



# Capping of marine sediments with valuable industrial by-products: Evaluation of inorganic pollutants immobilization<sup>☆</sup>

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## ABSTRACT

In-situ capping of polluted sediment is considered as an inexpensive and effective treatment technology to immobilize contaminants in a short time. In this remediation technique sediments are capped by placing a layer of sand, clean sediment or other materials over sediments in order to mitigate risk. In this study, low cost industrial by products (bauxaline, steel slag, and mixture of the two products) were applied as capping agents. A bench scale laboratory experiment in aquariums was performed to evaluate their effects on Cd, Zn, As, and Cr mobility from an artificially contaminated marine sediment. Without capping, all the contaminants are constantly released with various kinetic depending of mineral oxidation or dissolution or leaching. Nevertheless, release did not exceed 31% of the initial amount of pollutant. Capping sediment with steel slag, bauxaline and their mixture totally captured Cd, Zn, and As. In the case of Cr, only steel slag actively blocked its release. A kinetic model was developed to model As and Cr release, with and without capping. The release times for Cr and As from the sediment were close to 6 days. In the presence of capping agents, the capture time for Cr was found to be 57 days for steel slag, and 7 days for bauxaline. Despite a high capture time, steel slag was the best capping agent since bauxaline matrix was a source of Cr and rapidly released it (release time = 1 day). The results indicated that steel slag and its mixture additive can be used as potential capping materials for the remediation of contaminated sites due to their significant entrapping of Cd, Zn, As, and Cr.

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## 1. Introduction

Coastal ecosystems, particularly the continental shelf and rocky reefs, are the most threatened, due to their high vulnerability to the cumulative effects of coastal urbanization and overfishing (Halpern et al., 2008). Artificial reef deployment started in Europe 40 years ago for conservation and restoration of fish habitats. France was among the first European countries to carry out experiments on artificial reefs. In earlier 1968, approximately 90,000 m<sup>3</sup> of reefs have been deployed at 20 sites along the French Mediterranean coast. Artificial reef construction in the Mediterranean Sea focused on fish stock enhancement and fishery management. Mostly,

artificial reefs are made of specially designed concrete units while some pilot reefs were made of waste materials (Tessier et al., 2015). In early 1980's, around 8000 m<sup>3</sup> of tyre reefs were deployed between 24 and 37 m deep on 24 ha in the marine protected area of Vallauris-Golfe Juan (NW Mediterranean Sea) (Risso-de Faverney et al., 2010). Construction of artificial tyre reefs aimed to improve habitat conservation but in later year's concern about pollution arise due to possible leaching of tyre components. Tyre are synthesized of natural rubber, synthetic polymers, carbon black, high aromatic oils, sulphur, zinc oxide, heavy metals, organic peroxides (Collins et al., 1995). Leaching of tyre components have toxic effects on various aquatic organisms (Gualtieri et al., 2005; Nelson et al., 1994; Wik et al., 2009). However, few studies have examined the effects of leached chemical compounds from tyre reefs in seawater (Collins et al., 1995; Collins et al., 2002; Risso-de Faverney et al., 2010). On the request of Alpes-Maritimes General Council in 2007, potential toxic effects of tyre reefs in the Golfe Juan were investigated using mussels (*Mytilus galloprovincialis*)

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transplantation at two tyre stations, and reference site. Biochemical analysis and physiological status of mussels indicate that the mussels transplanted at tyre reef station were more affected by pollutants leaching than at reference site (Risso-de Faverney et al., 2010).

To avoid further pollution in the marine protected area of Golfe Juan, these results advocate in favor of tyre dredging. Therefore, in May 2015, authorities have started a pilot scheme to remove around 2000 tyres from the French Riviera coast, i.e. 10% of the total number of tyres. The main objective is to assess the feasibility of such a dredging and its consequences on the natural habitat. Indeed, dredging and natural events can enhance mobilization of contaminants trapped in the sediment matrix and these contaminants have adverse effects on aquatic organisms (Eggleton and Thomas, 2004). Trace metals are bound with different geochemical fractions in sediment such as carbonate, reducible, organic matter, sulfide and residual phases (Tessier et al., 1979). The mobility and eco-toxicity of trace metals in natural system is dependent on environmental conditions which in turn influence chemical speciation and reactivity towards carrier phases rather than their total content (Du Laing et al., 2009; Qian et al., 2009). Therefore, these contaminants need to be immobilized or remediated from sediments to minimize harmful impacts. The sediments quality in France is evaluated on the basis of two regulatory levels naming N1 and N2 fixed for the concentration of heavy metals and PCBs (Alzieu, 2005).

Generally adopted sediment remediation approaches include dredging, capping of contaminated areas, and monitored natural recovery (MNR). Dredging can remove contaminated sediments but it requires large areas of land for sediment disposal. It is expensive and may cause secondary contamination of the water column during re-suspension (Francingues et al., 2008; Ghosh et al., 2011). MNR relies on ongoing naturally occurring processes to decrease the bioavailability or toxicity of contaminants in sediment. These processes may include physical, biological, and chemical mechanisms that act together to reduce environmental risks posed by contaminated sediments. MNR require longer time monitoring and can be even more expensive than for dredging and capping (Magar et al., 2009). Capping consist of in situ covering of clean or suitable isolating material over contaminated sediments layer to limit leaching of contaminants, and to minimize their re-suspension and transport. Compared to MNR and dredging this treatment method is cost effective, less disruptive and less time consuming (Ghosh et al., 2011; Palermo, 1998).

Activated carbon (AC) is widely used for remediation purposes due to its high adsorption abilities for a large number of heavy metal ions. However, since the price of AC is relatively high their usage for capping operations is not an economically valid alternative. This has led many researchers to seek for low cost materials such as naturally occurring mineral and industrial by-products (Cetin and Pehlivan, 2007). In case of organic pollutant, AC in laboratory and field trial reduced the risk of polycyclic aromatic hydrocarbons (PAHs), mercury and methyl mercury uptake in contaminated sediments by the organisms. Strong sorption to AC particles leads to reductions in pore water concentration (Cornelissen et al., 2011; Ghosh et al., 2011; Gilmour et al., 2013). Other mineral amendments (sand, limestone, aquablok™, and bauxite residue) also gave satisfactory results for organic pollutants (Eek et al., 2008; Ghosh et al., 2011; Lampert et al., 2011; Randall et al., 2013). On the other hand, metals are non-biodegradable and are difficult to immobilize, the efficacy of treatment may vary due to the critical combination of elements. Addition of mineral additives such as zeolites, apatite, fly ash, zero valent iron, goethite, hematite, and ferrihydrite can reduced the availability of contaminants and toxicity by changing their chemical speciation in

sediments (Mamindy-Pajany et al., 2013; Qian et al., 2009; Shin and Kim, 2016). Industrial by-products (fly ash and bauxite residue) can attenuate cationic pollutants (Cd, Zn, Cu, Pb, and Ni) bioavailability, and reduce plant metal uptake in multi-contaminated acidic soils and sediments (Qiu et al., 2012; Shin and Kim, 2016; Taneez et al., 2016). High rate of amendments (bauxite residues) significantly decreased the eco-toxicity of marine dredged sediment by stabilizing the cationic elements (Hurel et al., 2017), but some limitations in case of As, Cr and Mo were highlighted (Taneez et al., 2015, 2016).

Mostly researchers used mineral additives as mix additive in sediments for ex-situ application but the research on the use of low cost industrial by-products for in-situ capping of multi-metal contaminated sediments is limited. Recently, naturally occurring minerals Cd and Pb were stabilized in laboratory microcosms tests by mixing natural calcium-rich clay minerals (sepiolite and attapulgite) with sediments for short period of time i.e., 15 days and observed that  $\text{Ca}^{2+}$  rich minerals can induce metal exchange in sediments and reduce metal uptake by benthic organisms (Yin and Zhu, 2016). Zeolite, montmorillonite, and steel slag were placed as cap layer of varying thickness from 1 cm to 5 cm over metal contaminated marine sediments in flow tank systems for 30 days. The study reported that cationic pollutants were effectively blocked but the bioavailability of arsenic was not reduced (Kang et al., 2016). However, additional research is needed to determine future requirements for improved results, mode of amendments application (capping directly or mixed with sediment), thickness and use of new minerals amendments in different mixtures ratios. Therefore, in the present study selection of capping agents (industrial by-products) was made on the basis of their mineral composition, high sorption efficiency, low cost, and easy availability. The effectiveness of these industrial by-products i.e., bauxaline, steel slag and mixture of steel slag/bauxaline as thin caps to capture toxic pollutants; cadmium (Cd), zinc (Zn), arsenic (As), and chromium (Cr) was evaluated in aquariums for 90 days. The results of the study will help to provide cost effective in-situ remedial measure for multi-contaminated sediments.

## 2. Materials and methods

### 2.1. Capping materials

Industrial by-products are produced in large quantities all over the world and their utilization in pollution control has attracted global attention due to their low cost. Two industrial by-products such as bauxaline (red mud) and steel slag were used for capping and their composition is given in Table 1. The bauxaline sample was provided by an international company ALTEO Gardanne plant, (France) that supplies specialty alumina. Bauxaline is an alkaline material (pH 10.6), reddish brown in color, with fine particle size containing iron, aluminum and titanium oxides. The specific surface area of bauxaline was found to be  $23 \text{ m}^2/\text{g}$  (Taneez et al., 2015). Bauxaline has strong tendency for adsorption of metals from wastewater, acid mine drainage, soil and sediments (Doye and Duchesne, 2003; Garau et al., 2007; Taneez et al., 2015; Vaclavikova et al., 2005).

Steel slag (0/10 mm) was provided by Phoenix Services, Inc. Steel slags are metallurgical by-products presenting a complex mixture of  $\text{SiO}_2$ , CaO,  $\text{Fe}_2\text{O}_3$ , FeO,  $\text{Al}_2\text{O}_3$ , MgO, MnO, and  $\text{P}_2\text{O}_5$ . Limestone (CaO) and silica ( $\text{SiO}_2$ ) are primary components of iron and steel slag as shown in Table 1 (Grubb et al., 2011). Being alkaline (pH 10–12), porous in nature with a large surface area and a wide particle distribution, steel slag has potential for removal of Cd, Cu, Pb, Zn, and As metal ions from water and acid mine water (Huiifen et al., 2011; Oh et al., 2012), soil aquifer treatment, soil metal

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