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

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Original Citation:

A life cycle assessment study on the stabilization/solidification treatment processes for contaminated marine sediments / George, Barjoveanu; De Gisi, Sabino; Casale, Rossella; Todaro, Francesco; Notarnicola, Michele; Carmen, Teodosiu. - In: JOURNAL OF CLEANER PRODUCTION. - ISSN 0959-6526. - STAMPA. - 201:(2018), pp. 391-402. [10.1016/j.jclepro.2018.08.053]

Availability:

This version is available at <http://hdl.handle.net/11589/138116> since: 2021-03-06

Published version

DOI:10.1016/j.jclepro.2018.08.053

Publisher:

Terms of use:

(Article begins on next page)

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

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24
25 12 **ABSTRACT**

26
27 13 Contaminated marine sediment management strategies involves *in situ* and *ex situ* options
28
29 14 for preventing pollutants from re-entering the water column, thus becoming available to
30
31 15 benthic organisms and subsequently entering aquatic food chains. These pollution
32
33 16 abatement strategies can cause significant secondary environmental impacts which in some
34
35 17 cases have been considered to be even higher than the primary ones. This study aims at
36
37 18 identifying and quantifying through life cycle assessment (LCA) the environmental
38
39 19 impacts of the application of Stabilization/Solidification (S/S) options for the remediation
40
41 20 of contaminated marine sediments from the Mar Piccolo in Taranto (Southern Italy). The
42
43 21 analysis considers all the stages involved in marine sediments processing (dredging,
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45 22 transport, storage, treatment, safe disposal of the treated sediments) but focuses on several
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47 23 S/S options (4 S/S mixes with cement and 4 mixes with lime). These S/S options were
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49 24 tested at lab scale with different results in immobilizing heavy metals and organic
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25 pollutants. The LCA suggests that the ex-situ treatment could contribute to improving the
26 current situation and that the marine sediments S/S operation generates a complex
27 environmental profile which is dominated by the treatment phase, which in turn shows that
28 optimization of this stage could lower these impacts.

29
30 **Keywords:** Ex-situ treatment; LCA; Leaching test; Marine sediments contamination;
31 Organic clay; Portland cement

32 33 1. INTRODUCTION

34
35 Sediment-bound pollutants pose major concerns for human health and the environment,
36 because these contaminants can re-enter the overlying water column and become available
37 to benthic organisms and subsequently enter aquatic food chains. Sediment acts as both
38 carriers and long-term secondary sources of contaminants to aquatic ecosystems.

39 Sediment management strategies may involve *in situ* and *ex situ* options. In situ remedial
40 alternatives generally involve Monitored Natural Recovery (MNR) (De Gisi *et al.*, 2017a)
41 and in situ containment and treatment (Lofrano *et al.*, 2016). While the MNR is based on
42 the assumption that natural processes can reduce risk over time in a reasonably safe
43 manner, in containment and in situ treatments, contaminated sediments are physically and
44 chemically isolated from aquatic ecosystems or contaminants in sediments and further
45 sequestered and degraded. An example of in situ containment and treatment is In Situ
46 Capping (ISC) (De Gisi *et al.*, 2017b; Lin *et al.*, 2011). Ex situ remedial alternatives
47 typically require several component technologies to dredging or excavation, transport, pre-
48 treatment, treatment, and/or disposal of sediments and treatment residues. Among the most
49 widely applied are Stabilization/Solidification (S/S) (Tang *et al.*, 2015; Wang *et al.*, 2015),
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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 (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

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75 sediment offered an interesting valorisation potential as cemented mortars for non-
76 structural applications. Colangelo and Cioffi (2017) analysed the mechanical properties
77 and durability of mortar containing fine fraction of construction and demolition waste
78 (CDW), that generally are problematic waste materials. They use of superplasticizer
79 combined with selective demolition can improve significantly the mechanical properties of
80 mortars produced with CDW aggregate. Recently, Wang *et al.* (2018) developed a
81 remediation method for contaminated sediment using S/S with calcium-rich/low-calcium
82 industrial by-products and CO₂ utilization. This study represented an additional example of
83 how S/S processes can be a suitable way to transform contaminated sediment into value-
84 added materials. However, the study of this research highlighted the growing importance
85 of assessing the impacts of these new products on the environment.

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

93

94 **Table 1. Life cycle assessments of contaminated marine sediments treatment options.**

95

96 Although LCA has been used previously to evaluate various treatment options for
97 contaminated sites (Morais and Delerue-Matos, 2010), in the case of marine sediments,
98 there are few studies that mention LCA as an environmental performance assessment tool,
99 except the ones presented in Table 1. Most of these studies focus mainly on comparing

100 different options for marine sediments manipulation: in-situ vs. natural remediation
101 (Sparrevik *et al.*, 2011; Choi *et al.* (2016), in-situ vs. ex-situ placement (Bates *et al.*, 2015),
102 primary vs. secondary vs. tertiary impacts (Hou *et al.*, 2014). The study of Falciglia *et al.*
103 (2018) compares actual treatment technologies for the removal (destruction) of
104 hydrocarbons from MS by heat. To our current knowledge, information on the assessment
105 by life cycle assessment of impacts associated to the use of ex-situ S/S for the remediation
106 of contaminated sediments is currently limited.

107 In this context, the article presents the implementation of a complex LCA study aimed at
108 identifying, quantifying and analysing the primary, secondary and tertiary environmental
109 impacts of the remediation options for marine sediments coming from Mar Piccolo in
110 Taranto (Southern Italy). This area is known for its economic and tourism activities as well
111 as for sea-food production, but also for being one of the most polluted in Europe. This
112 assessment is intended to evaluate various MS stabilization/solidification options in a
113 wider context that considers the current local situation (primary impacts); the manipulation
114 of sediments (including dredging, transport and on-shore operations); the specific
115 performance of solidification/stabilization mixes (secondary impacts) and finally the
116 tertiary impacts due to final MS disposal. The sensitivity analysis (SA) considered the
117 measured variability of key flows (i.e. pollutant releases and material consumption,
118 measured as standard deviations) and default variability of background processes in the
119 inventory. Alongside the LCA study, the paper discusses the technical performance of 8
120 S/S mixes that use various proportions of Portland cement, lime, activated carbon and
121 organic clays.

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123 **2. MATERIALS AND METHODS**

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125 *2.1 Background information*

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2 126 Taranto is a coastal city in Southern Italy, an important commercial port as well as the
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5 127 main Italian naval base. Taranto faces the Ionian Sea and is known as the “city of two seas”
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7 128 because it is extended around the Big Sea and the vast reservoir of the Little Sea,
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10 129 composed of the two internal basins (Fig. 1).

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14 131 **Figure 1. The “Mar Piccolo of Taranto” study area (Southern Italy): sampling area,**
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17 132 **main phases of the intervention (dredging, intermediate storage and treatment) and**
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19 133 **the S/S pilot treatment plant located at the Taranto Bellavista municipal wastewater**
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21 134 **treatment plant.**

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24 136 The relatively shallow waters in the Gulf of Taranto yield large numbers of mussels
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27 137 *Mytilus Galloprovincialis* so, Taranto seas are a noteworthy economic resource, being the
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30 138 site of intensive mussel farming. In addition to the commercial aspect, this activity has a
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33 139 close connection to the traditions of the city as its history that dates back to the sixteenth
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36 140 century. In fact, the mussel breeder is the oldest job of the tarantine tradition. This industry
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39 141 has grown from the idea of an enterprising local to become a big export earner. Until 2007,
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41 142 the annual output amounted to 30,000 tonnes of mussels. Only a part of the locally
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43 143 harvested seafood was used for home consumption, while most was exported to European
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46 144 Economic Community countries (Cardellicchio *et al.*, 2007a).

47
48 145 The trade of this typical product, renamed “black gold of Taranto”, has been repeatedly hit
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51 146 by restrictions because of the strong contamination. The picking and handling of mussels
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53 147 grown in the first basin (in Italian, *Primo Seno*), has been forbidden for three years (Decree
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56 148 of the Health Authority n. 1989 of the 22/07/2011) and then its collection and destruction
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58 149 has been ordered (Decree of the Health Authority n. 1765 of the 11/06/2012). Now mussels

150 are still farmed in Taranto Sea, but most of them have been moved to the Mar Grande and
151 all the others can only be kept in the first basin water for the initial phase of ripening, then
152 they need to be moved in the Mar Grande too for the last maturation, in a different
153 temperature and water condition. Although there were conducted some studies to evaluate
154 the local impacts of intensive mussel's production even through LCA (Iribarren *et al.*,
155 2010), the objective of this paper is driven on investigating how the causes of declining
156 mussel's quality may be addressed through marine sediments stabilization and
157 solidification.

158 The city is one of the areas declared as "at high risk of environmental crisis" by the
159 national government (Italian Law n. 349. 1986) because it represents one of the most
160 complex industrial sites in Europe, located near urban areas of high population density. All
161 the industrial activities are responsible for the high environmental contamination, mainly
162 due to heavy metals and organic pollutants. This explains why Taranto has been recently
163 included into the list of polluted Sites of National Interest (SIN) by the Italian Government
164 (Italian Law n. 426, 1998), for which the environmental remediation has been identified as
165 a national priority (Ministerial Decree n. 468, 2002) (Vitone *et al.*, 2016).

166 In the last ten years, the seabed of the basins has been investigated through a widespread
167 survey. The submarine sediments in the Mar Piccolo contain high concentrations of heavy
168 metals (i.e., Hg, Pb, Cd, Cu and Zn) and organic pollutants (PCBs, PAHs and dioxins)
169 (Bellucci *et al.*, 2016; Kralj *et al.*, 2016; Matturro *et al.*, 2016, Cardellicchio *et al.*, 2007b).

170

171 2.2 Experimentation plan

172 The investigation presented in this study has involved two main phases, namely S/S testing
173 and LCA evaluation as presented in Figure 2, which shows the schematization of the main
174 lab-scale studies, LCA phases, the MS treatment life cycle steps (processes that were

175 considered in the life cycle inventory) and the specific impact categories of Recipe 2008
176 mid-point method which was used for the life cycle impact assessment.

177

178 **Figure 2. Experimental framework for marine sediments treatment and LCA**
179 **approach.**

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181 *2.2.1 S/S testing*

182 Sediments, coming from one of most contaminated areas of Mar Piccolo, were taken up to
183 depths of about 1.5 m from the seafloor, that is about the depth of interest in view of any
184 mitigation solution. These were passed through a 2 cm sieve, homogenized by mixing and
185 stored at 4 °C until use. The standard protocols of ISPRA (the Italian Institute for the
186 Environmental Protection and Research) was used for determining grain-size, moisture
187 content and organic matter of sediments (ICRAM-APAT, 2007). The concentrations of
188 metals were obtained by ICP-OES (Inductively Coupled Plasma-Optical Emission
189 Spectrometry) in accordance to EPA method 200.8 (EPA, 1994). For the determination of
190 the total PAHs and PCBs concentrations as well as each compound or homologue group, a
191 Gas Chromatograph - Mass Spectrometer (GC-MS) and EPA method 8275A was used. In
192 the present case, the sediment samples were only contaminated by inorganic pollutants,
193 shown in Table 2.

194

195 **Table 2. Physical-chemical properties of the sediments samples used for the tests.**

196

197 The mixtures were prepared by using different contents (by dry soil weight) of several
198 additives, namely CEM I 42.5 R Portland cement (C), lime (L), activated carbon (AC) and
199 organoclay (OC) (Table 3).

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2 **201 Table 3. Mixture design for S/S testing.**

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7 **203** All the materials were initially mixed for 5 min with a standard mixer and, then, a steel
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9 **204** trowel to ensure a homogeneous paste was used. In the casting phase, the prepared mixture
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11 **205** was introduced into different silicone molds with hemispherical shape. The samples, in the
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13 **206** curing phase, were kept at 20 ± 5 °C and 80% moisture.

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17 **207** The leaching tests were carried out according to the EN standard 12457-2 (EN 12457-2,

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19 **208** 2002). For several samples, a 40g portion was sampled and transferred to a polyethylene

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21 **209** bottle. Distiller water was added with a solid-liquid ratio of 1:10 by weight and the bottles

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24 **210** was keep in rotation at 12 rpm for 24 hours using Rotax 6.8 (Velp Scientifica). To end of

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26 **211** the 24 hours, a short retention time was given to the extraction vessels for the settlement of

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28 **212** suspended coarse solids; then, the leachate was filtered for the removal of suspended

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30 **213** solids. The soluble concentrations of heavy metals of interest (As, Co, Cr, Ni, V and Zn)

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32 **214** were analysed by using ICP-OES.

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34 **215**

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36 **216** *2.2.2 LCA evaluation*

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39 **217** The objective of the LCA study was to investigate the environmental impacts associated to

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41 **218** the current situation in Mar Piccolo (primary impacts), the impacts associated to the S/S

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43 **219** options (secondary impacts), and the potential impacts that appear during the post-

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45 **220** treatment phases (tertiary impacts)

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48 **221** The functional unit of the LCA study was chosen to be one square meter of sea bed in Mar

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50 **222** Piccolo from which the top layer of 50 cm was considered in the next phases of the LCA

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52 **223** analysis. The characteristics of this sediment are presented in Table 2. This surface-based

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54 **224** functional unit definition is motivated by the need to improve the local sea-bed quality.

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1 225 The system under study was organized as pre-treatment operations which included the
2 226 dredging process, dockyard operations (unloading and transfer to a storage site); marine
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4 227 sediment treatment (8 options of S/S with various mixes stabilizers, according to Table 3)
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7 228 and post-treatment operations which included final placement in a specially designed
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9 229 storage facility.

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11 230 Dredging was modelled considering a hydraulic dredger, which is very efficient when
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14 231 working with fine materials, because they can easily be held in suspension (Blažauskas *et*
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16 232 *al.*, 2006), so it suits to this case study as the characterization of the seabed shows a high
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18 233 percentage of silt and clay. This category of dredge has been chosen as it is suitable for
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21 234 navigational dredging and environmental dredging, even if they present a quite high water
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24 235 content of the removed material (Blažauskas *et al.*, 2006).

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26 236 The dredge characteristics that were included in the life cycle inventory have considered
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29 237 the dredge transport power (1950 kW), the dredge jet pump power (800 kW) and a
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31 238 maximum production rate of 581.53 m³/h. Also, modelling of this process has considered
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34 239 that the dredger aspires a mix of water and sediment in a 5:1 proportion. The Jet pump
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36 240 collects the top layer of sediments (50 cm) and then the two phases are separated on a
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39 241 barge while the excess of water is expelled back to the sea.

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41 242 The volume of sediments to be dredged has been estimated to about 900,000 m³, covering
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44 243 a surface of about 180 ha.

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46 244 Then, the life cycle inventory included a transfer process in which the dredged marine
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49 245 sediments are moved from the Port of Taranto by lorry to a Pilot Technology Platform for
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51 246 treatment and temporary storage. This platform is located at the municipal wastewater
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53 247 treatment plant (WWTP) of Taranto Bellavista, less than 4 km away from the port (Fig. 1).

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56 248 For the LCA modelling, the S/S treatment was imagined as a mixing process that it would
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58 249 take place in the Pilot Technology Platform. Because of the high moisture content, the
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1 250 sediment needs to lose some of the excess water. This process was modelled by using the
2 251 sludge drying beds in the WWTP of Taranto Bellavista. Once the moisture rate reaches a
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4 252 suitable value, the sediment is moved to the hopper and the treating begins. After mixing, a
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7 253 granulation step is provided, then the granular material needs a maturation phase of 28
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9 254 days, then it can be reused or deposited in landfill.

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12 255 The post-treatment phase was modelled in LCA as a landfill of residual materials. Detailed
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14 256 information of the life cycle inventory organization is presented in the supplementary
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17 257 material (Table S1 – inventory data).

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20 21 259 **3. RESULTS AND DISCUSSION**

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25 26 261 *3.1 S/S testing*

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29 262 This study proposed a remediation approach to treat and recycle the contaminated
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31 263 sediment by means of stabilization/solidification enhanced by the addition of absorbent
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34 264 materials. Stabilization/solidification of contaminated sediments has proved to be an
35
36 265 appealing technology for metal immobilization, such that the treated sediments can be
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39 266 recycled (Couvidat *et al.*, 2016; Pinto *et al.*, 2011).

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41 267 For the beneficial reuse of contaminated marine sediments, the leaching of each metal has
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44 268 to be lower than limits imposed by legislation. In Italy, the chemical parameters must be
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46 269 under the threshold levels defined by the Italian Ministerial Decree 5/2/1998. The leaching
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49 270 tests results of S/S treated marine sediments after 28 days are given in Table 4.

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52 53 272 **Table 4. Lab-scale S/S performance in terms of metals removal and leaching test.**

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58 274 In general, the addition of binders and reagents to the contaminated marine sediments
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275 resulting shows positive effects on decreasing the mobility of heavy metals. The Vanadium
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2 276 (V) from mixtures with cement (Mix 1, Mix 2, Mix 3 and Mix 4) and the Copper (Cu)
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4 277 from Mix 5 and Mix 7 are released with concentrations higher than the legal limits.
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7 278 However, with greater curing times (i.e., 56 days) the leaching of metals was well
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9 279 controlled, especially it was less than 0.02 mg/L.

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12 280 Usage of 10% lime in combination with 5% AC (Mix 2) or with 2.5% AC and 2.5% OC
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14 281 (Mix 8) is effective such that all metal concentrations meet the regulatory standards.

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17 282 The main results shown in this study indicate that, despite the total concentrations of heavy
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19 283 metals in the studied marine sediment, the release of contaminants after contact with
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21 284 deionized water is very limited. This is due to the low metals solubility and to the stability
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24 285 of their solid phases under slightly basic conditions (Chatain *et al.*, 2013). In particular,
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26 286 mobility of the metals appears to be governed by pH. However, the adding of cement
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29 287 appears to increase the leaching of vanadium; whereas the adding of lime appears to
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31 288 increase the leaching of copper. A possible effect of the contaminants (i.e., organic matter
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34 289 and heavy metals) that interfered with the chemistry of binder's hydration, compromising
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36 290 the effectiveness of metal stabilization and development of hardening (Wang *et al.*, 2015).

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40 41 292 *3.2 Life cycle impact assessment*

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43 293 The life cycle impact assessment was performed using the Recipe 2008 mid-point method
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46 294 with the impact categories and the normalization values presented in Table 5. This method
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49 295 was selected based on a preliminary LCIA method screening considering aspects like
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51 296 impacts relevance and data representability. The ReCiPe 2008 method covers a multitude
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53 297 of environmental aspects and it has a good inventory data coverage as it provides
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56 298 characterization factors (which are particularized for the sea compartment) for more
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58 299 pollutant species than the other considered methods.

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Table 5. Impacts categories defined in the Recipe 2008 midpoint method.

3.2.1 Reference case and environmental benefices

Since the goal of the LCA study was to evaluate the performance of the eight options for marine sediment stabilization, it was important to determine the impacts of a “no-action” scenario and to use it as a reference case against which all other actions would be compared. This scenario characterizes the “primary impacts” of the contaminated marine sediments in Mar Piccolo, as it was important to have this reference case impact done with the same evaluation tool.

In a similar way to the previous studies, (Sparrevik *et al.*, 2011, Hou *et al.*, 2014) which have used multi-compartment fate models, in the “no-action” scenario considered in this study, pollutants from the marine sediments cause impacts to the local marine eco-system via a resuspension process in which pollutants are released from the solid phase of the sediments to the liquid phase of the sea water. This resuspension process was modelled in LCA as a discharge into the seawater of a virtual wastewater containing pollutant concentrations corresponding to the complex transport and transformation phenomena of the resuspension process. Although there were some laboratory data available for the marine sediment leaching behaviour, these were not used, as they were performed in standard lab conditions (with deionized water at neutral pH), which are completely different from the real sea-bed situation. In this case, the released pollutant concentrations were determined with the help of a model (Martín-Torre *et al.*, 2015) which considers the complex processes (redox reactions of the metal species found in the marine sediments, pollutant release and sorption processes), as well as the local conditions (pH, ionic strength).

325 In Fig. 3a a comparison between the “no-action” scenario and the marine sediments
326 removal is presented and it shows that the modelled resuspension of the marine sediments
327 would cause an impact in the marine toxicity category, as expected.

Fig. 3. (a) Environmental comparison of three marine sediments management options; (b) Comparison of potential environmental toxicity-related impacts of the 8 S/S options. Symbols in the figure: HT = Human toxicity; TTOX = Terrestrial ecotoxicity; FTOX = Freshwater ecotoxicity; MTOX = Marine ecotoxicity.

334 By removing the contaminated sediments from the sea-bed, the impact in this
335 environmental compartment was greatly diminished (from 0.235 to 0.0007 impact points)
336 which shows that the ex-situ treatment of sediments is a viable option for solving the Mar
337 Piccolo pollution problem. At the same time, landfilling the marine sediments without
338 treatment represents a pollution transfer from the sea to the land, as impacts in the human
339 toxicity, terrestrial toxicity and freshwater toxicity increase. This showed that the
340 stabilization of the pollutants is required for a safe landfilling, as demonstrated by the great
341 decrease of the impact values in these categories after a S/S with Portland cement.

342 In figure 3b a performance comparison of the eight S/S options based on the stabilization
343 potentials determined by laboratory testing (Fig. 4a) highlights how the highest impacts
344 were caused in the terrestrial toxicity (TTOX) category, with much smaller impacts in the
345 other toxicity compartments. In general, the quick lime S/S mixes generated considerable
346 smaller impacts than the cement options in the toxicity-related categories, but it has to be
347 noted that these impacts consider only to the leaching potential of the treated marine
348 sediments.

350 3.2.2 Evaluation of S/S options

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2 351 Fig. 4a presents the full environmental impact profile of the marine sediments S/S options
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4 352 considering the specific materials inputs, as well as the S/S potential. The results showed
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7 353 that the impacts are higher with increasing quantity of additives in the S/S mix. Thus, the
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10 354 smallest impacts were caused by Mix 5 in most impact categories (quicklime 10%, 0% AC,
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12 355 0% OC), followed by Mix 1 (cement 10%, 0% AC, 0% OC), while the highest were caused
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14 356 by Mixes 2 and 6 (due to the addition of AC).
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19 358 **Fig. 4. Potential additional impacts due to sediment treatment options (a)**
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21 **(characterization) and (b) (normalization); Symbols in the figure are those designated**
22 359 **in Table 5.**
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29 362 An impact analysis which considers the specific impact categories was possible after the
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31 363 normalization step (Fig. 4b). The highest impacts appeared in the natural land
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34 364 transformation (NLT), followed by impacts in the toxicity related categories (Mtox, Ftox,
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36 365 Htox) and freshwater eutrophication. The main contributors in these categories were
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39 366 related to the use of AC and OC.
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44 368 **Figure 5. Comparison of secondary environmental impacts (treatment costs)/tertiary**
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46 369 **environmental impacts (benefices).**
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51 371 As presented before, the treatment of marine sediments with various mixes was successful
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53 372 in stabilizing/immobilizing various metal pollutants, but it also introduced some secondary
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56 373 environmental impacts due to the use of stabilizers (cement, quicklime, activated carbon
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58 374 and organic clay). From this point of view, it was important to identify and understand the
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375 trade-offs that need to be made to find an optimal solution. In Fig. 5, a comparison
376 between the environmental benefices (orange bars, expressed as % of the maximal impact
377 per treatment mix) and the secondary environmental impacts due to treatment (blue bars,
378 expressed as % of the maximal impacts) is presented. This comparison quantified the
379 environmental benefices as the potential impacts due to pollutant leaching in accordance
380 with the treatment options (Table 3), so the best treatment options referred to the ones with
381 the smallest value (quicklime mixes). The secondary environmental impacts due to marine
382 sediment treatment (blue bars) were quantified considering the consumption of treatment
383 agents and, again, the most environmentally advantageous were the ones with the smallest
384 values. In consequence, from Fig. 4b one may notice that the quicklime treatment options
385 have considerably smaller potential direct impacts (via pollutant leaching), compared to
386 the cement treatment mixes. At the same time, the options involving the addition of
387 activated carbon induced the greatest additional impacts, followed by the mixes with both
388 activated carbon, then organoclay and finally, the lowest environmental impacts appeared
389 when using only cement or quicklime, respectively.

390

391 *3.2.3 Complete environmental profiles*

392 The environmental analysis of the treatment options was refined to include data regarding
393 all the steps; these environmental profiles were included in Figs. 6a and 6b, respectively.

394

395 **Figure 6. Environmental profile of marine sediment treatment with (a) Mix 2 (cement
396 10%, GAC 5%) and (b) Mix 5 (quicklime 10%); Symbols in the figure are those
397 presented in Table 5.**

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399 These results presented the fact that the determining treatment step in the total

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400 environmental impact balance was the ex-situ treatment step, because it shows a great
401 variability of impact values with changing treatment options. For example, in Fig. 5a the
402 environmental profile of marine sediment treatment option 2, which is the least favourable
403 option from an environmental point of view (mix with 10% cement, and 5% activated
404 carbon), generated much higher impacts compared to the other treatment steps, as well as
405 compared to the most favourable option (approximately one order of magnitude), which is
406 presented in Fig. 5b. These higher impacts were associated in all the impact categories with
407 the use of the activated carbon. This showed that the stabilization material has a high
408 importance in this environmental balance (in most of the impact categories, as it can be
409 seen in the snippet of Fig. 5a) and its choice and dosage can be a good option for the
410 environmental optimization of marine sediments treatment options. This is supported by
411 the fact that in the life cycle inventory, the activated carbon was modelled as being
412 produced from a carbon-based source, thus giving the high impacts, whereas if it had been
413 modelled from bio-based source the impacts would have been lower, as suggested in other
414 studies (Sparrevik *et al.*, 2011).

415 The environmental impact profile of the most environmentally friendly treatment option
416 (Mix 5), presented in Fig. 5a showed a more balanced distribution of impact contributors.
417 Higher impacts appeared in the land use categories (NLT and ULO) due to landfilling and
418 in the environmental pollution categories (FE, HT, Ftox, Mtox) mainly due to the use of
419 solidification agents.

420 It was important to note that the pre-treatment operations which include the marine
421 sediments dredging, transfers and storage account for small portions in the overall impact
422 budgets.

423

424 3.2.4 Sensitivity analysis

425 The many uncertainty sources that arise in LCA studies are usually grouped into three
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2 426 categories (European Commission 2010): stochastic uncertainty, choice uncertainty.
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4 427 Stochastic uncertainty refers to inventory and assessment data uncertainty and is usually
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7 428 estimated with the help of probability distributions that statistically describe how a variable
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10 429 varies around a value (i.e. mean and standard deviation). Choice uncertainty refers to
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12 430 discrete values which usually are modelled in LCA as independent scenarios, while the
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14 431 lack of knowledge uncertainty reflects omission of data or incorrect assumptions (Sabia *et*
15
16
17 432 *al.*, 2016).
18
19 433 Because in this study we consider only some stochastic uncertainties which refer to
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21 434 inventory data variability, the uncertainty analysis is restrained to a sensitivity analysis
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24 435 which considers the measured variability of key flows (i.e. pollutant releases and material
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26 436 consumption, measured as standard deviations) and default variability of background
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29 437 processes in the inventory (default standard deviations and probability distributions of
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31 438 flows in the Eco-Invent data base). While other sensitivity issues may be important, like
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34 439 impact assessment factors uncertainty, these were not included in our sensitivity analysis as
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36 440 all the scenarios were compared against the same reference (characterization factors).
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39 441 The SA was performed to investigate how different contributor's variation affect the
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41 442 impact results by means of Monte Carlo simulations (in 10,000 points).
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44 443 With respect to the input data, a preliminary sensitivity analysis was carried out in order to
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46 444 evaluate how different data quality aspects would impact the LCIA results. In particular,
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48 445 the impacts in the marine toxicity category (MTOX) were investigated for the reference
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51 446 case as a function of pollutant release data variability. This was investigated by developing
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53 447 3 scenarios: 2 data scattering sets, (measured as 2 sets of standard deviation of the mean)
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56 448 and 1 scenario for the type of distribution (normal vs. uniform distributions). The
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58 449 configuration of the SA analysis and results are presented in Table 6.
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2 451 **Table 6. Sensitivity analysis for Marine ecotoxicity, reference case (sediment**
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5 452 **resuspension).**

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10 454 The first two input data sets refer to two independent sampling locations for which time
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12 455 series of determinations were performed (so they would fit a normal distribution), while
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15 456 the last one was obtained from samples collected all over the Mar Piccolo, and which were
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17 457 better described with a uniform distribution, due to their more random character. The
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20 458 output of Monte Carlo analysis show that the first two cases fit very well a normal
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22 459 distribution (indicated by the low chi squared values), and that the variability of results is
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25 460 very low (coefficient of variation of 1.31 %). For the third case, and the best fit was a beta
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27 461 probability distribution which describes better the much greater variability of the results
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30 462 (coefficient of variation of 27.78%). In terms of contributions, data in Table 6 shows that in
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32 463 all 3 cases the major source of variance is V, followed by Cu and Ni, while the other
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34 464 species have negligible impact on the variability of the total impact.

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36
37 465 Individual SA were performed to investigate how different sources of variability influence
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39 466 the complex profiles that consider multiple environmental categories. In figure 7 a
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42 467 comparison of uncertainties between the option with the highest (mix 2) and smallest (mix
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44 468 5) overall impacts is represented as percentage of variation (with a 95% confidence
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47 469 interval around the most probable value). Data shows in general low variability, the highest
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49 470 deviation is 23% for natural land transformation for mix 2, and 20% for freshwater
50
51 471 eutrophication for mix 5, which indicates a high confidence in the LCA model and its
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54 472 results. The deviation profiles in Figure 7 are backed up by data regarding the contribution
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56 473 sources, for each impact category, which are presented in the Supplementary material (file
57
58
59 474 S2) and may be explained by the relative low number of variables that generate a high

475 sensitivity in the majority of impact categories.

476

477 **Figure 7. Uncertainty comparison between mix 2 and mix 5 (expressed as a variation**
478 **coefficient calculated for a 95% confidence interval around the most probable value).**

479

480 This SA was then used to compare the impact values obtained for the MS treatment
481 options, especially in the categories where these impacts were closer. Data in Table 7
482 presents the results of Monte Carlo analysis, the goodness of fit parameters, and the
483 probability for each impact category, showing that impacts of mix 2 are higher than those
484 of mix 5 (for highest overall impacts and lowest ones, respectively). While for most
485 categories, mix 2 has 100% chances to have higher impacts than mix 5, for ozone depletion
486 this probability is only 63% and for natural land transformation is 86%, which indicates
487 that interpretation of greater this data should be done attentively.

488

489 **Table 7. Monte Carlo analysis comparison across multiple impact categories for the**
490 **total impacts of mix 2 and mix 5.**

491

492 **4. CONCLUSIONS**

493

494 This study approaches the environmental analysis of ex-situ marine sediment treatment
495 options by stabilization-solidification with the use of the life cycle assessment
496 methodology. The study is applied on sea region in the Mar Piccolo of Taranto in Italy,
497 where the historic pollution with heavy metals and persistent organic pollutants cause
498 significant impacts to the local sea-farming economies, as these pollutants that originate in
499 the top-layer of the marine sediments tend to bio-accumulate in the tissues of some

500 molluscs that are commercially farmed in the area.

501 This research analyses from an environmental standpoint the potential impacts caused by
502 all the marine sediment treatment steps (dredging, transport, transfer, storage, actual
503 treatment and final disposal) and discusses the types and values environmental impacts that
504 are associated to these processes.

505 By means of LCA it was possible to demonstrate that the ex-situ treatment of these marine
506 sediments can lead to an improvement of the local situation. With respect to the evaluation
507 of the treatment options it has to be noted that, based on their performances, these can
508 diminish greatly the potential risks associated to metals leaching, but they induce
509 additional impacts in various other categories that are associated to the use of stabilizers
510 and solidification agents.

511

512 **ACKNOWLEDGEMENTS**

513

514 The study was carried out under the ERASMUS+ Agreement between the Polytechnic
515 University of Bari and the “Gheorghe Asachi” Technical University of Iasi.

516

517 **DECLARATION OF INTEREST**

518

519 None

520

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Figure 1. The “Mar Piccolo of Taranto” study area (Southern Italy): sampling area, main phases of the intervention (dredging, intermediate storage and treatment) and the S/S pilot treatment plant located at the Taranto Bellavista municipal wastewater treatment plant.

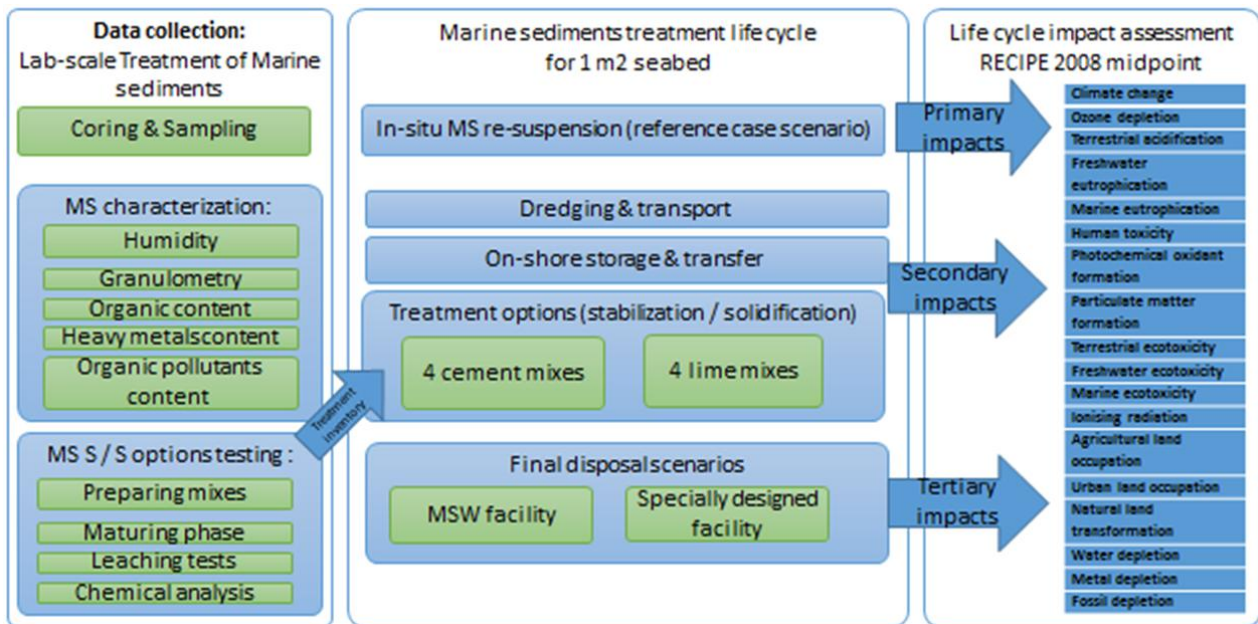
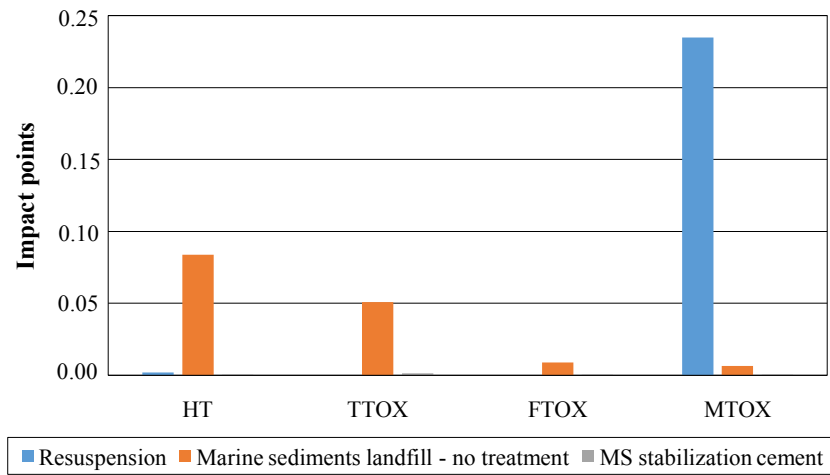
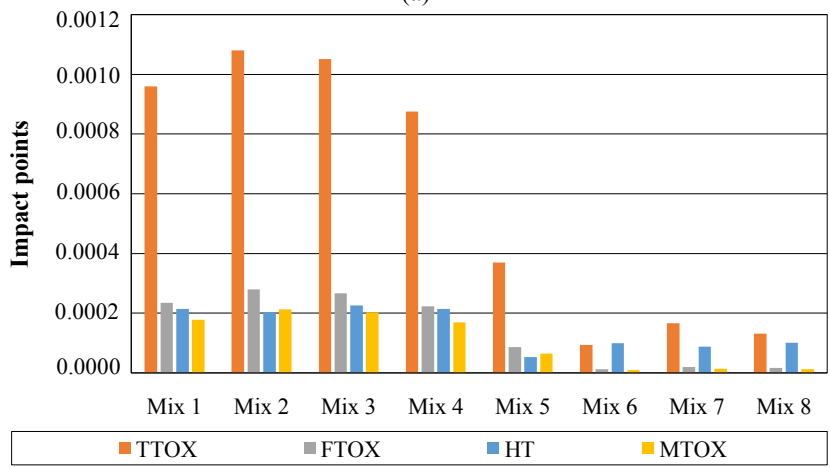


Figure 2. LCA framework for MS treatment and LCA approach.

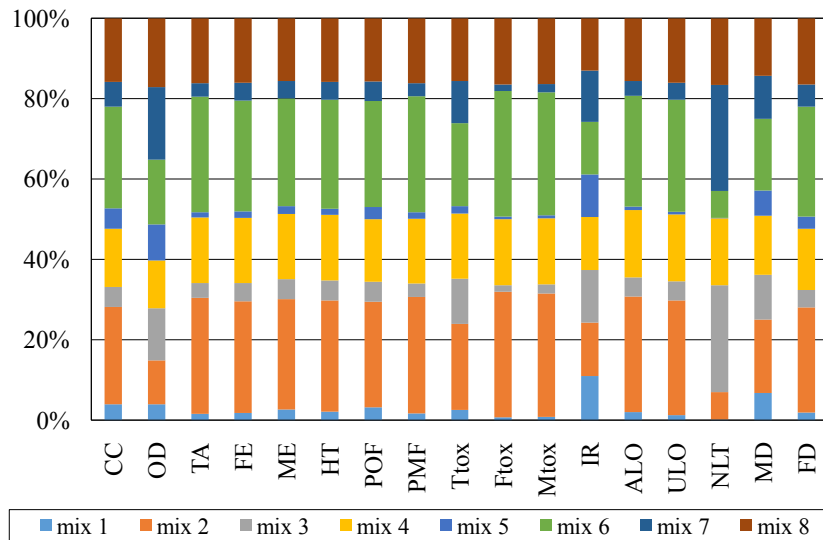


(a)

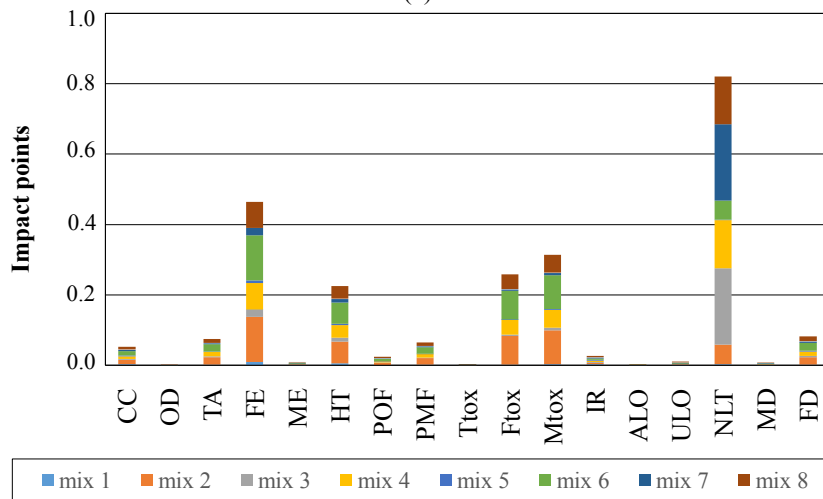


(b)

Figure 3. (a) Environmental comparison of three marine sediments management options; (b) Comparison of potential environmental impacts of various S/S options. Symbols in the figure: HT = Human toxicity; TTOX = Terrestrial ecotoxicity; FTOX = Freshwater ecotoxicity; MTOX = Marine ecotoxicity.



(a)



(b)

Figure 4. Potential additional impacts due to sediment treatment options (a) (characterization) and (b) (normalization); Symbols in the figure, as presented in Table 4.

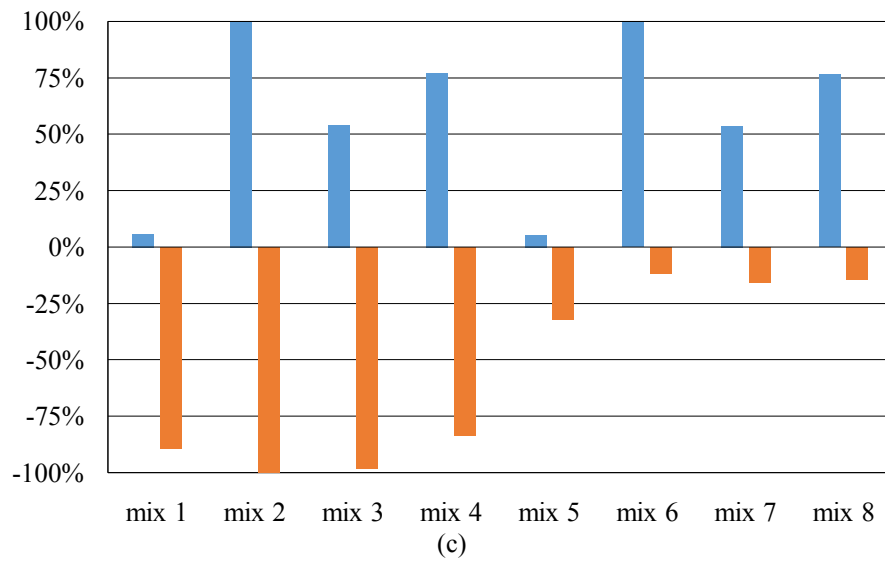
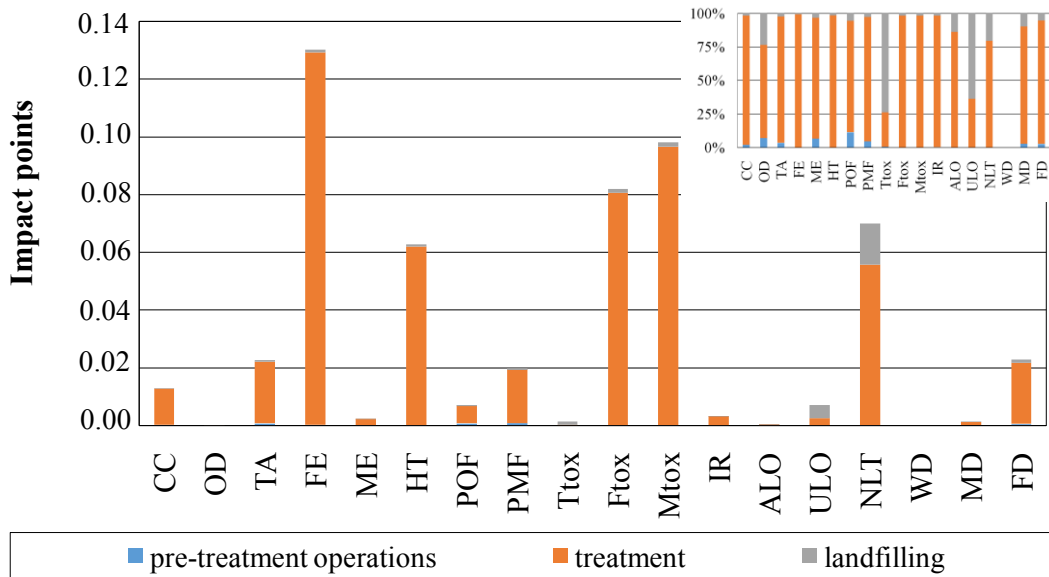
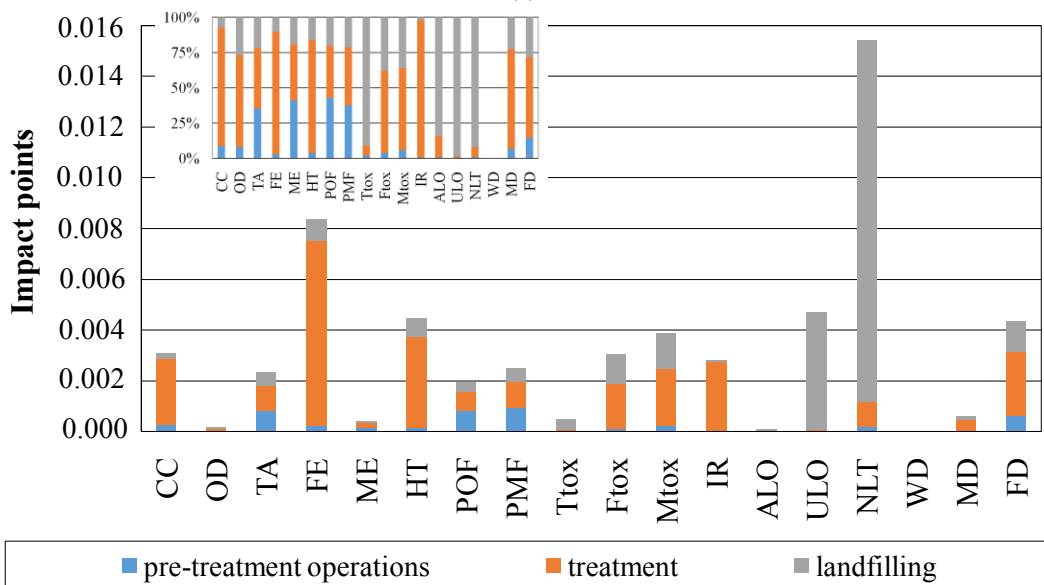


Figure 5. Comparison of secondary environmental impacts (treatment costs)/tertiary environmental impacts (benefices).



(a)



(b)

Figure 6. Environmental profile of marine sediment treatment with (a) Mix 2 (cement 10%, GAC 5%) and (b) Mix 5 (quicklime 10%). Symbols in the figure as those presented in Table 4.

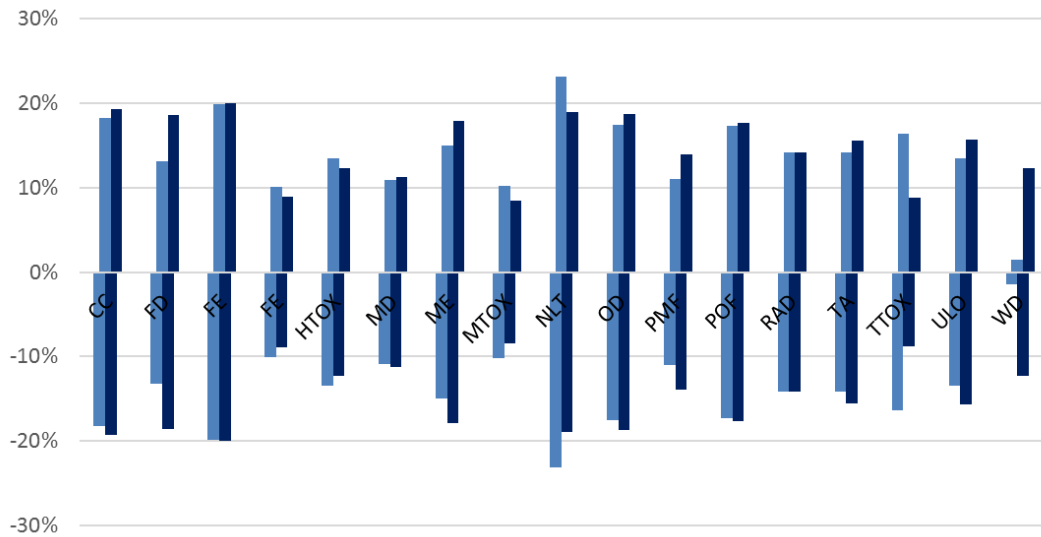


Figure 7. Uncertainty comparison between mix 2 and mix 5 (expressed as a variation coefficient calculated for a 95% confidence interval around the most probable value).

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	Grenland 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 <i>et al.</i> (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 <i>et al.</i> (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 <i>et al.</i> (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 <i>et al.</i> (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 <i>et al.</i> (2018)

Table 2. Physical-chemical properties of the sediments samples used for the tests.

Parameter	Unit	Sample			Assessment			
		1	2	3	Min	Max	Average value	St. Dev.
pH	u. pH	8.50	8.62	8.98	8.50	8.98	8.70	±0.25
Eh	mV	-95.3	-103.0	-105.0	-105.0	-95.3	-101.1	±5.12
Conductivity	mS/cm	3.2	3.6	3.4	3.2	3.6	3.4	±0.20
Moisture content	%	45.1	42.7	46.9	42.7	46.9	44.9	±2.10
Ashes at 600°C	%	83.7	88.3	84.5	83.7	88.3	85.5	±2.50
Organic matter content	%	12.0	16.9	14.6	12.0	16.9	14.5	±2.50
Particle size distribution								
Sand fraction	%	19.4	15.3	23.1	15.3	23.1	19.3	±3.9
Silt fraction	%	43.2	42.0	44.4	42.0	44.4	43.2	±1.2
Clay fraction	%	37.4	42.7	32.5	32.5	42.7	37.5	±5.1
Metals								
As	mg/kgSS	11.85	11.90	11.92	11.85	11.92	11.89	±0.036
Co	mg/kgSS	7.07	7.10	7.10	7.07	7.10	7.09	±0.017
Cr	mg/kgSS	57.20	57.49	57.60	57.20	57.60	57.43	±0.207
Ni	mg/kgSS	38.80	38.90	38.55	38.55	38.90	38.75	±0.180
Pb	mg/kgSS	83.29	83.57	83.07	83.07	83.57	83.31	±0.251
V	mg/kgSS	57.35	57.38	57.02	57.02	57.38	57.25	±0.200
Cu	mg/kgSS	79.97	79.94	80.40	79.90	80.40	80.08	±0.278
Zn	mg/kgSS	205.08	205.93	206.24	205.08	206.24	205.75	±0.601

Table 3. Mixture design for S/S testing.

Mixture element	Mixture design (% of dry sediment)							
	Mix 1	Mix 2	Mix 3	Mix 4	Mix 5	Mix 6	Mix 7	Mix 8
Additive								
Portland cement	10.0	10.0	10.0	10.0	-	-	-	-
Lime	-	-	-	-	10.0	10.0	10.0	10.0
Reagent								
Activated Carbon	-	5.0	-	2.50	-	5.0	-	2.50
Organic Clay	-	-	5.0	2.50	-	-	5.0	2.50
A+R content^(a)	10.0	15.0	15.0	15.0	10.0	15.0	15.0	15.0
Water content	70.0	70.0	70.0	70.0	70.0	70.0	70.0	70.0

^(a): Sum of the Additive (A) and Reagent (R) contents.

Table 4. Lab-Scale stabilization performance in terms of metals removal and leaching test.

Mixes	Parameter	Unit	Metals							
			As	Co	Cr	Ni	Pb	V	Cu	Zn
Mix 1	leachate concentration	mg/l	0.009	0.001	0.009	0.003	0.012	0.255	0.063	< LOD
	Stabilization potential ^(a)	%	99.924	99.986	99.984	99.992	99.985	99.555	99.921	100
	Stabilization goals ^(b)	-	✓	✓	✓	✓	✓	✗	✓	✓
Mix 2	leachate concentration	mg/l	0.007	< LOD ^(d)	0.011	< LOD	0.013	0.314	0.034	< LOD
	Stabilization potential	%	99.941	100	99.981	100	99.984	99.452	99.958	100
	Stabilization goals ^(b)	-	✓	✓	✓	✓	✓	✗	✓	✓
Mix 3	leachate concentration	mg/l	0.009	< LOD	0.010	0.002	0.013	0.297	0.045	< LOD
	Stabilization potential	%	99.924	100	99.983	99.995	99.984	99.481	99.944	100
	Stabilization goals ^(b)	-	✓	✓	✓	✓	✓	✗	✓	✓
Mix 4	leachate concentration	mg/l	0.009	< LOD	0.008	0.001	0.013	0.250	0.035	< LOD
	Stabilization potential	%	99.924	100	99.986	99.997	99.984	99.565	99.956	100
	Stabilization goals ^(b)	-	✓	✓	✓	✓	✓	✗	✓	✓
Mix 5	leachate concentration	mg/l	< LOD	0.001	0.010	0.005	0.008	0.086	0.089	< LOD
	Stabilization potential	%	100	99.986	99.983	99.987	99.990	99.850	99.889	100
	Stabilization goals ^(b)	-	✓	✓	✓	✓	✓	✓	✗	✓
Mix 6	leachate concentration	mg/l	0.006	< LOD	0.007	0.001	0.006	0.006	0.032	< LOD
	Stabilization potential	%	99.949	100	99.988	99.997	99.993	99.989	99.960	100
	Stabilization goals ^(b)	-	✓	✓	✓	✓	✓	✓	✓	✓
Mix 7	leachate concentration	mg/l	0.005	0.001	0.010	0.003	0.006	0.006	0.064	< LOD
	Stabilization potential	%	99.958	99.986	99.983	99.992	99.993	99.989	99.920	100
	Stabilization goals ^(b)	-	✓	✓	✓	✓	✓	✓	✗	✓
Mix 8	leachate concentration	mg/l	0.006	0.001	0.007	0.003	0.006	0.006	0.048	< LOD
	Stabilization potential	%	99.949	99.986	99.988	99.992	99.993	99.989	99.940	100
	Stabilization goals ^(b)	-	✓	✓	✓	✓	✓	✓	✓	✓
Legal limit value leaching test ^(c)		mg/l	0.05	0.25	0.05	0.01	0.05	0.25	0.05	3.00

Symbols in Table 4: ^(a): Stabilization potential is evaluated as: % = $[(C_{TOT} - C_{LEACHATE}) / C_{TOT}] \times 100$; ^(b): Stabilization goals is evaluated: positively (✓) if the metal concentrations are lower than the limit of law; negatively (✗) if the metal concentrations are higher than the limit of law; ^(c): According to the Ministerial Decree 5/02/1998; ^(d): Limit of Detection (LOD) < 0.001 mg/l.

Table 5. Impacts categories defined in the Recipe 1.13. midpoint method.

N.	Impact Category	Symbol	Unit	Normalization values (European set)
1	Climate change	CC	kg CO ₂ eq	0.0000892
2	Ozone depletion	OD	kg CFC-11 eq	45.4
3	Terrestrial acidification	TA	kg SO ₂ eq	0.0291
4	Freshwater eutrophication	FE	kg P eq	2.41
5	Marine eutrophication	ME	kg N eq	0.0988
6	Human toxicity	HT	kg 1,4-DB eq	0.00159
7	Photochemical oxidant formation	POF	kg NMVOC	0.0176
8	Particulate matter formation	PMF	kg PM ₁₀ eq	0.0671
9	Terrestrial ecotoxicity	Ttox	kg 1,4-DB eq	0.121
10	Freshwater ecotoxicity	Ftox	kg 1,4-DB eq	0.091
11	Marine ecotoxicity	Mtox	kg 1,4-DB eq	0.115
12	Ionising radiation	IR	kBq U235 eq	0.00016
13	Agricultural land occupation	ALO	m ² a	0.000221
14	Urban land occupation	ULO	m ² a	0.00246
15	Natural land transformation	NLT	m ²	6.19
16	Water depletion	WD	m ³	0
17	Metal depletion	MD	kg Fe eq	0.0014
18	Fossil depletion	FD	kg oil eq	0.000643

Table 6. Sensitivity analysis for Marine ecotoxicity, reference case (sediment resuspension).

No	Substance	Input							Output							
		Scenario 1 normal			Scenario 2 normal		Scenario 3 uniform		Contribution to variance (%)			Parameter	Scenario 1	Scenario 2	Scenario 3	
		Mean	St. Dev. 1	Dev. CV (%)	St. Dev. 2	CV (%)	Min	Max	Scenario 1	Scenario 2	Scenario 3	Distribution	Normal	Normal	Beta	Normal
-	Total	5.57E-02	-	-	-	-	-	-	-	-	-	Most likely value	0.00901	0.00901	0.07849	0.08
1	Arsenic	1.75E-04	5.29E-07	0.303	5.29E-07	0.303	3.49E-05	8.93E-04	0.0	0.0	0.0	St. Dev.	0.000118	0.000122	0.021809	0.020000
2	Chromium	2.29E-05	8.00E-08	0.350	8.24E-08	0.360	5.84E-06	5.24E-05	0.0%	0.0	1.0	Skewness	-0.00318	-0.01914	0.01885	0.01880
3	Cobalt	8.70E-03	1.72E-05	0.197	2.09E-05	0.240	8.66E-03	8.73E-03	0.0%	1.8	0.0	Kurtosis	2.99	2.96523	2.24876	2.25000
4	Copper	7.54E-03	2.64E-05	0.350	2.62E-05	0.347	8.26E-04	3.64E-02	4.4%	4.3	20.8	CV	1.31%	1.36%	27.78%	27.78%
5	Lead	1.02E-05	3.07E-08	0.300	3.08E-08	0.301	9.43E-07	3.18E-05	0.0%	0.0	0.0	IL, 95%	0.0088	0.0088	0.0349	0.0400
6	Nickel	5.03E-03	1.51E-05	0.299	2.34E-05	0.465	1.58E-03	1.09E-02	1.4%	3.0	0.0	SL, 95%	0.0092	0.0093	0.1221	0.1200
7	Vanadium	3.28E-02	1.15E-04	0.349	1.15E-04	0.349	9.83E-03	7.51E-02	92.1%	90.8	77.8	MSE	1.2E-06	1.2E-06	0.000218	
8	Zinc	1.38E-03	3.46E-06	0.252	4.02E-06	0.292	1.31E-04	4.44E-03	0.1%	0.0	0.3	Chi sq.	74.434	56.8812	90.8176	488.76

Table 7. Monte Carlo analysis comparison across multiple impact categories for the total impacts of mix 2 and mix 5.

Impact category	Most probable value	St. Dev.	CV (%)	Most probable value	St. Dev.	CV (%)	P Mix 2 > mix 5 (%)
CC	199.47	18.250	9.1	86.88	8.366	9.6	100.0
FD	51.81	3.413	6.6	22.68	2.114	9.3	100.0
FE	0.06	0.006	10.0	0.01	0.001	10.0	100.0
FTOX	1.10	0.055	4.5	0.23	0.011	4.5	100.0
HTOX	45.95	3.102	6.8	9.19	0.567	6.2	100.0
MD	3.99	0.218	5.5	3.41	0.191	5.6	97.8
ME	0.05	0.004	7.5	0.03	0.002	8.9	100.0
MTOX	1.05	0.053	5.1	0.23	0.010	4.2	100.0
NLT	0.06	0.007	11.6	0.05	0.005	9.4	86.4
OD	0.00	0.000	8.7	0.00	0.000	9.4	63.0
PMF	0.47	0.026	5.5	0.21	0.015	6.9	100.0
POF	1.03	0.089	8.6	0.72	0.063	8.8	99.7
RAD	25.71	1.817	7.1	21.53	1.521	7.1	96.1
TA	1.18	0.084	7.1	0.48	0.037	7.8	100.0
TTOX	0.01	0.001	8.2	0.01	0.000	4.4	94.2
ULO	5.85	0.395	6.7	4.82	0.379	7.9	97.1
WD	3.31	0.024	0.7	1.54	0.095	6.2	100.0