French Decree of June 14, 2000. The physico-chemical characteristics of the sediments were obtained previously at the end of SEDIMARD program by using several protocols or standards, i.e. NF EN ISO 6468 for PCB determination, NF EN ISO 11885 for trace element determination, ISO/WD/7981 for PAH, and ISO 17353 for organotin compounds.

These two sediments present high levels of contamination in comparison to thresholds N1 and N2 (Table 1). Thus the values of Cu, Pb, PCB (e.g. PCB 118 and PCB 153), tributyltin and Benzo[k]fluoranthene and Benzo[a]pyrene were higher than thresholds N1 and N2 (Table 1). These sediments with high levels of contamination were chosen at the beginning of the study to generate a clear response from the plants in view to validating the methodology. Thus the level used does not correspond to a level of contaminant pollution for sediments capable of being used to fill-in terrestrial quarries (for example, pollutant concentrations below levels of acceptance in landfills for inert waste).

2.1.2. Sediment ageing

The objective was to simulate the ageing of sediments treated by subjecting sediment samples to given quantities of “contact water” (representative of a given rainwater exposure time) so as to have available sediments representative of different storage durations under site conditions. In the framework of the scenario considered, given an average rainfall of 1 m/year, i.e. $1 \text{ m}^3/\text{m}^2/\text{year}$, and with an evapotranspiration rate of 70%, i.e. 30% of infiltration in the sediment layer, we obtained an annual liquid/solid ratio of 0.3 for the upper soil layer of the deposit supporting the vegetation.

¹ <http://www.sedigest.org>.

² SEDIMARD Program: http://www.ports-developpementdurable.com/ports_plaisance_developpement_durable/2009/res/aqua.pdf 2.

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