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A life cycle assessment study on the stabilization/solidification treatment processes for contaminated marine sediments

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ABSTRACT

Contaminated marine sediment management strategies involves *in situ* and *ex situ* options for preventing pollutants from re-entering the water column, thus becoming available to benthic organisms and subsequently entering aquatic food chains. These pollution abatement strategies can cause significant secondary environmental impacts which in some cases have been considered to be even higher than the primary ones. This study aims at identifying and quantifying through life cycle assessment (LCA) the environmental impacts of the application of Stabilization/Solidification (S/S) options for the remediation of contaminated marine sediments from the Mar Piccolo in Taranto (Southern Italy). The analysis considers all the stages involved in marine sediments processing (dredging, transport, storage, treatment, safe disposal of the treated sediments) but focuses on several S/S options (4 S/S mixes with cement and 4 mixes with lime). These S/S options were tested at lab scale with different results in immobilizing heavy metals and organic pollutants. The LCA suggests that the *ex-situ* treatment could contribute to improving the current situation and that the marine sediments S/S operation generates a complex environmental profile which is dominated by the treatment phase, which in turn shows that optimization of this stage could lower these impacts.

1. Introduction

Sediment-bound pollutants pose major concerns for human health and the environment, because these contaminants can re-enter the overlying water column and become available to benthic organisms and subsequently enter aquatic food chains. Sediment acts as both carriers and long-term secondary sources of contaminants to aquatic ecosystems.

Sediment management strategies may involve *in situ* and *ex situ* options. In situ remedial alternatives generally involve Monitored Natural Recovery (MNR) (De Gisi et al., 2017a) and in situ containment and treatment (Lofrano et al., 2016). While the MNR is based on the assumption that natural processes can reduce risk over time in a reasonably safe manner, in containment and in situ treatments, contaminated sediments are physically and chemically isolated from aquatic ecosystems or contaminants in sediments and further sequestered and degraded. An example of in situ containment and treatment is In Situ Capping (ISC) (De Gisi et al., 2017b; Lin et al., 2011). Ex situ remedial alternatives typically require several component technologies to dredging or excavation, transport, pre-treatment, treatment, and/or disposal of sediments and treatment residues. Among the most widely applied are Stabilization/Solidification (S/S) (Tang et al., 2015; Wang et al., 2015), Nano-scale Zero Valent Iron (nZVI) treatment (De Gisi et al., 2017c), landfarming (NSW EPA, 2014), composting (Mattei et al., 2017), sediment washing (Stern et al., 2007), thermal desorption (Bortone and Palumbo, 2007), vitrification (Colombo et al., 2009), biological treatment (Matturro et al., 2016) and/or their combination

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(Careghini et al., 2010).

Long-applied, S/S is based on adding chemical compounds to dredged material in order to chemically immobilize contaminants and thus reduce leachability and bioavailability. Therefore, S/S does not remove the contaminants from the dredged material, but they are transformed into a less mobile, and less harmful species (Akcil et al., 2015; Bonomo et al., 2009). The simplest form of treatment involves Portland cement although further materials can be added such as calcium aluminates, fly ashes, bentonite or other clays, phosphates, lime, oil residue and silicate fume (Marques et al., 2011). However, the additive used depends on the type of contaminants, water content and characteristics of the dredged material. In the last years, innovative binders and mixtures, alone or in combination with cement, have been tested (Roviello et al., 2017).

Today, S/S is experiencing renewed importance; the use of treated sediments for other applications (material recovery) is an interesting solution in line with the philosophy of the circular economy (Todaro et al., 2016; Wang et al., 2015). In this regard, Colangelo et al. (2017, 2015) investigated the recycling of several waste such as municipal solid waste incinerator fly ash by means of cold bonding palletisation based on the use of cement, lime and coal fly ash as components of the binding systems. The showed how the obtained lightweight porous aggregates were mostly suitable for recovery in the field of building materials with enhanced sustainability properties. Couvidat et al. (2016) studied the feasibility to use dredged sediments as substitute for sand in non-structural cemented mortars. The obtained results confirmed that the reuse of the coarser fraction of a marine sediment offered an interesting valorisation potential as cemented mortars for non-structural applications. Colangelo and Cioffi (2017) analysed the mechanical properties and durability of mortar containing fine fraction of construction and demolition waste (CDW), that generally are problematic waste materials. They use of superplasticizer combined with selective demolition can improve significantly the mechanical properties of mortars produced with CDW aggregate. Recently, Wang et al. (2018) developed a remediation method for contaminated sediment using S/S with calcium-rich/low-calcium industrial by-products and CO_2 utilization. This study represented an additional example of how S/S processes can be a suitable way to transform contaminated sediment into value-added materials. However, the study of this research highlighted the growing importance of assessing the impacts of these new products on the environment.

Life Cycle Assessment (LCA) is one of the most important methods for evaluating the environmental performance of alternative treatment systems considering their entire life cycle (De Feo and Ferrara, 2017; Colangelo et al., 2018). LCA allows to compare different systems considering the consumption of resources as well as the emission of pollutants that may occur during their life cycle (secondary impacts), which may include the extraction of raw materials, the production and processing of materials, the transport, the phase of use and, finally, the end of life (ISO 14040, 2006; ISO 14044, 2006).

Although LCA has been used previously to evaluate various treatment options for contaminated sites (Morais and Delerue-Matos, 2010), in the case of marine sediments, there are few studies that mention LCA as an environmental performance assessment tool, except the ones presented in Table 1. Most of these studies focus mainly on comparing different options for marine sediments manipulation: in-situ vs. natural remediation (Sparrevik et al., 2011; Choi et al. (2016), in-situ vs. ex-situ placement (Bates et al., 2015), primary vs. secondary vs. tertiary impacts (Hou et al., 2014). The study of Falciglia et al. (2018) compares actual treatment technologies for the removal (destruction) of hydrocarbons from MS by heat. To our current knowledge, information on the assessment by life cycle assessment of impacts associated to the use of ex-situ S/S for the remediation of contaminated sediments is currently limited.

Table 1
LCA studies of marine sediments decontamination operations.

No.	Location/main contaminants	Goal and scope, functional unit (FU)	LCIA method	Results/impacts	Reference
1	Greenland fjord, Norway polluted with polychlorinated dibenzo-p-dioxins and -furans	Comparison of natural remediation and capping, and in-situ treatment with various materials FU: whole inner fjord area (23.4 km^2)	Modified Recipe to account for local toxicity conditions	Secondary impacts due to capping are higher than primary impacts (natural remediation)	Sparrevik et al. (2011)
2	London Olympic Park, London, UK. Sediments contaminated with lubricating range organics (LRO) and polycyclic aromatic hydrocarbons (PAHs)	Comparison of "primary impacts" associated with the state of the site (e.g. site contamination), "secondary impacts" associated with remediation operations, and "tertiary impacts" associated with the effects of the post-rehabilitation fate of the site FU: 2500 m of waterways for 100 years; 30,000 m^3 of sediment when evaluating the different treatment methods	IO-based hybrid LCA coupled with social and economic data. default ReCiPe endpoint method, hierarchist version for environmental assessment	adverse secondary environmental impacts can exceed environmental benefit resulting from contamination removal, but the consequential benefit (i.e. tertiary impact) resulting from site use change can far exceed the secondary environmental impact	Hou et al. (2014)
3.	Long Island Sound, NewYork, USA Dredged material is considered uncontaminated	Comparison of three types of placement alternatives (open water, containment island, and upland) for dredged material at three different transport distances. FU: 100,000 cubic yards (cy) of uncontaminated sediment	IMPACT 2002+ Recipe	Transport-related impacts (climate change, fossil fuel depletion, etc.)	Bates et al. (2015)
4	Hunters Point Shipyard, San Francisco (USA) polluted with polychlorinated biphenyls (PCBs)	Comparison of dredge-and-fill; capping, and in-situ activated carbon. FU: for 1000 m^2 of remediated area.	Eco-Indicator 95	Comparable impacts for dredge-and-fill and in-situ AC amendment using C-VAC, and smaller for capping.	Choi et al. (2016)
5	Augusta Bay (Sicily, Southern Italy), marine sediment contaminated with hydrocarbons	Evaluation decontamination by citric acid enhanced-microwave heating and electrokinetic processes. Dredging and transport not included FU: 1 ton of sediments	Impact 2002+	MW technology is 75.74% lower the electrokinetic decontamination Electricity consumption related impacts	Falciglia et al. (2018)