

# Environmental Geotechnics

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## Characterization, Remediation and Valorization of Contaminated Sediments: A Critical review

ENGE-2022-201-R2 | Paper

Submitted on: 11-05-23

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Keywords: UNITED NATIONS SUSTAINABLE DEVELOPMENT GOALS, DREDGING & RECLAMATION, CONTAMINATED MATERIAL, REMEDIATION



**1 Characterization, Remediation and Valorization of Contaminated Sediments** A**2 Critical review**

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63 **Number of Table:** 3

64 **Number of Figures:** 23

65 **Word count in main body of manuscript:** 7375 words (excluding Abstract, Tables, Figures,  
66 Acknowledgements and Reference section)

67 Submitted for peer review and possible publication in the *Journal of Environmental*  
68 *Geotechnics*

69 Date of revised manuscript Submission: 11<sup>th</sup> JulyMay 2023

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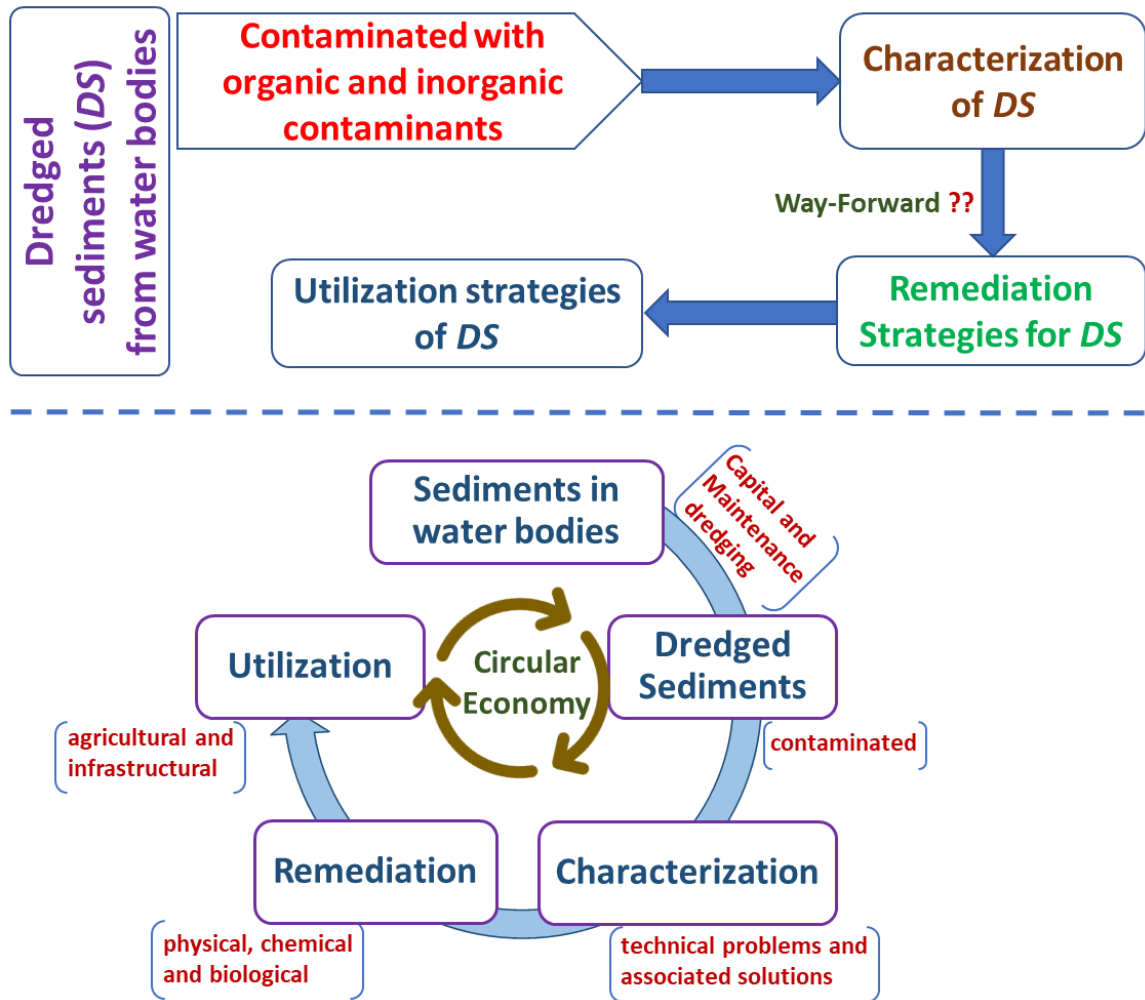
71 **Abstract**

72           The constraints associated with the availability of huge amounts of natural resources  
73 for infrastructure and agricultural development calls for the reuse and recycling of  
74 anthropogenically created geomaterials, which is in line with the UN Sustainable Development  
75 Goals. In this context, valorization of dredged sediments (*DS*), obtained from water bodies such  
76 as rivers, lakes, oceans, etc., as a resource material is worth considering. Unfortunately, *DS*  
77 might be contaminated and exhibit a higher moisture-holding capacity due to higher organic  
78 matter and clay minerals/colloids. These attributes pose a serious question towards dumping of  
79 the *DS* in the deep sea (in the case of marine sediments), a practice which though prevails  
80 presently but endangers marine life. Hence, the way forward would be to characterize them  
81 holistically, followed by adequate treatment to make them ecologically synergetic before  
82 developing a strategy for their valorization. In this regard, many studies have been focused on  
83 the characterization and treatment of *DS* to make them environmentally safe manmade  
84 resource. With this in view, a critical synthesis of the published literature pertaining to the (i)  
85 characterization, (ii) treatment, remediation, and immobilization of contaminants, and (iii)  
86 utilization of *DS* has been conducted, and the salient findings are presented in this paper. Based  
87 on this study, it was observed that the *DS* acts as a sink for emerging contaminants for which  
88 no remediation strategies are available. Moreover, the study highlighted the lacuna in upscaling  
89 the existing treatment and stabilization techniques to field conditions while highlighting the  
90 concept of circular economy.

91 **Keywords:** *sustainable development goals; dredged sediments; contamination; toxicity;*  
92 *remediation; utilization*

93 **Graphical Abstract**

94



95

96 **Highlights**

- 97 • Source, concentration and effect of contaminants.
- 98 • Sustainable development and circular economy perspective of dredged sediments with
- 99 Technology Readiness Levels.
- 100 • Strategies for remediation of contaminants and utilization of dredged sediments.
- 101 • Prospects and recommendations considering policy and guideline issues.

102

## 103 **1 Introduction**

104         The increasing human activities and natural conditions are responsible for the  
105 contamination of water bodies, viz., harbors, ports, estuaries, rivers, lakes, etc. (Akcil *et al.*,  
106 2015). In the realm of the dredging industry (capital and maintenance), the dredged sediments  
107 (*DS*) act like a by-product that has the potential to be utilized to replace natural mineral  
108 aggregates (Achour *et al.*, 2014; ~~Dubois *et al.*, 2009;~~ Loudini *et al.*, 2020). Furthermore, the  
109 emergent demand for construction materials in the infrastructure sector and the environmental  
110 constraints on opening new quarries create an unavoidable need for unconventional  
111 geomaterials like *DS*.

112         The perspective towards dredged material has changed over the past few years from a  
113 waste to a resource, and the utilization of the same is being explored considering the circular  
114 economy and sustainable development (~~Dubois *et al.*, 2009;~~ Gebert and Groengroeft, 2020;  
115 Mehdizadeh *et al.*, 2021). However, *DS* are complex materials due to the presence of salts,  
116 organic matter (*OM*), and contaminants (Rakshith and Singh, 2017). The major contaminants  
117 in *DS* could be classified as (i) inorganic pollutants (potentially toxic elements, viz., zinc,  
118 copper, iron, manganese, cadmium, lead, etc.) and (ii) organic pollutants [viz., polycyclic  
119 aromatic hydrocarbons (*PAHs*), polychlorinated biphenyl (*PCBs*), etc.]. Also, *OM* impacts the  
120 geomechanical performance of sediments due to the increase in voids ratio induced by *OM*  
121 decomposition (Hamouche and Zentar, 2020a, 2020b). In this context, the assessment of the  
122 effects of *OM* on the geotechnical parameters and their evolution in conjunction with *OM*  
123 transformations is one of the relevant aspects to be faced within engineering practice.  
124 However, their potential impact on the environment needs also to be established to design  
125 proper treatments if necessary.

126         Considering the high amounts of *DS* produced worldwide mainly from the marine  
127 environment and the legal constraints associated with their management, their direct disposal

128 in confined disposal facilities/landfilling is no longer economically, socially, and  
129 environmentally feasible (Mehdizadeh *et al.*, 2021; Pal and Hogland, 2022). Further, hydraulic  
130 fills have been utilized in many land reclamation projects, for example, Kansai International  
131 Airport in Osaka Bay, Changi Airport Singapore, etc., were constructed on a hydraulic fill  
132 made of *DS* and soil, which not only allowed for the expansion of the airport but also  
133 contributed to reducing the amount of waste sent to landfills (Choa, 1994; Douglas and  
134 Lawson, 2003; Matsui, 1996). However, it should be noted that the hydraulic fills are not  
135 always the best option for managing the contaminated *DS*. The selection of a management  
136 option is based on various factors including level of contamination, volume of sediments, local  
137 regulations etc. Therefore, a more sustainable fate for *DS* prompts novel research and  
138 management challenges for researchers, management, policymakers, and administrators  
139 (Crocetti *et al.*, 2022; Loudini *et al.*, 2020). Furthermore, the potential utilization of *DS* in  
140 infrastructural and agricultural applications has been tried by earlier researchers (Crocetti *et*  
141 *al.*, 2022; Hamouche and Zentar, 2020a; Rakshith and Singh, 2017). However, *DS* toxicity  
142 and contamination level being case- and site-specific, more extensive studies focusing on its  
143 utilization schemes need to be performed by the research communities.

144 From the existing literature, it was realized that the number of publications considering  
145 the valorization of *DS* with a focus on sustainable development is less, whereas that for the  
146 circular economy perspective is almost negligible. The reviews conducted till date on *DS* are  
147 limited to either contamination, or management, or application aspects, which does not give a  
148 broader perspective about contaminated sediments. Keeping in view of these mentioned  
149 findings, this paper synthesizes the recent developments in the field of contamination  
150 associated with the dredged material, their characterization, followed by remediation and  
151 utilization strategies considering the sustainable development and circular economy aspects.



152 Furthermore, the necessities associated with the policy and guidelines have been critically  
153 evaluated, and a brief account of the same has been discussed in the following sections.

## 154 **2 Source, concentrations, and effects of emerging contaminants in dredged sediments**

155 Emerging contaminants (*ECs*) are ‘any synthetic or naturally occurring chemical or any  
156 microorganism that is not commonly monitored in the environment, but has the potential to  
157 enter the environment and cause known or suspected adverse ecological and/or human/aquatic  
158 life/wildlife health effects’ (Smital, 2008). The *ECs* need not been found in the environment in  
159 the recent past but may persist over the decades in small concentrations (i.e.,  $\mu\text{g/L}$  and  $\text{ng/L}$ )  
160 and found to be of concern due to (i) exponential growth in the utilization of products  
161 contributing to them, and (ii) increase in their adverse effects on the environment and life on  
162 the planet. For instance, the per- and polyfluoroalkyl substances (*PFAS*) based products such  
163 as paints, sealants, water-resistant clothing, grease-resistant papers, fast food containers, and  
164 nonstick cookware are being used since the 1950s, but widely found in different environmental  
165 systems after development and improvement in the sensitivity of mass spectrometers in 1980s,  
166 which subsequently led to their classification as *ECs* in early 2000s (Richardson and Kimura,  
167 2017). Hence, the *ECs* are also known as ‘chemical of emerging concerns’ or ‘contaminants of  
168 emerging concerns’ (Rosenfeld and Feng, 2011).

169 The sources of *ECs* in *DS* can be classified as primary and secondary. The primary sources can  
170 be defined as the initial point of contact wherein the *ECs* are used in the manufacturing of the  
171 products to attain the desired properties. The primary sources of *ECs* include pharmaceutical  
172 and personal care products, biocides (including agricultural and plant protection products),  
173 disinfection by-products, industrial chemicals (viz., lubricants, flame retardants, gasoline,  
174 antimicrobial agents, surfactants, food additives, and plasticizers), bioterrorism and sabotage  
175 agents, algal toxins, etc. (Barber, 2014; Rosenfeld and Feng, 2011).

176           The secondary sources of *ECs* include industrial sludges and wastewater, surface water  
177 bodies, municipal solid waste, industrial by-products and soils contaminated with industrial  
178 discharges and chemicals (refer to *Figure S1*). Furthermore, micro(nano)plastics can also be  
179 considered as the potential secondary source of *ECs* because they can fragment, degrade and  
180 leach one or more of the *ECs*, such as persistent organic pollutants (Goli *et al.*, 2021; O’Kelly  
181 *et al.*, 2021). The primary sources majorly contaminate the *DS* through their deposition,  
182 leaching, and sorption, while the secondary sources would contaminate by the sorption  
183 mechanism. However, the dominant mechanisms which contribute to *ECs* in *DS* would  
184 completely depend on the characteristics of the latter and environmental conditions to which  
185 the primary sources are exposed.

186           The contamination of *DS* through primary sources can be more often observed in the  
187 developing and under-developed countries where the guidelines for liquid and solid waste  
188 collection, transportation, and treatment are not enforced strictly or not available. Unlike the  
189 primary sources, secondary sources of *ECs* are the major pathways for contamination of the  
190 *DS* in all countries due to the fact that the removal of the *ECs* is not the primary motive of the  
191 domestic and industrial wastewater treatment plants, municipal solid waste leachates and  
192 sludges up to the recent past.

193           Furthermore, the determination of concentrations of *ECs* in *DS* is mostly limited to a  
194 few compounds based on *PAHs* and *PCBs* because these are major contaminants emitted during  
195 the vehicular and vessel movements that are essential for offshore transportation, recreational  
196 activities and nearby industrial activities (Kafilzadeh, 2015; Norén *et al.*, 2020) (refer to *Table*  
197 *1*). The *ECs* contamination, their possible sources, the source of *DS*, and the detection  
198 techniques studied by earlier researchers have been presented in the *Table 1*.

199

200

**Table 1.** Summary of the studies conducted on the concentrations of *ECs* in *DS*

Reference	Study area	Source of <i>DS</i>	<i>ECs</i> detected with concentration	Possible source of <i>ECs</i>	Detection techniques
Torres <i>et al.</i> (2009)	Port of Santos, Brazil	Marine sediments (18 samples from dredged areas and disposal sites, 4 samples from hopper dredge)	<i>PAH</i> (27.86 to 679.35 µg/kg); <i>PCB</i> (0.17 to 12.33 µg/kg)	Emissions and activities of steel plant and industrial complex	Gas Chromatography/Mass Spectroscopy ( <i>GC/MS</i> )
Rocha <i>et al.</i> (2011)	Porto region, Portugal	4 river estuary and 2 marine beach sediments	<i>PAH</i> [Estuary (98.40 to 156.50 µg/kg dw); Marine sediment (52.00 to 54.80 µg/kg dw)]	-	<i>GC/MS</i>
Tavakoly Sany <i>et al.</i> (2014)	Klang strait, Malaysia	Coastal sediment	16 compounds of <i>PAHs</i> (994.02±918.10 µg/ kg dw)	Contamination due to cargo transport, petrogenic spillage and pyrogenetic combustion	<i>GC/MS</i>
Kafilzadeh (2015)	Soltan Abad river, Iran	River sediment (4 sampling locations at a depth of 5 cm from the bed)	16 compounds of <i>PAHs</i> (180.30 to 504.00 µg/kg)	Pyrogenic combustion and petrogenic spillage	Gas Chromatography/Flame Ionisation Detection ( <i>GC/FID</i> )
Couvidat <i>et al.</i> (2018)	Port in the south of France	Harbour sea bed (Top 50-80 cm)	16 compounds of <i>PAHs</i> (62.18-62.40 mg/kg) 7 compounds of <i>PCBs</i> (0.96-0.97 mg/kg) 3 compounds of Organotin compounds (65.50 mg/kg)	Extensive anthropogenic activity for centuries and contamination due to industrial activity	<i>GC/MS</i> and low-resolution <i>MS</i>
Shilla and Routh (2018)	Rufiji Estuary, Tanzania	River sediment (top 1-2 cm sediment was scrapped on South, middle and north parts of Rufiji Delta)	19 compounds of <i>PAHs</i> (128 to 377 µg/kg)	Petrogenic spillage and pyrogenic combustion of coal and biomass (mainly grass and wood)	<i>GC/MS</i>
Norén <i>et al.</i> (2020)	Two ports, three marina and one waterway leading to the marina in Sweden	Marine environment	Tributyltin: ports (150230 mg/kg); marina (50±50 and 310±240 mg/kg); waterway (70±60 mg/kg)	Pollutants released by recreational and public transport boats, cargo vessels. Effluents from Cu production, wastewater treatment, battery production industries and shipyards.	-

201

### 202 **3 Geotechnical characterization: technical problems and adopted solutions**

203 The characterization of contaminated sediments to address environmental issues related to  
204 the remediation of polluted areas is aimed to build the so-called Conceptual Design Site Model  
205 (*CDSM*). The *CDSM*, includes the most relevant site features (i.e., water, soil/sediment and  
206 biota properties, together with land waterway use) as well as the processes ongoing within the  
207 system. Stemming from the traditional Conceptual Site Model (*CSM*), the *CDSM* is originally  
208 meant to be an updated model including chemical, geo-hydro-mechanical and environmental  
209 engineering knowledge about the processes ongoing within the relevant volume of the system.  
210 It supports a more sustainable choice of remedial strategies since it is capable of taking account  
211 of at least two (Environment and Engineering) of the *four-E* (Environment, Economy, Equity  
212 and Engineering) criteria of the multi-dimensional approach towards *sustainability* (Basu *et al.*,  
213 2015). Moreover, being centered on the knowledge of processes, it can more efficiently  
214 support the first predictions of the system evolution, both in the short and in the long term, that  
215 would accompany the remediation phase (Vitone *et al.*, 2020).

216 It follows that it becomes a strategic tool to address both the selection of sustainable  
217 remedial strategies and the technology screening phase of contamination (Reible, 2014;  
218 USEPA, 2019). In this model, the geo-hydro-mechanical characterization of the sediments  
219 provide geotechnical parameters which have a direct effect on the feasibility of all remedial  
220 technologies and supports the predictions of the *DS* behaviour before and after treatment  
221 (Adamo *et al.*, 2018; Roque *et al.*, 2022; Vitone, 2020).

222 For example, in situ capping is a remediation option that can be selected and designed only  
223 after a site characterisation which includes geotechnical considerations (Vitone *et al.*, 2016).  
224 Usually, contaminated sediments are predominantly fine-grained and often have high water  
225 content and compressibility, and low shear strength. Cap stability and settlement due to  
226 consolidation are geotechnical issues that may be important for cap effectiveness. After

227 placement of a cap, consolidation of both the underlying contaminated sediment and the cap  
228 layer usually occurs (Reible *et al.*, 2014). The consolidation of the cap is typically small. On  
229 the other hand, the consolidation of the underlying contaminated sediments may be significant,  
230 especially when dealing with soft soils, and expresses porewater pressure from the  
231 contaminated layer up into the cap. Moreover, the fluid expelled during the consolidation  
232 process should be evaluated for the investigation of a contaminated marine site. In fact,  
233 contaminant migration can change sediment properties (e.g., consistency limits), influencing  
234 capping design (Erten *et al.*, 2011). The impact of these processes depends on the sediment  
235 geotechnical properties that should be known for an efficient design of capping (Reible, 2014).

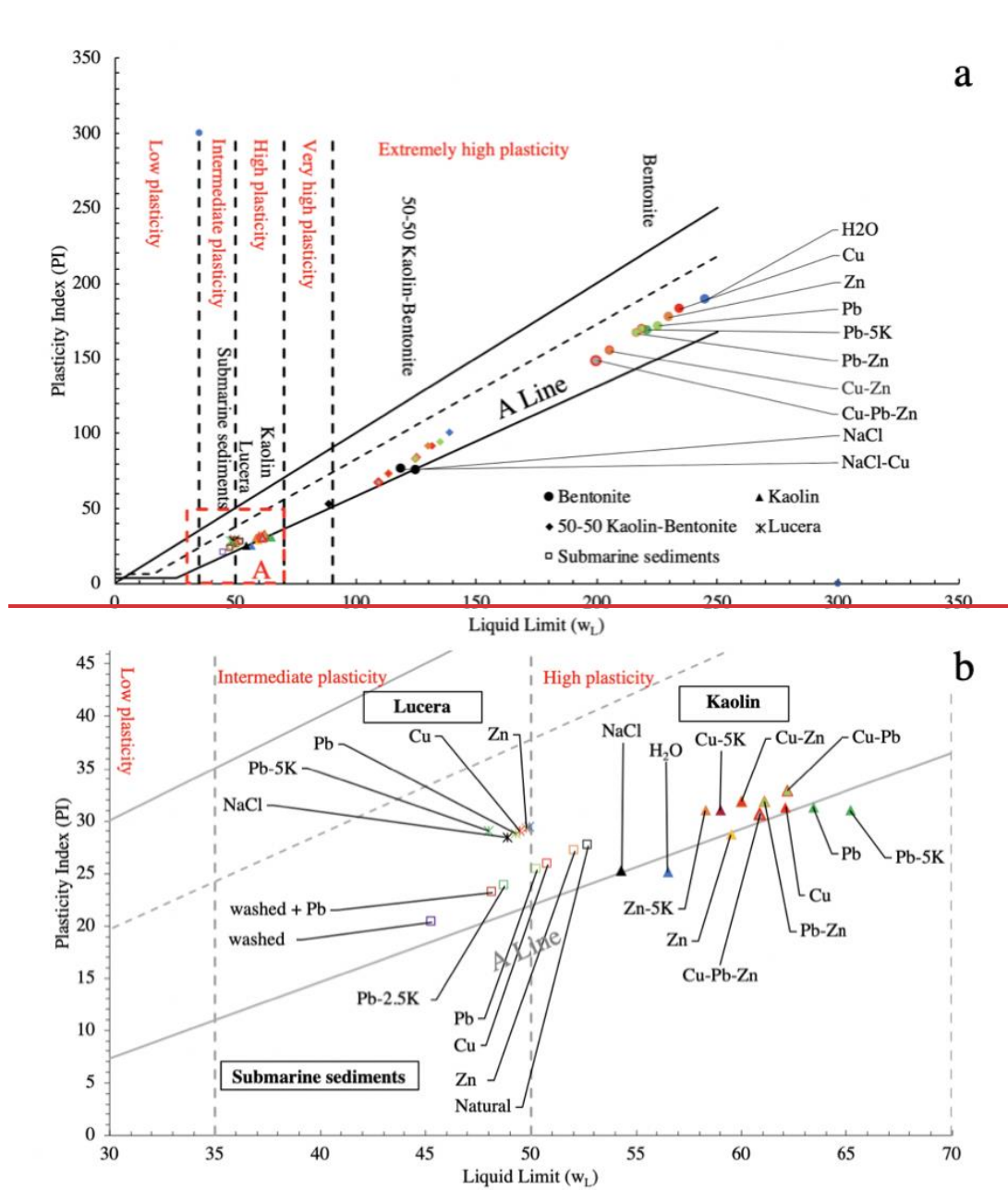
236 However, marine sediments may contain *OM*, shells, microfossils and diatoms, salts, heavy  
237 metals, and organic pollutants, which may induce some bias in the measurement and  
238 classification of fine-grained soils, which makes geotechnical characterization quite  
239 challenging. The framework defined for normally-consolidated natural clays and the laboratory  
240 standards does not focus on soils containing sources of complexity such as those typical of  
241 contaminated marine sediments. As reported in the following paragraphs, some novelties need  
242 to be introduced in the phase of soil testing and data analysis to accurately measure the state,  
243 physical and mechanical properties of contaminated sediments.

### 244 **3.1 Impact of organic matter, heavy metals, fossils, and other pollutants**

245 *DS* from natural environments may be characterized by a significant content of buried *OM*  
246 coming from the biosynthesis of organisms existing in the water column. Moreover, in shallow  
247 marine basins, near the coast, the terrigenous contribution of *OM* (allochthone *OM*) might  
248 occur. Organic particles can be adsorbed by negatively charged mineral surfaces and promote  
249 the aggregation of clay-size particles to form a more open fabric. If *OM* content is high, soils  
250 may be characterized by unusually high-water contents, plasticity, and activity index, with  
251 exceptionally low wet bulk densities and high compressibility (Levesque *et al.* 2007).

252 Furthermore, non-decomposed organic substances and microbial populations will have a  
253 binding effect on the soil particles (Bobet *at al.*, 2011), which reduces the soil plasticity and  
254 activity indexes of clays (Sollecito *et al.*, 2021).

255 Recent research by Muththalib (2020) and Muththalib and Baudet (2019) on kaolin,  
256 bentonite, mixtures of kaolin and bentonite, illite rich Lucera clay, and submarine sediments  
257 from the Port of Taranto (Mar Piccolo) showed the effect of heavy metal contamination on  
258 their physical and mechanical properties. In *Figure S2*, the plasticity of kaolin is seen to  
259 increase with heavy metal contamination, with a reverse effect observed in bentonite and hardly  
260 noticeable effects in the illite rich Lucera clay. The reason behind this observation might be the  
261 difference in their *pH* and electrical conductivity, and the alterations in these properties due to  
262 the presence of heavy metals (*viz.*, Cu, Zn, Pb, etc.) in soluble form.



263  
 264 **Figure 1.** (a) Casagrande's Plasticity Chart for all the clays studied, (b) Zoomed in to show  
 265 area A marked in (a) (Muththalib, 2020) (Note: The legends for Figure 1 b are same as  
 266 mentioned in Figure 1 a; and 'K' in Figure 1 represents 1000 ppm)

267

268 *Figure AS3* summarizes the variation in the plasticity of different clays with the presence of  
269 heavy metal contamination. Pure kaolin, pure bentonite, kaolin-bentonite mixtures, submarine  
270 sediments from the Port of Taranto and an illite-smectite rich clay (Lucera) were tested with  
271 salt or heavy metals used single or combined. Copper, Lead and Zinc were chosen at  
272 concentrations of 1000 ppm unless specified. In *Figure AS3*, the symbol shapes characterize  
273 the soil tested while the colour represents the added salt or metal(s).

274 The thermogravimetry tests could be coupled to geotechnical testing to explore the sediment  
275 skeleton's nature and its *OM* content, based on the main thermal reactions occurring within  
276 different temperature ranges (Sollecito *et al.*, 2021). The *DS* having a substantial quantity of  
277 *OM* should not be oven dried before testing for the Atterberg limit determination because the  
278 liquid limit decreases when the organic soil is oven-dried before testing (ASTM D2487-ASTM,  
279 2011). Furthermore, the sieving procedure at 425- $\mu\text{m}$  (No. 40) sieve required for the  
280 preparation of material for the Atterberg limit determination (ASTM D4318-ASTM, 2017),  
281 may remove the organic components and alter the sediment plasticity (Roque *et al.*, 2022).

282 The testing on marine sediments may be further compounded by the widespread presence  
283 of lapideous elements and fragments of shells, mussels, fossils, and diatoms, whose dimensions  
284 could vary from some centimeters to a few micrometers. The presence of these elements in the  
285 soil matrix has been found to alter the soil fractions of sediments retrieved in the Mar Piccolo,  
286 a highly polluted marine basin in southern Italy (Cotecchia *et al.*, 2021).

287 Also, inclusions only visible at the micro-scale can introduce some bias in the sediment  
288 characterization. The presence of microfossils and diatoms (*Figure S34(e)*), of high intra-  
289 skeletal voids space can provide an apparent increase of the soil plasticity and activity indexes,  
290 as well as the soil compressibility, irrespective of the clay fraction size and typology (Caicedo  
291 *et al.*, 2018; Sollecito *et al.*, 2021).



292 The effect of the remediation treatment depends on several factors. For example, in the case  
293 of ex situ stabilization/solidification treatments, the quantity of additive, the curing time,  
294 composition and physical properties of the sediments and water chemistry. In particular, the  
295 contaminants can interfere with the sediment properties (e.g., consistency limits)  
296 compromising the effectiveness of the stabilization. It follows that the optimization of the  
297 treatment depends on the type of contaminants, soil physical properties, composition, and the  
298 required performance (Todaro *et al.*, 2020; Vitone *et al.*, 2020; Wang *et al.*, 2018).

### 299 3.2 Effect of pore water salinity

300 The effects of chemo-mechanical coupling in soils are usually interpreted according to the  
301 Gouy-Chapman diffuse double layer (*DDL*) theory (Chapman, 1913; Gouy, 1910). According  
302 to this theory, the thickness of the *DDL* in clays decreases when either the pore water ion  
303 concentration or the cation valence increases and the dielectric constant decreases, as for clays,  
304 including high concentrations of salts, metal ions, and organic pollutants. These conditions  
305 favor clay particle flocculation and prompt significant variations of the soil index properties,  
306 mechanical parameter values, and testing procedures, with respect to those consisting  
307 uncontaminated pore solution, and thus the salt concentration in the pore-fluid should also be  
308 considered (Di Maio and Scaringi, 2016; Jang and Santamarina, 2016; Mitchell and Soga,  
309 2005; Sollecito *et al.*, 2019a). A reduction of liquid limit,  $w_L$ , and compression index,  $C_c$ , is  
310 generally recorded in active clays when pore fluid salinity increases (Di Maio *et al.*, 2004).  
311 Special consideration should therefore be paid to sediments from the marine environment  
312 where high soluble salt concentrations are present, especially when remediation strategies such  
313 as washing and decontamination must be undertaken since they may change the pore fluid  
314 chemistry and, in turn, the soil behavior.

315 To take into account the presence of salts, the water content data obtained through oven-  
316 drying (ASTM D2216-ASTM, 2019) should be corrected for the salinity values (ASTM

317 D4542-ASTM, 2015; Imai et al., 1978; Nash et al., 1992; Sollecito et al., 2021). Furthermore,  
318 the use of a fluid with the same salinity of the pore water for laboratory experiments is  
319 recommended by several earlier researchers (Di Maio and Onorati, 2000; Di Maio et al., 2004;  
320 Baudet and Ho, 2004; Sollecito et al., 2019a). This is the case of the preparation of the material  
321 for the liquid limit determination (Di Maio et al., 2004; Sollecito et al., 2019a); the preparation  
322 of reconstituted samples (Baudet and Ho, 2004); and the filling of oedometer or direct shear  
323 tests cells, where differences in the fluid composition may induce the flow of water or ions  
324 through the soil (Di Maio and Onorati, 2000). The use of water with salt concentration same  
325 as the sample pore fluid is also recommended to apply the cell pressure during triaxial tests to  
326 avoid building up osmotic pressures across the sample membrane because of differences in  
327 salinity (Baudet and Ho, 2004).

### 328 3.3 Use of statistical techniques for integrated sediment characterization

329 As previously reported, the characterization of contaminated sediments requires assessing  
330 several variables, including contaminant source, contaminant type, the sedimentary up to even  
331 the hydrologic environment, or natural features such as sediment grain-size distribution,  
332 composition, the effect of transportation (including here the cross-shore and long-shore  
333 transport of sediment). Due to the potentially high costs associated with the management of  
334 contaminated sediments and their remediation process, the assessment of the degree of  
335 contamination becomes paramount as (i) the inaccurate determination can result in wasting of  
336 considerable financial resources related with unnecessary treatment measures, and (ii) poses  
337 both ecological and human health risks. In this sense, the characterization phase and  
338 determination of the contamination degree becomes critical and the subject of the intense  
339 inspection.

340 During the characterization phase, a deterministic approach in analyzing the parameters  
341 selected to describe the ecosystem can surely provide a great deal of information and drive the

342 remediation strategies (Cotecchia *et al.*, 2021). Nevertheless, the inspection of the complex  
343 dataset that is generated from the investigation campaign cannot easily allow the understanding  
344 of the factors of key relevance, which impact and control the spatial and temporal distribution  
345 of contaminants in the ecosystem. Aiming to address such complexity, environmental scientists  
346 have started employing multivariate statistical approaches that constitute an advantage in the  
347 assessment and modelling of contamination patterns of highly contaminated areas on a large  
348 scale and thus could contribute to effective and economical monitoring of their quality.

349 The statistical techniques have been widely exploited in the literature since they support the  
350 generation of spatial pollution maps and identify potential interaction stressors in contaminated  
351 areas (Hopke 2015; Mali *et al.*, 2016, 2017, 2022). Successful examples can be reported, such  
352 as the characterization of the *EC* distribution in the sediments in one of the most polluted  
353 Mediterranean coastal basins, Mar Piccolo (Mali *et al.*, 2017); or the influence of Sarno river  
354 discharges onto Gulf of Naples, a marine basin subjected to a highly anthropized coastal area  
355 (~~Montuori *et al.*, 2013~~; Mali *et al.*, 2022).

356 It has been reported by Mali *et al.* (2017) that the combination of principal component  
357 analysis (*PCA*) and analysis of variance (*ANOVA*) revealed synergistic effects of independent  
358 factors such as total organic carbon (*TOC*) and Grain Size and allows to understand the *TOC*  
359 concentration resulted to be dominant conditioning factor with respect to granulometry.

360 Therefore, the characterization of complex matrices such as marine and harbor sediments  
361 needs advanced tools that are able to investigate the complex pattern that arises from the  
362 superposition of natural and anthropogenic processes and from multiple factors acting  
363 simultaneously on a local scale.

## 364 **4 Remediation strategies for contaminated sediments**

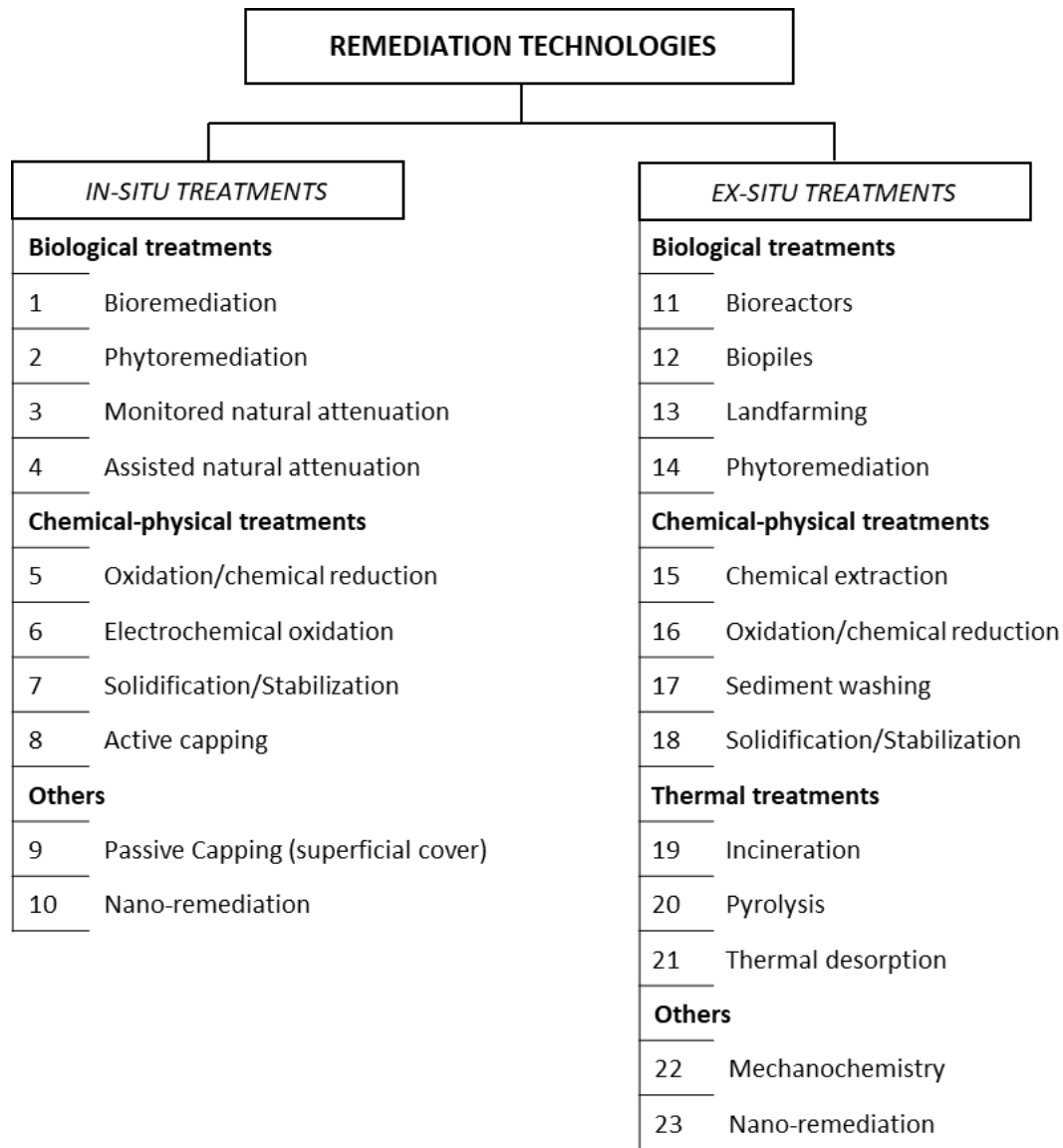
### 365 **4.1 Technologies overview**

366 The remediation of contaminated marine sediments is more complex than managing  
367 contaminated soil or groundwater sites. Hence, the choice of approach(es) is generally broad  
368 and complex, frequently conflicting, and often controversial (Todaro *et al.*, 2018). As a result,  
369 the management and remediation of contaminated sediments is a significant issue faced by  
370 environmental policymakers, scientists, and engineers (Labianca *et al.*, 2021).

371 Sediment remediation techniques are commonly classified as in-situ (i.e., treatments  
372 operating without sediment dredging) and ex-situ (i.e., treatments including sediment  
373 dredging). In-situ technologies allow soil and sediment to be treated without being excavated  
374 and transported, with potentially significant savings. However, in-situ treatments generally  
375 require a longer remediation time (Lofrano *et al.*, 2017). Instead, ex-situ treatments require the  
376 dredging of sediments, leading to increased costs and engineering for equipment, possible  
377 permitting, and material handling/worker exposure considerations. However, the main  
378 advantage of ex-situ treatment is that it requires shorter periods than in-situ treatment, and there  
379 is more certainty about the uniformity of treatment because of the ability to screen and mix the  
380 sediments (Zhang *et al.*, 2021).

381 Moreover, treatment methods can be categorized into three major groups: (a)  
382 physical/chemical, (b) biological, and (c) thermal (Todaro *et al.*, 2016; De Gisi *et al.*, 2017a).  
383 Furthermore, the classification of the remediation technologies has been proposed in *Figure*  
384 [21](#). In *Figure 21*, various in-situ and ex-situ treatment techniques for sediment remediation  
385 have been presented, which need to be adopted based on the contamination level and type and  
386 thus need further extensive research in this context. Furthermore, this paper focuses on the  
387 more competitive technologies in the direction of sustainable development.

388



389

390 **Figure 21.** Sediments remediation technologies for in-situ and ex-situ treatments

#### 391 4.2 In situ capping

392 In situ remedial alternatives generally involve: (i) natural attenuation, which is based  
 393 on the assumption that, although sediments pose some risk, it is low enough that natural  
 394 processes can reduce risk over time in a reasonably safe manner (De Gisi *et al.*, 2017b); (ii) in  
 395 situ containment and treatment via capping, in which contaminated sediment is physically and

396 chemically isolated from aquatic ecosystems or the contaminants in sediment are sequestered  
397 and degraded (Todaro *et al.*, 2018).

398 Capping is the process of placing a layer of clean materials over contaminated  
399 sediments to isolate the contaminant from the overlying water column and biota, to reduce  
400 contaminant flux into the biologically active portion of the sediment, and to create new habitats  
401 for aquatic organisms (Reible, 2014). Conventional (passive) caps consist of placing a layer of  
402 clean neutral materials (such as sand, silt, clay, and crushed rock debris) that rely on  
403 containment, rather than treatment, of contaminated sediment. The cap may also include  
404 geotextiles to aid in layer separation or geotechnical stability, amendments (that is of  
405 chemically reactive materials) to enhance protectiveness, or additional layers to armor and  
406 maintain its integrity or enhance its habitat characteristics. When these amendments are added  
407 to cap material, the technology is called an “Active Cap” (or “Reactive Cap”), and the  
408 amendments enhance the performance of the cap material. The use of chemically reactive  
409 materials allows sequestering and/or degrading of sediment contaminants, reducing their  
410 mobility, toxicity, and bioavailability, performing both containment and treatment of  
411 contaminated sediment. The comparison between passive capping and active capping is listed  
412 in *Table S1*. Active/Reactive Cap presents several advantages; however, the capping  
413 technology selection depends on site characteristics (e.g., contamination levels and  
414 geotechnical properties of sediments).

415 Sand capping has been widely investigated, being mostly utilized for large availability  
416 and ease of placement of sand (Jiao *et al.*, 2020). In Bortone *et al.* (2018) a sand cap was  
417 investigated to reduce the exposure of the aquatic ecosystem to *PCB*-contaminated sediments  
418 in Lake Hartwell, US. Specifically, it was demonstrated that *PCB* transport was extremely  
419 dependent on the cap characteristics, and that the cap thickness could be reduced at 20 cm-  
420 thick by using a high sorbent cap. In the study by Meric *et al.* (2014), a 7.5 cm thick sand cap

421 reduced the bioavailability of *PCBs* by a factor of 100 compared to the no-capping scenario,  
422 but it did not influence the bioavailability of naphthalene.

423 However, a passive cap might partially allow dissolved contaminants into the overlying  
424 water column and consequently still pose a risk to the benthic environment and the trophic  
425 chain. In these cases, reactive materials can be used. Murphy and Lowry (2004) demonstrated  
426 that a thin layer of the adsorptive amendment (i.e., activated carbon) could have a more  
427 performing pollution containment capacity for *PCBs*, equal to over 100 times sand adsorption  
428 effectiveness. Other reactive amendments (such as calcite, zeolite, apatite, organoclay, and  
429 biopolymers) can also sequester or degrade a variety of contaminants and control their mobility  
430 to the water column (*Table 2*).

431 **Table 2.** Summary of amendments used to treat organic and inorganic contaminants

Function	Amendment	Contaminant targeted	Reference
Sequestering	Activated Carbon, AC	Organics/inorganic	Choi (2018); Silvani <i>et al.</i> (2017)
	Apatites	Metals	Knox <i>et al.</i> (2012); Xing <i>et al.</i> (2016); Zhang <i>et al.</i> (2016)
	Bauxite	Metals	Taneez <i>et al.</i> (2018)
	Biochars	Organics/inorganic	Bianco <i>et al.</i> (2021); Janssen and Beckingham (2013); Ting <i>et al.</i> (2020)
	Organoclays	Organics/inorganic	Erten <i>et al.</i> (2012); Olsta (2010); Pagnozzi <i>et al.</i> (2020)
	Zeolites	Metals	Gu <i>et al.</i> (2019); Kang and Park (2015)
Degrading	Bioremediation agents	Organics	Atashgahi <i>et al.</i> (2014); Sun <i>et al.</i> (2010); Wang <i>et al.</i> (2014)
	Zero-Valent Iron, ZVI	Organics	Chapman <i>et al.</i> (2020); Hu <i>et al.</i> (2020)

432

433 In this regard, an innovative use of reactive capping entails encapsulation of the reactive  
434 amendments between two geotextile layers by further reducing reactive layer thickness to about  
435 1 cm (Meric *et al.*, 2014; Bortone *et al.*, 2020). It includes additional benefits such as uniform  
436 consolidation and defined mass per area. Alternatively, innovative reactive capping is  
437 represented by reactive granular materials where the reactive amendments cover an inner inert  
438 core more stable and slough off the aggregate during hydration, enabling mixing with the  
439 contaminated sediment.

#### 440 **4.3 Stabilisation/Solidification (S/S)**

441 The immobilization and stabilisation of the metals and other contaminants from the *DS*  
442 can be achieved using lime, cement, silicates, and other additives, which subsequently enhance  
443 the matrix's compressive strength. In this context, Stabilisation/Solidification (S/S) has been  
444 elaborately discussed in the following sections.

##### 445 **4.3.1 Traditional chemical reagents**

446 The contamination of aquatic sediments with organic and/or inorganic pollutants is a  
447 widespread environmental problem. Stabilisation/Solidification (S/S) is a chemo-mechanical  
448 treatment that makes use of chemical reagents, such as cement, lime, and other binders.  
449 Cement-based S/S is a chemical treatment process that aims to either bind compounds of a  
450 hazardous waste stream into a stable insoluble form (stabilisation) or to entrap the waste within  
451 a solid cementitious matrix (solidification) (Wiles, 1987). It is widely established as an  
452 effective method for both improving the engineering properties of sediments and encapsulating  
453 contaminants (Barjoveanu *et al.*, 2018; Dermatas and Meng, 2003; Wang *et al.*, 2012; Levacher  
454 *et al.*, 2011; Miqueleiz *et al.*, 2012; Palansooriya *et al.*, 2020; Qian *et al.*, 2008; Silitonga *et al.*,  
455 2009; Valls and Vázquez, 2000; Wang *et al.*, 2012; Zentar *et al.*, 2012). The US Environmental  
456 Protection Agency (USEPA) defines S/S treatment as “a process that encapsulates waste to  
457 form a solid material” (USEPA, 1997).



458           The contaminated sediments are converted into solid forms and entrapped within a  
459 granular or monolithic matrix through the chemical reactions developed during the process of  
460 solidification. The treatment limits the mobility or solubility of the hazardous components and  
461 does not necessarily alter the physical nature of the contaminants (USEPA, 2004). The  
462 combined application of the solidification and stabilisation process ensures the mixing of the  
463 contaminated sediments with the treatment agents and consequently, both the physical and  
464 chemical immobilisation of the hazardous components occurs. The ultimate objective of *S/S* is  
465 to complete the transformation of toxic components into nontoxic forms. However, the  
466 objective of *S/S* technology not only includes limiting the solubility of the contaminant and  
467 decreasing the surface area across which contaminant transport might occur, but also the  
468 improvement of the mechanical properties of the sediments (soils).

469           In addition, *S/S* treatments offer several advantages over other treatment technologies  
470 (e.g., Oh et al., 2011), including: i) costs, and ii) implementability. As exemplified in Table 3,  
471 several are the examples of their use to improve the physical, mechanical, and environmental  
472 properties of *DS*. The authors show that *S/S* treatments are effective to: (a) reduce the initial  
473 fluid content of sediments, (b) eliminate or stabilize the hazardous compounds, such as heavy  
474 metals and *OM*, (c) improve the mechanical properties of the sediments and (d) prompt the  
475 production of new geomaterials or granular materials to address novel options of sediment  
476 management, i.e., base materials for pavement construction, cement production, light-weight  
477 concrete production and brick fabrication. However, a notable disadvantage of *S/S* is that, the  
478 contaminants, although immobilised, are still present in the sediments. Moreover, organic oily  
479 compounds can represent a threat for the efficacy of cement stabilisation.

480

**Table 3.** S/S treatments of sediments in the literature

Reference	Material	Binder	Effect	Purpose
Boutouil and Levacher (2000)	Sediment contaminated by heavy metals (port of Le Havre)	Cement	- Increase in compressive strength - Decrease in leaching of heavy metals	Road or civil construction materials
Colin (2003)	Sediment (Rouen harbour)	Cement	- Improvement of physical, mechanical, and environmental properties	Road bed materials
Scordia <i>et al.</i> (2008)	Sediment contaminated by heavy metals and organic matter (channel linking Charleroi to Brussels)	Roc Sol (commercial product) and lime	- Increase in bearing capacity, compressive strength and Brazilian tension - Decrease in expansive behaviour	
Silitonga <i>et al.</i> (2010)	Sediment contaminated by heavy metals (port En-Bessin)	Cement and silica fume	-Increase in compressive strength - Decrease in leaching of heavy metals	
Wang <i>et al.</i> (2012)	Marine sediment (Dunkirk)	Lime and cement	- Increase in unconfined compressive strength and tensile strength	
Zentar <i>et al.</i> (2012)	Marine sediment (Dunkirk)	Cement and fly-ash	- Increase in tensile strength and compressive strength - Restrain of the swelling potential	
Kogbara (2014)	Contaminated sediment	Cement and blends of cement–fly ash, cement–slag, lime–slag, lime–fly ash	- Increase in compressive strength. - Decrease in leaching of heavy metals	Sediment management
Radenović <i>et al.</i> (2019)	Highly contaminated sediment, dominantly by heavy metals (Great Bačka canal)	Kaolinite, quicklime and cement	- Decrease in leaching of heavy metals	
Mastoi <i>et al.</i> (2022)	Sediment (Nanhu lake located in the Chinese city of Wuhan)	Cement	- Increase in compressive strength	Civil construction materials

481

482 With respect to the binders and additives used for *S/S* treatments of sediments, the  
483 literature reports a variety of solutions: lime or cement alone (e.g., Jaubertie *et al.*, 2010,  
484 Federico *et al.*, 2015), lime combined with high alkali and slag cements, fly ashes (Grubb *et*  
485 *al.*, 2010) or pozzolana (e.g., Zoubir *et al.*, 2013), to cite a few of them. Due to the heterogeneity  
486 in the properties of *DS* and physicochemical reactions between binders and metal and organic  
487 contaminants, different binders show different efficiencies for pollutant immobilisation. The  
488 ability of Portland cement to improve the sediments' geotechnical characteristics (Wang *et al.*,  
489 2012; Zentar *et al.*, 2012) and immobilise contaminants has been widely documented (Xue *et*  
490 *al.*, 2017; Wang *et al.*, 2018). However, there are some cases in which the mobility of  
491 contaminants in marine sediments does not reduce when treated with either lime and cement  
492 or additives (Saussaye *et al.*, 2016; Taneez *et al.*, 2016; Xu, 2017). Overall, most studies to  
493 date have focused on individual parameters and there remains a lack of systematic research  
494 considering the combination of factors affecting the properties of stabilised sediments in a  
495 unified way. In the *S/S* treatment of organic compounds using cement alone, the contaminants  
496 are physically trapped within the pores in the cement matrix and are not reacting with the polar  
497 inorganic components of the cement constituents. The use of adsorbents such as organophilic  
498 clays and AC, either as a pre-treatment or as additives in the cement mix, can more effectively  
499 immobilize organic compounds in the cement matrix (Paria and Yuet, 2006). However, organic  
500 compounds have been found to retard the cement setting process by forming a protective layer  
501 around the cement grain, thus hindering the formation of calcium hydroxide (Sora *et al.*, 2005).

#### 502 **4.3.2 Sustainable solutions**

503 Despite cement being largely used in several *S/S* treatments, it is responsible for 5–8%  
504 of global anthropogenic CO<sub>2</sub> emissions and accounts for 12–15% of total industry energy use  
505 (Ali *et al.*, 2011; Scrivener and Kirkpatrick, 2008). This finding has prompted new research to  
506 investigate more environmentally friendly and sustainable materials for *S/S* applications. For

507 example, in recent years, research has focused on proposing a partial or total replacement of  
508 traditional cement with natural additives to treat contaminated sediments (Patmont *et al.*, 2015;  
509 Lofrano *et al.*, 2017). For example, a variety of waste shells, including eggshells, mussel shells,  
510 and oyster shells, were analysed by researchers to verify their efficacy to immobilise pollutants  
511 (in particular heavy metals) in contaminated soils (Islam *et al.*, 2017; Liu *et al.*, 2018).

512 Moreover, Paleologos *et al.* (2022) have proposed the results regarding the mechanical  
513 stabilisation of fine marine sediments with mixtures formed by cement partially substituted by  
514 mussel shell powder produced without calcination. From the findings of microstructural  
515 investigations and scanning electron microscopy (*SEM*) images, it is clear that shell powder is  
516 completely encapsulated in the cement-sediment matrix, acting as a binder due to the elongated  
517 shape of the mussel shell fabric. This microstructural feature of mussel shells enhances the  
518 electrolytic exchanges between sediments and cement, and thus increases the contact areas  
519 between the mineral particles promoting the chemical hydration reactions. Such peculiarity of  
520 mussel shells makes them a valuable substitute for cement in stabilisation of *DS*, and provides  
521 a viable alternative that can reduce the consumption of natural resources (such as crushed rock,  
522 sand and gravel, extracted through highly impactful quarrying or river exploitation activities)  
523 and lower the amount of binders used in traditional sediment stabilisation practice.

#### 524 **4.4 Electrochemical remediation**

525 The electrochemical remediation includes the passage of electric current between the  
526 cathode and anode rods inserted in the slurry of the *DS* (Pedersen *et al.*, 2015). The positively  
527 charged particles start moving towards the cathode, while the negatively charged particles  
528 move towards the anode due to the influence of generated electric field (Pal and Hogland, 2022;  
529 Pedersen *et al.*, 2015). However, as the fine-grained sediments have more affinity to adsorb  
530 metal ions on their surface, electrochemical remediation finds its utility for their remediation  
531 (Pal and Hogland, 2022; Peng *et al.*, 2009). The basic mechanisms involved in this remediation

532 include (i) electro-osmosis, (ii) electromigration, (iii) electrolysis, and (iv) electrophoresis,  
533 which can remove even soluble metal ions and ions bounded with sediment oxides, hydroxides,  
534 carbonates, nitrates, and cyanide (Peng *et al.*, 2009).

535 Furthermore, the influencing factors responsible for heavy metal extraction include  
536 agitation rate, sediment properties, moisture content, *OM*, current flow, and extraction duration  
537 (Pedersen *et al.*, 2015). It should be noted that the *pH* of the sediment slurry controls the  
538 electrochemical remediation, viz., if the *pH* of the slurry is basic, then the precipitation of metal  
539 ions forms hydroxides or oxy-hydroxides, whereas, in the case of acidic nature, the metal ions  
540 are more likely to get desorb or solubilize (Pal and Hogland, 2022; Pedersen *et al.*, 2015). Also,  
541 it should be noted that the extraction ability of heavy metals from the *DS* slurry can be enhanced  
542 by using desorbing agents, viz. acidification and surfactants, which has the ability to solubilize  
543 the metal oxides, nitrates, hydroxides, carbonates, etc. adsorbed on the sediment surface (Peng  
544 *et al.*, 2018). The electrochemical remediation process results in chemical transformations that  
545 change the accessibility and mobility of the toxic substances making them more hazardous for  
546 living organisms and making it necessary to perform toxicity analysis (Benamar *et al.*, 2019).  
547 It has been reported by earlier researchers that due to the low mobility of charged particles in  
548 the process of electric remediation, the effect of electrophoresis can be ignored. Thus, the action  
549 of electric migration and electro-osmosis is used for actual migration of heavy metal ions in  
550 soil pore water under the influence of an external direct current electric field (Han *et al.*, 2021).  
551 Therefore, it is highly recommended to conduct elaborate and extensive studies to optimize  
552 electrochemical techniques with desorbing agent modifications for heavy metals extraction.

#### 553 **4.5 Biological remediation**

554 The biological processes (read bioremediation) include the action of microorganisms  
555 or plants (viz., phytoremediation) for the remediation of contaminated *DS* by oxidation of the  
556 *PAHs*, hydrocarbons, and mineral oils, converting them into non-hazardous compounds (Feng

557 *et al.*, 2022). Bioremediation can be achieved due to naturally occurring indigenous microbes  
558 by introducing nutrients in the form of water-soluble, slow-release, and oleophilic fertilizers  
559 and oxygen (biostimulation) in the contaminated sediments or can also be achieved by the  
560 addition of alien microorganisms (bioaugmentation viz., external microorganisms, enzymes,  
561 nutrients, etc.) to the DS (Doni *et al.*, 2018; Maletić *et al.*, 2019). As the microorganisms  
562 available in sediments plays a major role in the biodegradation of the contaminants, this process  
563 is known as natural attenuation (Maletić *et al.*, 2019). However, the duration required for  
564 contaminant degradation is noticeably high, but this treatment presents a low impact on carbon  
565 footprint (Crocetti *et al.*, 2022). Phytoremediation comprises remediation by plants and the root  
566 colonizing microbes to degrade the toxic compounds to non-toxic metabolites and is effective  
567 for the immobilisation of Zn, Fe, Mn, Cd, etc. (Peng *et al.*, 2009). Unfortunately,  
568 bioremediation is less predictable than other processes (Maletić *et al.*, 2019). Therefore, more  
569 extensive research should be conducted considering different plants, microorganisms,  
570 enzymes, nutrients, and environmental conditions to establish the suitability and effectiveness  
571 of this method and proper guidelines and regulations.

572 The permissible limits of heavy metals and PAHs have been presented in *Table S2*,  
573 which might be achieved with the methodologies discussed above in *Section 4* of this paper.  
574 The regulatory conditions that necessitate to be achieved by allowing proper treatment would  
575 be helpful in promoting DS use as raw material for on-shore and off-shore applications.  
576 Furthermore, establishing of various sediment remediation techniques on the basis of their type  
577 and concentration of contaminants is a future scope of work.

## 578 **5 Utilization strategies for Dredged sediments**

579 The DS finds its utilization in soil filling, coastal nourishment, construction purposes,  
580 horticulture, forestry, agriculture, etc., a few of which have been discussed elaborately in the

581 following sections. Here, it should be noted that the *DS* should comply with pollutant-specific  
582 regulations before utilization.

### 583 **5.1 Agricultural applications**

584 The contaminated *DS* can be used for horticulture, forestry, and agricultural  
585 applications (Crocetti *et al.*, 2022; Rakshith and Singh, 2017). The presence of micro-nutrients  
586 (viz., Cu, Fe, Mn, Zn, etc.) and macro-nutrients (viz., C, Ca, K, N, P, Mg, etc.) induces inherent  
587 fertility in the *DS* (Renella, 2021). Furthermore, freshwater *DS* application to agricultural land  
588 increases the *OM* content, cation exchange capacity of the soil mass, improves soil structure,  
589 enhances water retention, and thus increases overall soil microbiological, chemical, and  
590 physical fertility, apart from improvement in sorption properties and nutrient concentrations  
591 (Leue and Lang, 2012; Renella, 2021). Kazberuk *et al.* (2021) performed a pot experiment on  
592 white mustard as a test plant to evaluate the possibility of using bottom sediments from  
593 reservoirs and rivers contaminated with heavy metals (viz., Zn, Cu, Cd, Pb) and its impact on  
594 soil and plants. The obtained results motivate further research as the bottom sediment added  
595 soil yield was higher than the control soil. Thus, there is a dire need for field-scale experiments  
596 to understand the behavior of various crops, contaminant transmission in food chain, etc., with  
597 the addition of *DS* having different concentrations of *OM*, organic carbon, nutrients, heavy  
598 metals, etc.

### 599 **5.2 Infrastructure development**

600 The transformation of *DS* into geomaterials is an attractive way to relieve the shortage of  
601 high-quality raw materials for various applications and projects, such as constructing coastal  
602 highways and manufacturing pavements (Couvidat *et al.*, 2016), road construction (Hussan et  
603 al., 2023), producing cleaner pervious concrete (Beddaa et al., 2023), constructing plant-  
604 growing substrate to cultivate lettuce (Ferrans et al., 2022), fill material (Wang *et al.*, 2018),  
605 bricks (Wang *et al.*, 2015), and breakwaters. The treatments techniques can be different and

606 vary depending on the target to be reached for the sediment reuse; for example: (i) chemically  
607 immobilize the contaminants, reducing the leachability and bioavailability, and (ii)  
608 mechanically stabilize the material for its reuse as new construction material. The reuse of  
609 contaminated *DS* would facilitate the recycling of dredged materials from local sources and  
610 save natural soil resources and transportation costs for construction, in line with the philosophy  
611 of the circular economy (Todaro *et al.*, 2016; Wang *et al.*, 2012). However, only 5% of the  
612 materials generated from recycling operations are currently used in public works (Wang *et al.*,  
613 2014). These data indicate that in the context of sustainable development, it is still necessary  
614 to further study solutions for recycling sediments as renewable geomaterials. CemShell-based  
615 solutions can provide the physics-based methodology for achieving the change of focus in  
616 relation to the management of *DS* and mussel shells (Paleologos *et al.*, 2022). In particular, the  
617 leaching test is one important aspect in the environmental assessment of the reuse options of  
618 treated sediments. The selection of an appropriate test or combination of tests (e.g., batch tests  
619 or column tests) is vital for predicting the long-term contaminants' release into the environment.  
620 Several authors have shown that to successfully transform a *DS* (i.e., a waste) into a geo-  
621 material, a multi-level testing program (e.g., with geotechnical and leaching tests) is required  
622 to investigate the effective recovery of treated sediments (Barjoveanu *et al.*, 2018; Todaro *et*  
623 *al.*, 2020).

624 In this context, it should be noted that the *DS* primarily consists of mineral, organic and  
625 liquid phases. It is well established that the presence of *OM* in sediments affects their  
626 engineering properties adversely (Benaissa *et al.*, 2016; Hamouche and Zentar, 2020a). It is  
627 worth mentioning here that the valorization of *DS* should be conducted considering the  
628 environmental, economic, geotechnical, and mechanical feasibility and sustainability. Also, it  
629 has been reported that the permissible limit of *OM* in road pavement material should be  $\approx 2\%$  -  
630  $4\%$ , while that for embankment material should be  $\approx 5\%$  -  $7\%$  (Hamouche and Zentar, 2020a).



631 The concern with the presence of *OM* is its decomposition with time, leading to an increase in  
632 the porosity and, thus, an increase in the compressibility, which is a controlling parameter for  
633 most of the infrastructure developments. Furthermore, *DS* utilization for infrastructure  
634 development, viz., road pavement, embankment, etc., could be an interesting and sustainable  
635 solution that covers sustainable development goals (*SDGs*), viz., *SDG-8*, *SDG-9*, *SDG-11*,  
636 *SDG-12*, *SDG-13*, *SDG-14* and *SDG-17* (Suedel *et al.*, 2022). Unfortunately, one of the major  
637 barriers is the perception of different stakeholders towards *DS* as a waste material that needs to  
638 be changed.

## 639 **6. Technology Readiness Level (*TRL*) and Circular Economy**

640 The technical maturity of any process/technique is assessed through the technology  
641 readiness level, *TRL*, a point-based framework system from concept to commercial use.  
642 Unfortunately, most configurations and processes used for the contaminated *DS* focusing  
643 circular economy objective is at low *TRL* and have only been tested in lab conditions (Crocetti  
644 *et al.*, 2022). Therefore, there is an urgent need to establish effective methodologies and  
645 upscaling the processes to pilot and field scale.

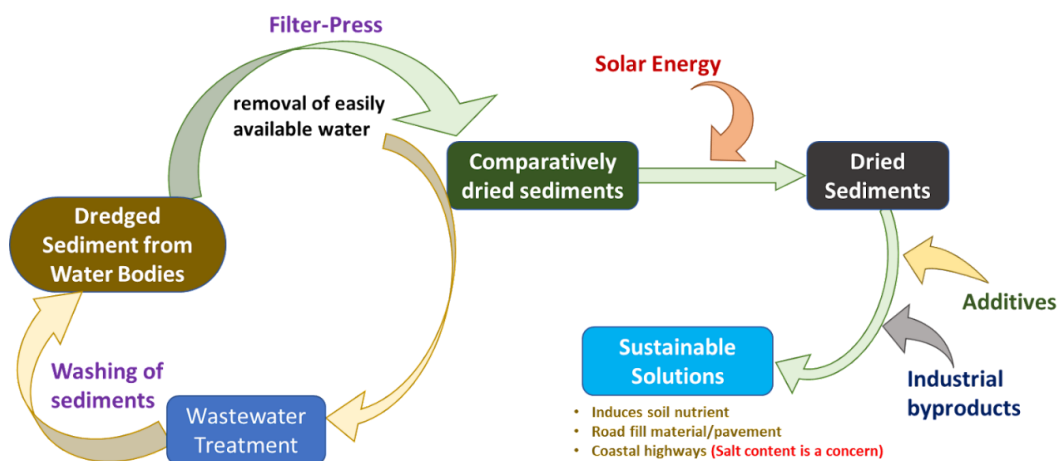
646 It should be noted that the major barrier behind the bulk utilization of *DS* include (i)  
647 policy/legal challenges at the local, national, and international levels, (ii) no guidelines against  
648 dumping of *DS* in the deep sea, which is catastrophic for marine flora and fauna and waste of  
649 money and material, (iii) lack of government initiative and policy framework in respect of  
650 promoting mission zero waste, and (iv) delay in approval from environmental agencies. Thus,  
651 there is an urgent requirement for the introduction of codes, standards, and guidelines for the  
652 utilization of *DS* as a secondary product that can reduce the use of raw materials and fill the  
653 gap created due to the scarcity of natural aggregates promoting circular economy and  
654 sustainable development.

655 Furthermore, the possible applications for *DS* utilization discussed should be modified,  
 656 bringing utilization of industrial by-products (*IBPs*) and other waste material replacing  
 657 conventional additives/modifiers, viz., cement, lime, sand, etc. (Singh et al., 2023; Singh and  
 658 Singh, 2023). It should be noted that this approach of using *IBPs* and waste will improve the  
 659 production chain, making it more environmentally friendly, sustainable, cost and energy  
 660 effective, apart from reducing carbon, water, and land footprints. Keeping in view of sediment  
 661 management and considering sustainable development, Figure 32 has been developed. From  
 662 the figure, it is clear that sustainable products can be generated by proper management of  
 663 sediments, and thus better *TRL*'s can be achieved. In this context, industries having *IBPs*,  
 664 government bodies/policymakers, researchers, and ports should come forward to make  
 665 sustainable management of *DS* a new reality.

666

667

668



669 **Figure 32.** Sediment management considering sustainable development and circular  
 670 economy

## 671 **7. Prospects and recommendations**

672 Based on the critical synthesis of the literature that deals with the (i) contamination  
673 assessment, (ii) testing and characterization methodology, (iii) remediation strategies, (iv)  
674 utilization strategies, and (v) *TRLs* and circular economy of the dredged sediments, the  
675 following generalized prospects and recommendations can be drawn:

- 676 (i) The major contaminants in the *DS* are heavy metals, *PAHs* and *PCBs*, which are  
677 case- and site-specific, and their concentration changes with prevailing  
678 environmental conditions and nearby contamination sources. In this context, the  
679 source, concentration level, and effect of emerging contaminants in the *DS* need to  
680 be further deepened case-by-case, which would help understand the primary and  
681 secondary sources of contaminants and address the most suitable remediation  
682 strategies.
- 683 (ii) The standard characterization and testing methodologies can fail when dealing with  
684 *DS* due to contaminants in the soil matrix and various pore fluid chemical  
685 compositions. In some cases, non-standard approaches need to be used to catch the  
686 complexity of the sediment matrix and fully understand the coupled related chemo-  
687 mechanical effects.
- 688 (iii) Based on a comparative procedure, the proposed framework appears transparent  
689 and interdisciplinary and represents a reliable way to select the most sustainable  
690 remediation alternative. The long-term performance is yet a matter of study for  
691 several remedial options (such as reactive capping and S/S). Moreover, evidence is  
692 provided about the need for further testing of these technologies at the real scale  
693 and to carry out a cost-benefit analysis considering the life-cycle impact analysis.

694 (iv) The possible utilization schemes of *DS* in agricultural applications and  
695 infrastructural development has been discussed. Also, the efforts tried by earlier  
696 researchers have been mentioned elaborately.

697 (v) The technology readiness level and circular economy perspective of *DS* utilization  
698 have been shown to open broad perspectives for scientists and policymakers to  
699 contribute and establish guidelines. Furthermore, such scientific advances will  
700 likely prompt a paradigm shift towards more sustainable industrial development.

701 Keeping this in view, the utilization of *DS* considering a circular economy perspective and  
702 *SDGs* needs to be established, which will further solve the issue being created due to the  
703 scarcity of natural aggregates and raw materials. Also, remediation techniques keeping in  
704 reference to type and concentration of the contaminants needs to be established. Furthermore,  
705 the local government should promote *DS* utilization by subsidizing transport facilities, creating  
706 a flexible licensing system for *DS* processing, policies to design life cycle assessments, and  
707 compulsory use of *DS* as secondary material.

708

### 709 **Acknowledgments**

710 Part of the research has brought about the filing of patent application (number IT  
711 102021000025103, title: “Stabilisation method of marine clays by shells and cements”) by  
712 POLIBA and ETH Zurich.

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1 **Characterization, Remediation and Valorization of Contaminated Sediments-A Critical**  
2 **review**

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63 **Number of Table:** 3

64 **Number of Figures:** 2

65 **Word count in main body of manuscript:** 7375 words (excluding Abstract, Tables, Figures,  
66 Acknowledgements and Reference section)

67 Submitted for peer review and possible publication in the *Journal of Environmental*  
68 *Geotechnics*

69 Date of revised manuscript Submission: 11<sup>th</sup> July 2023

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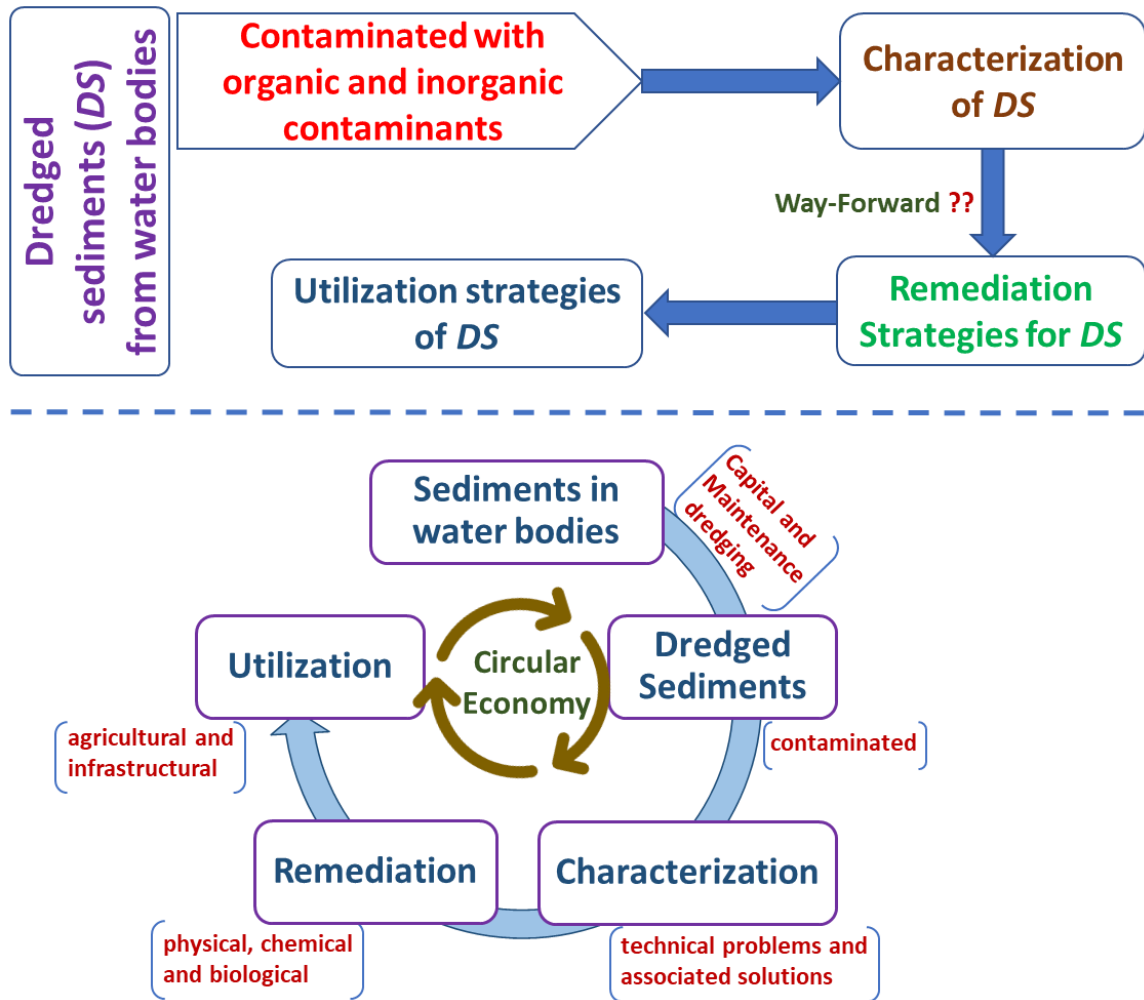
71 **Abstract**

72           The constraints associated with the availability of huge amounts of natural resources  
73 for infrastructure and agricultural development calls for the reuse and recycling of  
74 anthropogenically created geomaterials, which is in line with the UN Sustainable Development  
75 Goals. In this context, valorization of dredged sediments (*DS*), obtained from water bodies such  
76 as rivers, lakes, oceans, etc., as a resource material is worth considering. Unfortunately, *DS*  
77 might be contaminated and exhibit a higher moisture-holding capacity due to higher organic  
78 matter and clay minerals/colloids. These attributes pose a serious question towards dumping of  
79 the *DS* in the deep sea (in the case of marine sediments), a practice which though prevails  
80 presently but endangers marine life. Hence, the way forward would be to characterize them  
81 holistically, followed by adequate treatment to make them ecologically synergetic before  
82 developing a strategy for their valorization. In this regard, many studies have been focused on  
83 the characterization and treatment of *DS* to make them environmentally safe manmade  
84 resource. With this in view, a critical synthesis of the published literature pertaining to the (i)  
85 characterization, (ii) treatment, remediation, and immobilization of contaminants, and (iii)  
86 utilization of *DS* has been conducted, and the salient findings are presented in this paper. Based  
87 on this study, it was observed that the *DS* acts as a sink for emerging contaminants for which  
88 no remediation strategies are available. Moreover, the study highlighted the lacuna in upscaling  
89 the existing treatment and stabilization techniques to field conditions while highlighting the  
90 concept of circular economy.

91 **Keywords:** *sustainable development goals; dredged sediments; contamination; toxicity;*  
92 *remediation; utilization*

93 **Graphical Abstract**

94



95

96 **Highlights**

- 97
- Source, concentration and effect of contaminants.
- 98
- Sustainable development and circular economy perspective of dredged sediments with
- 99
- Technology Readiness Levels.
- 100
- Strategies for remediation of contaminants and utilization of dredged sediments.
- 101
- Prospects and recommendations considering policy and guideline issues.

102

## 103 **1 Introduction**

104       The increasing human activities and natural conditions are responsible for the  
105 contamination of water bodies, viz., harbors, ports, estuaries, rivers, lakes, etc. (Akcil *et al.*,  
106 2015). In the realm of the dredging industry (capital and maintenance), the dredged sediments  
107 (*DS*) act like a by-product that has the potential to be utilized to replace natural mineral  
108 aggregates (Achour *et al.*, 2014; Loudini *et al.*, 2020). Furthermore, the emergent demand for  
109 construction materials in the infrastructure sector and the environmental constraints on  
110 opening new quarries create an unavoidable need for unconventional geomaterials like *DS*.

111       The perspective towards dredged material has changed over the past few years from a  
112 waste to a resource, and the utilization of the same is being explored considering the circular  
113 economy and sustainable development (Gebert and Groengroeft, 2020; Mehdizadeh *et al.*,  
114 2021). However, *DS* are complex materials due to the presence of salts, organic matter (*OM*),  
115 and contaminants (Rakshith and Singh, 2017). The major contaminants in *DS* could be  
116 classified as (i) inorganic pollutants (potentially toxic elements, viz., zinc, copper, iron,  
117 manganese, cadmium, lead, etc.) and (ii) organic pollutants [viz., polycyclic aromatic  
118 hydrocarbons (*PAHs*), polychlorinated biphenyl (*PCBs*), etc.]. Also, *OM* impacts the  
119 geomechanical performance of sediments due to the increase in voids ratio induced by *OM*  
120 decomposition (Hamouche and Zentar, 2020a, 2020b). In this context, the assessment of the  
121 effects of *OM* on the geotechnical parameters and their evolution in conjunction with *OM*  
122 transformations is one of the relevant aspects to be faced within engineering practice.  
123 However, their potential impact on the environment needs also to be established to design  
124 proper treatments if necessary.

125       Considering the high amounts of *DS* produced worldwide mainly from the marine  
126 environment and the legal constraints associated with their management, their direct disposal  
127 in confined disposal facilities/landfilling is no longer economically, socially, and



128 environmentally feasible (Mehdizadeh *et al.*, 2021; Pal and Hogland, 2022). Further, hydraulic  
129 fills have been utilized in many land reclamation projects, for example, Kansai International  
130 Airport in Osaka Bay, Changi Airport Singapore, etc., were constructed on a hydraulic fill  
131 made of *DS* and soil, which not only allowed for the expansion of the airport but also  
132 contributed to reducing the amount of waste sent to landfills (Douglas and Lawson, 2003;  
133 Matsui, 1996). However, it should be noted that the hydraulic fills are not always the best  
134 option for managing the contaminated *DS*. The selection of a management option is based on  
135 various factors including level of contamination, volume of sediments, local regulations etc.  
136 Therefore, a more sustainable fate for *DS* prompts novel research and management challenges  
137 for researchers, management, policymakers, and administrators (Crocetti *et al.*, 2022; Loudini  
138 *et al.*, 2020). Furthermore, the potential utilization of *DS* in infrastructural and agricultural  
139 applications has been tried by earlier researchers (Crocetti *et al.*, 2022; Hamouche and Zentar,  
140 2020a; Rakshith and Singh, 2017). However, *DS* toxicity and contamination level being case-  
141 and site-specific, more extensive studies focusing on its utilization schemes need to be  
142 performed by the research communities.

143 From the existing literature, it was realized that the number of publications considering  
144 the valorization of *DS* with a focus on sustainable development is less, whereas that for the  
145 circular economy perspective is almost negligible. The reviews conducted till date on *DS* are  
146 limited to either contamination, or management, or application aspects, which does not give a  
147 broader perspective about contaminated sediments. Keeping in view of these mentioned  
148 findings, this paper synthesizes the recent developments in the field of contamination  
149 associated with the dredged material, their characterization, followed by remediation and  
150 utilization strategies considering the sustainable development and circular economy aspects.  
151 Furthermore, the necessities associated with the policy and guidelines have been critically  
152 evaluated, and a brief account of the same has been discussed in the following sections.

## 153 **2 Source, concentrations, and effects of emerging contaminants in dredged sediments**

154 Emerging contaminants (*ECs*) are ‘any synthetic or naturally occurring chemical or any  
155 microorganism that is not commonly monitored in the environment, but has the potential to  
156 enter the environment and cause known or suspected adverse ecological and/or human/aquatic  
157 life/wildlife health effects’ (Smital, 2008). The *ECs* need not been found in the environment in  
158 the recent past but may persist over the decades in small concentrations (i.e.,  $\mu\text{g/L}$  and  $\text{ng/L}$ )  
159 and found to be of concern due to (i) exponential growth in the utilization of products  
160 contributing to them, and (ii) increase in their adverse effects on the environment and life on  
161 the planet. For instance, the per- and polyfluoroalkyl substances (*PFAS*) based products such  
162 as paints, sealants, water-resistant clothing, grease-resistant papers, fast food containers, and  
163 nonstick cookware are being used since the 1950s, but widely found in different environmental  
164 systems after development and improvement in the sensitivity of mass spectrometers in 1980s,  
165 which subsequently led to their classification as *ECs* in early 2000s (Richardson and Kimura,  
166 2017). Hence, the *ECs* are also known as ‘chemical of emerging concerns’ or ‘contaminants of  
167 emerging concerns’ (Rosenfeld and Feng, 2011).

168 The sources of *ECs* in *DS* can be classified as primary and secondary. The primary sources can  
169 be defined as the initial point of contact wherein the *ECs* are used in the manufacturing of the  
170 products to attain the desired properties. The primary sources of *ECs* include pharmaceutical  
171 and personal care products, biocides (including agricultural and plant protection products),  
172 disinfection by-products, industrial chemicals (viz., lubricants, flame retardants, gasoline,  
173 antimicrobial agents, surfactants, food additives, and plasticizers), bioterrorism and sabotage  
174 agents, algal toxins, etc. (Barber, 2014; Rosenfeld and Feng, 2011).

175 The secondary sources of *ECs* include industrial sludges and wastewater, surface water  
176 bodies, municipal solid waste, industrial by-products and soils contaminated with industrial  
177 discharges and chemicals (refer to *Figure S1*). Furthermore, micro(nano)plastics can also be

178 considered as the potential secondary source of *ECs* because they can fragment, degrade and  
179 leach one or more of the *ECs*, such as persistent organic pollutants (Goli *et al.*, 2021; O’Kelly  
180 *et al.*, 2021). The primary sources majorly contaminate the *DS* through their deposition,  
181 leaching, and sorption, while the secondary sources would contaminate by the sorption  
182 mechanism. However, the dominant mechanisms which contribute to *ECs* in *DS* would  
183 completely depend on the characteristics of the latter and environmental conditions to which  
184 the primary sources are exposed.

185         The contamination of *DS* through primary sources can be more often observed in the  
186 developing and under-developed countries where the guidelines for liquid and solid waste  
187 collection, transportation, and treatment are not enforced strictly or not available. Unlike the  
188 primary sources, secondary sources of *ECs* are the major pathways for contamination of the  
189 *DS* in all countries due to the fact that the removal of the *ECs* is not the primary motive of the  
190 domestic and industrial wastewater treatment plants, municipal solid waste leachates and  
191 sludges up to the recent past.

192         Furthermore, the determination of concentrations of *ECs* in *DS* is mostly limited to a  
193 few compounds based on *PAHs* and *PCBs* because these are major contaminants emitted during  
194 the vehicular and vessel movements that are essential for offshore transportation, recreational  
195 activities and nearby industrial activities (Kafilzadeh, 2015; Norén *et al.*, 2020) (refer to *Table*  
196 *1*). The *ECs* contamination, their possible sources, the source of *DS*, and the detection  
197 techniques studied by earlier researchers have been presented in the *Table 1*.

198

199

**Table 1.** Summary of the studies conducted on the concentrations of *ECs* in *DS*

Reference	Study area	Source of <i>DS</i>	<i>ECs</i> detected with concentration	Possible source of <i>ECs</i>	Detection techniques
Torres <i>et al.</i> (2009)	Port of Santos, Brazil	Marine sediments (18 samples from dredged areas and disposal sites, 4 samples from hopper dredge)	<i>PAH</i> (27.86 to 679.35 µg/kg); <i>PCB</i> (0.17 to 12.33 µg/kg)	Emissions and activities of steel plant and industrial complex	Gas Chromatography/Mass Spectroscopy ( <i>GC/MS</i> )
Rocha <i>et al.</i> (2011)	Porto region, Portugal	4 river estuary and 2 marine beach sediments	<i>PAH</i> [Estuary (98.40 to 156.50 µg/kg dw); Marine sediment (52.00 to 54.80 µg/kg dw)]	-	<i>GC/MS</i>
Tavakoly Sany <i>et al.</i> (2014)	Klang strait, Malaysia	Coastal sediment	16 compounds of <i>PAHs</i> (994.02±918.10 µg/ kg dw)	Contamination due to cargo transport, petrogenic spillage and pyrogenetic combustion	<i>GC/MS</i>
Kafilzadeh (2015)	Soltan Abad river, Iran	River sediment (4 sampling locations at a depth of 5 cm from the bed)	16 compounds of <i>PAHs</i> (180.30 to 504.00 µg/kg)	Pyrogenic combustion and petrogenic spillage	Gas Chromatography/Flame Ionisation Detection ( <i>GC/FID</i> )
Couvidat <i>et al.</i> (2018)	Port in the south of France	Harbour sea bed (Top 50-80 cm)	16 compounds of <i>PAHs</i> (62.18-62.40 mg/kg) 7 compounds of <i>PCBs</i> (0.96-0.97 mg/kg) 3 compounds of Organotin compounds (65.50 mg/kg)	Extensive anthropogenic activity for centuries and contamination due to industrial activity	<i>GC/MS</i> and low-resolution <i>MS</i>
Shilla and Routh (2018)	Rufiji Estuary, Tanzania	River sediment (top 1-2 cm sediment was scrapped on South, middle and north parts of Rufiji Delta)	19 compounds of <i>PAHs</i> (128 to 377 µg/kg)	Petrogenic spillage and pyrogenic combustion of coal and biomass (mainly grass and wood)	<i>GC/MS</i>
Norén <i>et al.</i> (2020)	Two ports, three marina and one waterway leading to the marina in Sweden	Marine environment	Tributyltin: ports (150±30 mg/kg); marina (50±50 and 310±240 mg/kg); waterway (70±60 mg/kg)	Pollutants released by recreational and public transport boats, cargo vessels. Effluents from Cu production, wastewater treatment, battery production industries and shipyards.	-

200

### 201 **3 Geotechnical characterization: technical problems and adopted solutions**

202 The characterization of contaminated sediments to address environmental issues related to  
203 the remediation of polluted areas is aimed to build the so-called Conceptual Design Site Model  
204 (*CDSM*). The *CDSM*, includes the most relevant site features (i.e., water, soil/sediment and  
205 biota properties, together with land waterway use) as well as the processes ongoing within the  
206 system. Stemming from the traditional Conceptual Site Model (*CSM*), the *CDSM* is originally  
207 meant to be an updated model including chemical, geo-hydro-mechanical and environmental  
208 engineering knowledge about the processes ongoing within the relevant volume of the system.  
209 It supports a more sustainable choice of remedial strategies since it is capable of taking account  
210 of at least two (Environment and Engineering) of the *four-E* (Environment, Economy, Equity  
211 and Engineering) criteria of the multi-dimensional approach towards *sustainability* (Basu *et*  
212 *al.*, 2015). Moreover, being centered on the knowledge of processes, it can more efficiently  
213 support the first predictions of the system evolution, both in the short and in the long term, that  
214 would accompany the remediation phase (Vitone *et al.*, 2020).

215 It follows that it becomes a strategic tool to address both the selection of sustainable  
216 remedial strategies and the technology screening phase of contamination (Reible, 2014;  
217 USEPA, 2019). In this model, the geo-hydro-mechanical characterization of the sediments  
218 provide geotechnical parameters which have a direct effect on the feasibility of all remedial  
219 technologies and supports the predictions of the *DS* behaviour before and after treatment  
220 (Adamo *et al.*, 2018; Roque *et al.*, 2022; Vitone, 2020).

221 For example, in situ capping is a remediation option that can be selected and designed only  
222 after a site characterisation which includes geotechnical considerations (Vitone *et al.*, 2016).  
223 Usually, contaminated sediments are predominantly fine-grained and often have high water  
224 content and compressibility, and low shear strength. Cap stability and settlement due to  
225 consolidation are geotechnical issues that may be important for cap effectiveness. After

226 placement of a cap, consolidation of both the underlying contaminated sediment and the cap  
227 layer usually occurs (Reible *et al.*, 2014). The consolidation of the cap is typically small. On  
228 the other hand, the consolidation of the underlying contaminated sediments may be significant,  
229 especially when dealing with soft soils, and expresses porewater pressure from the  
230 contaminated layer up into the cap. Moreover, the fluid expelled during the consolidation  
231 process should be evaluated for the investigation of a contaminated marine site. In fact,  
232 contaminant migration can change sediment properties (e.g., consistency limits), influencing  
233 capping design (Erten *et al.*, 2011). The impact of these processes depends on the sediment  
234 geotechnical properties that should be known for an efficient design of capping (Reible, 2014).

235 However, marine sediments may contain *OM*, shells, microfossils and diatoms, salts, heavy  
236 metals, and organic pollutants, which may induce some bias in the measurement and  
237 classification of fine-grained soils, which makes geotechnical characterization quite  
238 challenging. The framework defined for normally-consolidated natural clays and the laboratory  
239 standards does not focus on soils containing sources of complexity such as those typical of  
240 contaminated marine sediments. As reported in the following paragraphs, some novelties need  
241 to be introduced in the phase of soil testing and data analysis to accurately measure the state,  
242 physical and mechanical properties of contaminated sediments.

### 243 **3.1 Impact of organic matter, heavy metals, fossils, and other pollutants**

244 *DS* from natural environments may be characterized by a significant content of buried *OM*  
245 coming from the biosynthesis of organisms existing in the water column. Moreover, in shallow  
246 marine basins, near the coast, the terrigenous contribution of *OM* (allochthonous *OM*) might  
247 occur. Organic particles can be adsorbed by negatively charged mineral surfaces and promote  
248 the aggregation of clay-size particles to form a more open fabric. If *OM* content is high, soils  
249 may be characterized by unusually high-water contents, plasticity, and activity index, with  
250 exceptionally low wet bulk densities and high compressibility (Levesque *et al.* 2007).

251 Furthermore, non-decomposed organic substances and microbial populations will have a  
252 binding effect on the soil particles (Bobet *et al.*, 2011), which reduces the soil plasticity and  
253 activity indexes of clays (Sollecito *et al.*, 2021).

254 Recent research by Muththalib (2020) and Muththalib and Baudet (2019) on kaolin,  
255 bentonite, mixtures of kaolin and bentonite, illite rich Lucera clay, and submarine sediments  
256 from the Port of Taranto (Mar Piccolo) showed the effect of heavy metal contamination on  
257 their physical and mechanical properties. In *Figure S2*, the plasticity of kaolin is seen to  
258 increase with heavy metal contamination, with a reverse effect observed in bentonite and hardly  
259 noticeable effects in the illite rich Lucera clay. The reason behind this observation might be the  
260 difference in their *pH* and electrical conductivity, and the alterations in these properties due to  
261 the presence of heavy metals (*viz.*, Cu, Zn, Pb, etc.) in soluble form.

262 *Figure S3* summarizes the variation in the plasticity of different clays with the presence of  
263 heavy metal contamination. Pure kaolin, pure bentonite, kaolin-bentonite mixtures, submarine  
264 sediments from the Port of Taranto and an illite-smectite rich clay (Lucera) were tested with  
265 salt or heavy metals used single or combined. Copper, Lead and Zinc were chosen at  
266 concentrations of 1000 ppm unless specified. In *Figure S3*, the symbol shapes characterize the  
267 soil tested while the colour represents the added salt or metal(s).

268 The thermogravimetry tests could be coupled to geotechnical testing to explore the sediment  
269 skeleton's nature and its *OM* content, based on the main thermal reactions occurring within  
270 different temperature ranges (Sollecito *et al.*, 2021). The *DS* having a substantial quantity of  
271 *OM* should not be oven dried before testing for the Atterberg limit determination because the  
272 liquid limit decreases when the organic soil is oven-dried before testing (ASTM D2487-ASTM,  
273 2011). Furthermore, the sieving procedure at 425- $\mu\text{m}$  (No. 40) sieve required for the

274 preparation of material for the Atterberg limit determination (ASTM D4318-ASTM, 2017),  
275 may remove the organic components and alter the sediment plasticity (Roque *et al.*, 2022).

276 The testing on marine sediments may be further compounded by the widespread presence  
277 of lapideous elements and fragments of shells, mussels, fossils, and diatoms, whose dimensions  
278 could vary from some centimeters to a few micrometers. The presence of these elements in the  
279 soil matrix has been found to alter the soil fractions of sediments retrieved in the Mar Piccolo,  
280 a highly polluted marine basin in southern Italy (Cotecchia *et al.*, 2021).

281 Also, inclusions only visible at the micro-scale can introduce some bias in the sediment  
282 characterization. The presence of microfossils and diatoms (*Figure S4*), of high intra-skeletal  
283 voids space can provide an apparent increase of the soil plasticity and activity indexes, as well  
284 as the soil compressibility, irrespective of the clay fraction size and typology (Caicedo *et al.*,  
285 2018; Sollecito *et al.*, 2021).

286 The effect of the remediation treatment depends on several factors. For example, in the case  
287 of *ex situ* stabilization/solidification treatments, the quantity of additive, the curing time,  
288 composition and physical properties of the sediments and water chemistry. In particular, the  
289 contaminants can interfere with the sediment properties (e.g., consistency limits)  
290 compromising the effectiveness of the stabilization. It follows that the optimization of the  
291 treatment depends on the type of contaminants, soil physical properties, composition, and the  
292 required performance (Todaro *et al.*, 2020; Vitone *et al.*, 2020; Wang *et al.*, 2018).

### 293 **3.2 Effect of pore water salinity**

294 The effects of chemo-mechanical coupling in soils are usually interpreted according to the  
295 Gouy-Chapman diffuse double layer (*DDL*) theory (Chapman, 1913; Gouy, 1910). According  
296 to this theory, the thickness of the *DDL* in clays decreases when either the pore water ion  
297 concentration or the cation valence increases and the dielectric constant decreases, as for clays,



298 including high concentrations of salts, metal ions, and organic pollutants. These conditions  
299 favor clay particle flocculation and prompt significant variations of the soil index properties,  
300 mechanical parameter values, and testing procedures, with respect to those consisting  
301 uncontaminated pore solution, and thus the salt concentration in the pore-fluid should also be  
302 considered (Mitchell and Soga, 2005; Sollecito *et al.*, 2019a). A reduction of liquid limit,  $w_L$ ,  
303 and compression index,  $C_c$ , is generally recorded in active clays when pore fluid salinity  
304 increases (Di Maio *et al.*, 2004). Special consideration should therefore be paid to sediments  
305 from the marine environment where high soluble salt concentrations are present, especially  
306 when remediation strategies such as washing and decontamination must be undertaken since  
307 they may change the pore fluid chemistry and, in turn, the soil behavior.

308 To take into account the presence of salts, the water content data obtained through oven-  
309 drying (ASTM D2216-ASTM, 2019) should be corrected for the salinity values (ASTM  
310 D4542-ASTM, 2015; Sollecito *et al.*, 2021). Furthermore, the use of a fluid with the same  
311 salinity of the pore water for laboratory experiments is recommended by several earlier  
312 researchers (Di Maio *et al.*, 2004; Baudet and Ho, 2004; Sollecito *et al.*, 2019a). This is the  
313 case of the preparation of the material for the liquid limit determination (Di Maio *et al.*, 2004;  
314 Sollecito *et al.*, 2019a); the preparation of reconstituted samples (Baudet and Ho, 2004); and  
315 the filling of oedometer or direct shear tests cells, where differences in the fluid composition  
316 may induce the flow of water or ions through the soil. The use of water with salt concentration  
317 same as the sample pore fluid is also recommended to apply the cell pressure during triaxial  
318 tests to avoid building up osmotic pressures across the sample membrane because of  
319 differences in salinity (Baudet and Ho, 2004).

### 320 **3.3 Use of statistical techniques for integrated sediment characterization**

321 As previously reported, the characterization of contaminated sediments requires assessing  
322 several variables, including contaminant source, contaminant type, the sedimentary up to even

323 the hydrologic environment, or natural features such as sediment grain-size distribution,  
324 composition, the effect of transportation (including here the cross-shore and long-shore  
325 transport of sediment). Due to the potentially high costs associated with the management of  
326 contaminated sediments and their remediation process, the assessment of the degree of  
327 contamination becomes paramount as (i) the inaccurate determination can result in wasting of  
328 considerable financial resources related with unnecessary treatment measures, and (ii) poses  
329 both ecological and human health risks. In this sense, the characterization phase and  
330 determination of the contamination degree becomes critical and the subject of the intense  
331 inspection.

332 During the characterization phase, a deterministic approach in analyzing the parameters  
333 selected to describe the ecosystem can surely provide a great deal of information and drive the  
334 remediation strategies (Cotecchia *et al.*, 2021). Nevertheless, the inspection of the complex  
335 dataset that is generated from the investigation campaign cannot easily allow the understanding  
336 of the factors of key relevance, which impact and control the spatial and temporal distribution  
337 of contaminants in the ecosystem. Aiming to address such complexity, environmental scientists  
338 have started employing multivariate statistical approaches that constitute an advantage in the  
339 assessment and modelling of contamination patterns of highly contaminated areas on a large  
340 scale and thus could contribute to effective and economical monitoring of their quality.

341 The statistical techniques have been widely exploited in the literature since they support the  
342 generation of spatial pollution maps and identify potential interaction stressors in contaminated  
343 areas (Hopke 2015; Mali *et al.*, 2016, 2017, 2022). Successful examples can be reported, such  
344 as the characterization of the *EC* distribution in the sediments in one of the most polluted  
345 Mediterranean coastal basins, Mar Piccolo (Mali *et al.*, 2017); or the influence of Sarno river  
346 discharges onto Gulf of Naples, a marine basin subjected to a highly anthropized coastal area  
347 (Mali *et al.*, 2022).

348 It has been reported by Mali *et al.* (2017) that the combination of principal component  
349 analysis (*PCA*) and analysis of variance (*ANOVA*) revealed synergistic effects of independent  
350 factors such as total organic carbon (*TOC*) and Grain Size and allows to understand the *TOC*  
351 concentration resulted to be dominant conditioning factor with respect to granulometry.

352 Therefore, the characterization of complex matrices such as marine and harbor sediments  
353 needs advanced tools that are able to investigate the complex pattern that arises from the  
354 superposition of natural and anthropogenic processes and from multiple factors acting  
355 simultaneously on a local scale.

## 356 **4 Remediation strategies for contaminated sediments**

### 357 **4.1 Technologies overview**

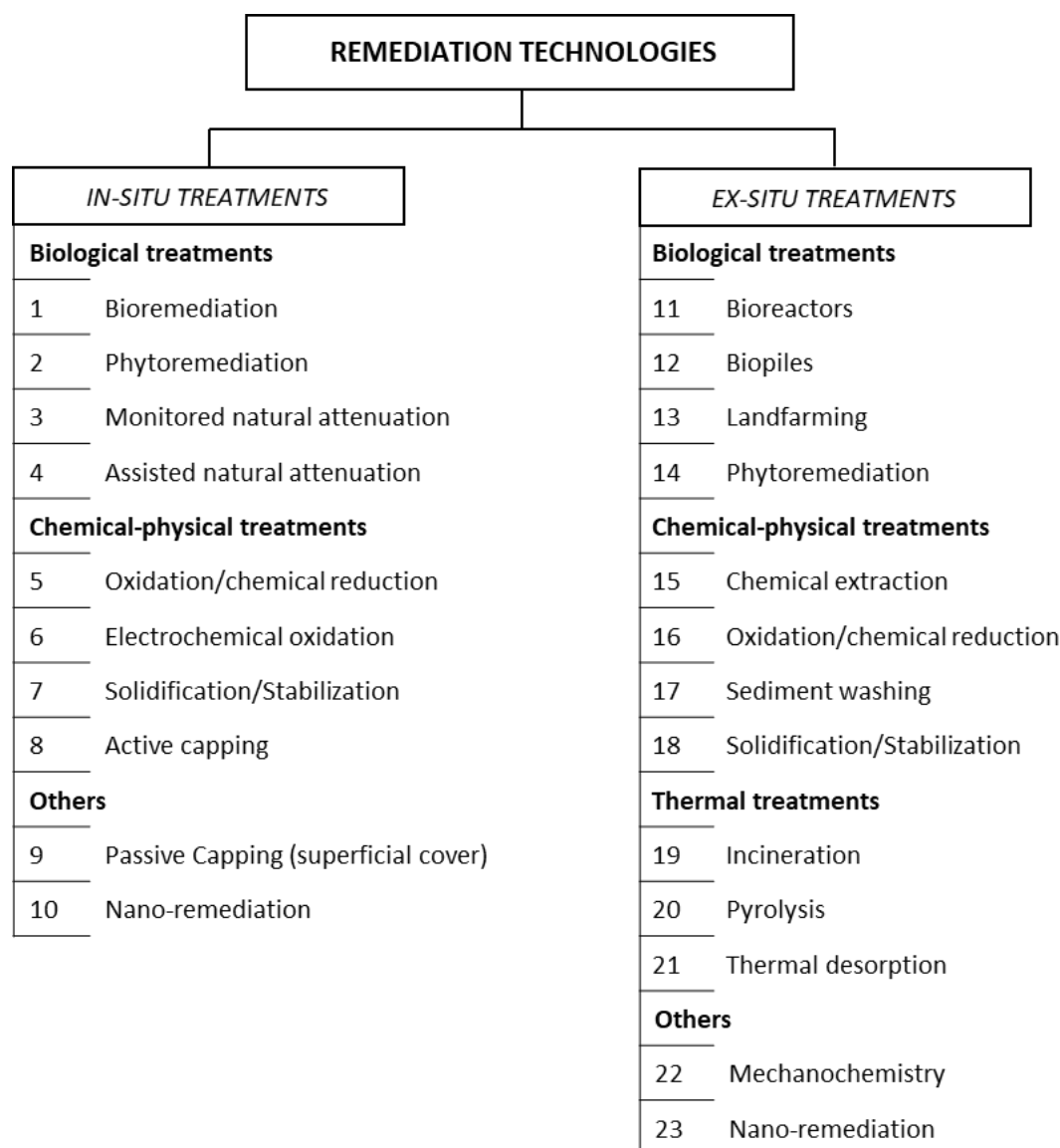
358 The remediation of contaminated marine sediments is more complex than managing  
359 contaminated soil or groundwater sites. Hence, the choice of approach(es) is generally broad  
360 and complex, frequently conflicting, and often controversial (Todaro *et al.*, 2018). As a result,  
361 the management and remediation of contaminated sediments is a significant issue faced by  
362 environmental policymakers, scientists, and engineers (Labianca *et al.*, 2021).

363 Sediment remediation techniques are commonly classified as in-situ (i.e., treatments  
364 operating without sediment dredging) and ex-situ (i.e., treatments including sediment  
365 dredging). In-situ technologies allow soil and sediment to be treated without being excavated  
366 and transported, with potentially significant savings. However, in-situ treatments generally  
367 require a longer remediation time (Lofrano *et al.*, 2017). Instead, ex-situ treatments require the  
368 dredging of sediments, leading to increased costs and engineering for equipment, possible  
369 permitting, and material handling/worker exposure considerations. However, the main  
370 advantage of ex-situ treatment is that it requires shorter periods than in-situ treatment, and there

371 is more certainty about the uniformity of treatment because of the ability to screen and mix the  
372 sediments (Zhang *et al.*, 2021).

373 Moreover, treatment methods can be categorized into three major groups: (a)  
374 physical/chemical, (b) biological, and (c) thermal (Todaro *et al.*, 2016; De Gisi *et al.*, 2017a).  
375 Furthermore, the classification of the remediation technologies has been proposed in *Figure 1*.  
376 In *Figure 1*, various in-situ and ex-situ treatment techniques for sediment remediation have  
377 been presented, which need to be adopted based on the contamination level and type and thus  
378 need further extensive research in this context. Furthermore, this paper focuses on the more  
379 competitive technologies in the direction of sustainable development.

380



381

382 **Figure 1.** Sediments remediation technologies for in-situ and ex-situ treatments383 **4.2 In situ capping**

384 In situ remedial alternatives generally involve: (i) natural attenuation, which is based  
 385 on the assumption that, although sediments pose some risk, it is low enough that natural  
 386 processes can reduce risk over time in a reasonably safe manner (De Gisi *et al.*, 2017b); (ii) in  
 387 situ containment and treatment via capping, in which contaminated sediment is physically and

388 chemically isolated from aquatic ecosystems or the contaminants in sediment are sequestered  
389 and degraded (Todaro *et al.*, 2018).

390 Capping is the process of placing a layer of clean materials over contaminated  
391 sediments to isolate the contaminant from the overlying water column and biota, to reduce  
392 contaminant flux into the biologically active portion of the sediment, and to create new habitats  
393 for aquatic organisms (Reible, 2014). Conventional (passive) caps consist of placing a layer of  
394 clean neutral materials (such as sand, silt, clay, and crushed rock debris) that rely on  
395 containment, rather than treatment, of contaminated sediment. The cap may also include  
396 geotextiles to aid in layer separation or geotechnical stability, amendments (that is of  
397 chemically reactive materials) to enhance protectiveness, or additional layers to armor and  
398 maintain its integrity or enhance its habitat characteristics. When these amendments are added  
399 to cap material, the technology is called an “Active Cap” (or “Reactive Cap”), and the  
400 amendments enhance the performance of the cap material. The use of chemically reactive  
401 materials allows sequestering and/or degrading of sediment contaminants, reducing their  
402 mobility, toxicity, and bioavailability, performing both containment and treatment of  
403 contaminated sediment. The comparison between passive capping and active capping is listed  
404 in *Table S1*. Active/Reactive Cap presents several advantages; however, the capping  
405 technology selection depends on site characteristics (e.g., contamination levels and  
406 geotechnical properties of sediments).

407 Sand capping has been widely investigated, being mostly utilized for large availability  
408 and ease of placement of sand (Jiao *et al.*, 2020). In Bortone *et al.* (2018) a sand cap was  
409 investigated to reduce the exposure of the aquatic ecosystem to *PCB*-contaminated sediments  
410 in Lake Hartwell, US. Specifically, it was demonstrated that *PCB* transport was extremely  
411 dependent on the cap characteristics, and that the cap thickness could be reduced at 20 cm-  
412 thick by using a high sorbent cap. In the study by Meric *et al.* (2014), a 7.5 cm thick sand cap

413 reduced the bioavailability of *PCBs* by a factor of 100 compared to the no-capping scenario,  
414 but it did not influence the bioavailability of naphthalene.

415 However, a passive cap might partially allow dissolved contaminants into the overlying  
416 water column and consequently still pose a risk to the benthic environment and the trophic  
417 chain. In these cases, reactive materials can be used. Murphy and Lowry (2004) demonstrated  
418 that a thin layer of the adsorptive amendment (i.e., activated carbon) could have a more  
419 performing pollution containment capacity for *PCBs*, equal to over 100 times sand adsorption  
420 effectiveness. Other reactive amendments (such as calcite, zeolite, apatite, organoclay, and  
421 biopolymers) can also sequester or degrade a variety of contaminants and control their mobility  
422 to the water column (*Table 2*).

423 **Table 2.** Summary of amendments used to treat organic and inorganic contaminants

Function	Amendment	Contaminant targeted	Reference
Sequestering	Activated Carbon, AC	Organics/inorganic	Choi (2018); Silvani <i>et al.</i> (2017)
	Apatites	Metals	Knox <i>et al.</i> (2012); Xing <i>et al.</i> (2016); Zhang <i>et al.</i> (2016)
	Bauxite	Metals	Taneez <i>et al.</i> (2018)
	Biochars	Organics/inorganic	Bianco <i>et al.</i> (2021); Janssen and Beckingham (2013); Ting <i>et al.</i> (2020)
	Organoclays	Organics/inorganic	Erten <i>et al.</i> (2012); Olsta (2010); Pagnozzi <i>et al.</i> (2020)
	Zeolites	Metals	Gu <i>et al.</i> (2019); Kang and Park (2015)
Degrading	Bioremediation agents	Organics	Atashgahi <i>et al.</i> (2014); Sun <i>et al.</i> (2010); Wang <i>et al.</i> (2014)
	Zero-Valent Iron, ZVI	Organics	Chapman <i>et al.</i> (2020); Hu <i>et al.</i> (2020)

424

425 In this regard, an innovative use of reactive capping entails encapsulation of the reactive  
426 amendments between two geotextile layers by further reducing reactive layer thickness to about  
427 1 cm (Meric *et al.*, 2014; Bortone *et al.*, 2020). It includes additional benefits such as uniform  
428 consolidation and defined mass per area. Alternatively, innovative reactive capping is  
429 represented by reactive granular materials where the reactive amendments cover an inner inert  
430 core more stable and slough off the aggregate during hydration, enabling mixing with the  
431 contaminated sediment.

### 432 **4.3 Stabilisation/Solidification (S/S)**

433 The immobilization and stabilisation of the metals and other contaminants from the *DS*  
434 can be achieved using lime, cement, silicates, and other additives, which subsequently enhance  
435 the matrix's compressive strength. In this context, Stabilisation/Solidification (*S/S*) has been  
436 elaborately discussed in the following sections.

#### 437 **4.3.1 Traditional chemical reagents**

438 The contamination of aquatic sediments with organic and/or inorganic pollutants is a  
439 widespread environmental problem. Stabilisation/Solidification (*S/S*) is a chemo-mechanical  
440 treatment that makes use of chemical reagents, such as cement, lime, and other binders.  
441 Cement-based *S/S* is a chemical treatment process that aims to either bind compounds of a  
442 hazardous waste stream into a stable insoluble form (stabilisation) or to entrap the waste within  
443 a solid cementitious matrix (solidification) (Wiles, 1987). It is widely established as an  
444 effective method for both improving the engineering properties of sediments and encapsulating  
445 contaminants (Barjoveanu *et al.*, 2018; Wang *et al.*, 2012; Palansooriya *et al.*, 2020; Qian *et*  
446 *al.*, 2008; Wang *et al.*, 2012; Zentar *et al.*, 2012). The US Environmental Protection Agency  
447 (*USEPA*) defines *S/S* treatment as “a process that encapsulates waste to form a solid material”  
448 (*USEPA*, 1997).



449           The contaminated sediments are converted into solid forms and entrapped within a  
450 granular or monolithic matrix through the chemical reactions developed during the process of  
451 solidification. The treatment limits the mobility or solubility of the hazardous components and  
452 does not necessarily alter the physical nature of the contaminants (USEPA, 2004). The  
453 combined application of the solidification and stabilisation process ensures the mixing of the  
454 contaminated sediments with the treatment agents and consequently, both the physical and  
455 chemical immobilisation of the hazardous components occurs. The ultimate objective of *S/S* is  
456 to complete the transformation of toxic components into nontoxic forms. However, the  
457 objective of *S/S* technology not only includes limiting the solubility of the contaminant and  
458 decreasing the surface area across which contaminant transport might occur, but also the  
459 improvement of the mechanical properties of the sediments (soils).

460           In addition, *S/S* treatments offer several advantages over other treatment technologies  
461 (e.g., Oh et al., 2011), including: i) costs, and ii) implementability. As exemplified in Table 3,  
462 several are the examples of their use to improve the physical, mechanical, and environmental  
463 properties of *DS*. The authors show that *S/S* treatments are effective to: (a) reduce the initial  
464 fluid content of sediments, (b) eliminate or stabilize the hazardous compounds, such as heavy  
465 metals and *OM*, (c) improve the mechanical properties of the sediments and (d) prompt the  
466 production of new geomaterials or granular materials to address novel options of sediment  
467 management, i.e., base materials for pavement construction, cement production, light-weight  
468 concrete production and brick fabrication. However, a notable disadvantage of *S/S* is that, the  
469 contaminants, although immobilised, are still present in the sediments. Moreover, organic oily  
470 compounds can represent a threat for the efficacy of cement stabilisation.

471

**Table 3.** S/S treatments of sediments in the literature

Reference	Material	Binder	Effect	Purpose
Boutouil and Levacher (2000)	Sediment contaminated by heavy metals (port of Le Havre)	Cement	- Increase in compressive strength - Decrease in leaching of heavy metals	Road or civil construction materials
Colin (2003)	Sediment (Rouen harbour)	Cement	- Improvement of physical, mechanical, and environmental properties	Road bed materials
Scordia <i>et al.</i> (2008)	Sediment contaminated by heavy metals and organic matter (channel linking Charleroi to Brussels)	Roc Sol (commercial product) and lime	- Increase in bearing capacity, compressive strength and Brazilian tension - Decrease in expansive behaviour	
Silitonga <i>et al.</i> (2010)	Sediment contaminated by heavy metals (port En-Bessin)	Cement and silica fume	-Increase in compressive strength - Decrease in leaching of heavy metals	
Wang <i>et al.</i> (2012)	Marine sediment (Dunkirk)	Lime and cement	- Increase in unconfined compressive strength and tensile strength	
Zentar <i>et al.</i> (2012)	Marine sediment (Dunkirk)	Cement and fly-ash	- Increase in tensile strength and compressive strength - Restrain of the swelling potential	
Kogbara (2014)	Contaminated sediment	Cement and blends of cement-fly ash, cement-slag, lime-slag, lime-fly ash	- Increase in compressive strength. - Decrease in leaching of heavy metals	Sediment management
Radenović <i>et al.</i> (2019)	Highly contaminated sediment, dominantly by heavy metals (Great Bačka canal)	Kaolinite, quicklime and cement	- Decrease in leaching of heavy metals	
Mastoi <i>et al.</i> (2022)	Sediment (Nanhu lake located in the Chinese city of Wuhan)	Cement	- Increase in compressive strength	Civil construction materials

472

473           With respect to the binders and additives used for *S/S* treatments of sediments, the  
474 literature reports a variety of solutions: lime or cement alone (e.g., Jauberthie *et al.*, 2010,  
475 Federico *et al.*, 2015), lime combined with high alkali and slag cements, fly ashes (Grubb *et*  
476 *al.*, 2010) or pozzolana (e.g., Zoubir *et al.*, 2013), to cite a few of them. Due to the heterogeneity  
477 in the properties of *DS* and physicochemical reactions between binders and metal and organic  
478 contaminants, different binders show different efficiencies for pollutant immobilisation. The  
479 ability of Portland cement to improve the sediments' geotechnical characteristics (Wang *et al.*,  
480 2012; Zentar *et al.*, 2012) and immobilise contaminants has been widely documented (Xue *et*  
481 *al.*, 2017; Wang *et al.*, 2018). However, there are some cases in which the mobility of  
482 contaminants in marine sediments does not reduce when treated with either lime and cement  
483 or additives (Taneez *et al.*, 2016; Xu, 2017). Overall, most studies to date have focused on  
484 individual parameters and there remains a lack of systematic research considering the  
485 combination of factors affecting the properties of stabilised sediments in a unified way. In the  
486 *S/S* treatment of organic compounds using cement alone, the contaminants are physically  
487 trapped within the pores in the cement matrix and are not reacting with the polar inorganic  
488 components of the cement constituents. The use of adsorbents such as organophilic clays and  
489 *AC*, either as a pre-treatment or as additives in the cement mix, can more effectively immobilize  
490 organic compounds in the cement matrix (Paria and Yuet, 2006). However, organic compounds  
491 have been found to retard the cement setting process by forming a protective layer around the  
492 cement grain, thus hindering the formation of calcium hydroxide (Sora *et al.*, 2005).

#### 493 **4.3.2 Sustainable solutions**

494           Despite cement being largely used in several *S/S* treatments, it is responsible for 5–8%  
495 of global anthropogenic CO<sub>2</sub> emissions and accounts for 12–15% of total industry energy use  
496 (Ali *et al.*, 2011; Scrivener and Kirkpatrick, 2008). This finding has prompted new research to  
497 investigate more environmentally friendly and sustainable materials for *S/S* applications. For

498 example, in recent years, research has focused on proposing a partial or total replacement of  
499 traditional cement with natural additives to treat contaminated sediments (Patmont *et al.*, 2015;  
500 Lofrano *et al.*, 2017). For example, a variety of waste shells, including eggshells, mussel shells,  
501 and oyster shells, were analysed by researchers to verify their efficacy to immobilise pollutants  
502 (in particular heavy metals) in contaminated soils (Islam *et al.*, 2017; Liu *et al.*, 2018).

503 Moreover, Paleologos *et al.* (2022) have proposed the results regarding the mechanical  
504 stabilisation of fine marine sediments with mixtures formed by cement partially substituted by  
505 mussel shell powder produced without calcination. From the findings of microstructural  
506 investigations and scanning electron microscopy (*SEM*) images, it is clear that shell powder is  
507 completely encapsulated in the cement-sediment matrix, acting as a binder due to the elongated  
508 shape of the mussel shell fabric. This microstructural feature of mussel shells enhances the  
509 electrolytic exchanges between sediments and cement, and thus increases the contact areas  
510 between the mineral particles promoting the chemical hydration reactions. Such peculiarity of  
511 mussel shells makes them a valuable substitute for cement in stabilisation of *DS*, and provides  
512 a viable alternative that can reduce the consumption of natural resources (such as crushed rock,  
513 sand and gravel, extracted through highly impactful quarrying or river exploitation activities)  
514 and lower the amount of binders used in traditional sediment stabilisation practice.

#### 515 **4.4 Electrochemical remediation**

516 The electrochemical remediation includes the passage of electric current between the  
517 cathode and anode rods inserted in the slurry of the *DS* (Pedersen *et al.*, 2015). The positively  
518 charged particles start moving towards the cathode, while the negatively charged particles  
519 move towards the anode due to the influence of generated electric field (Pal and Hogland, 2022;  
520 Pedersen *et al.*, 2015). However, as the fine-grained sediments have more affinity to adsorb  
521 metal ions on their surface, electrochemical remediation finds its utility for their remediation  
522 (Pal and Hogland, 2022; Peng *et al.*, 2009). The basic mechanisms involved in this remediation

523 include (i) electro-osmosis, (ii) electromigration, (iii) electrolysis, and (iv) electrophoresis,  
524 which can remove even soluble metal ions and ions bounded with sediment oxides, hydroxides,  
525 carbonates, nitrates, and cyanide (Peng *et al.*, 2009).

526 Furthermore, the influencing factors responsible for heavy metal extraction include  
527 agitation rate, sediment properties, moisture content, *OM*, current flow, and extraction duration  
528 (Pedersen *et al.*, 2015). It should be noted that the *pH* of the sediment slurry controls the  
529 electrochemical remediation, viz., if the *pH* of the slurry is basic, then the precipitation of metal  
530 ions forms hydroxides or oxy-hydroxides, whereas, in the case of acidic nature, the metal ions  
531 are more likely to get desorb or solubilize (Pal and Hogland, 2022; Pedersen *et al.*, 2015). Also,  
532 it should be noted that the extraction ability of heavy metals from the *DS* slurry can be enhanced  
533 by using desorbing agents, viz. acidification and surfactants, which has the ability to solubilize  
534 the metal oxides, nitrates, hydroxides, carbonates, etc. adsorbed on the sediment surface (Peng  
535 *et al.*, 2018). The electrochemical remediation process results in chemical transformations that  
536 change the accessibility and mobility of the toxic substances making them more hazardous for  
537 living organisms and making it necessary to perform toxicity analysis (Benamar *et al.*, 2019).  
538 It has been reported by earlier researchers that due to the low mobility of charged particles in  
539 the process of electric remediation, the effect of electrophoresis can be ignored. Thus, the action  
540 of electric migration and electro-osmosis is used for actual migration of heavy metal ions in  
541 soil pore water under the influence of an external direct current electric field (Han *et al.*, 2021).  
542 Therefore, it is highly recommended to conduct elaborate and extensive studies to optimize  
543 electrochemical techniques with desorbing agent modifications for heavy metals extraction.

#### 544 **4.5 Biological remediation**

545 The biological processes (read bioremediation) include the action of microorganisms  
546 or plants (viz., phytoremediation) for the remediation of contaminated *DS* by oxidation of the  
547 *PAHs*, hydrocarbons, and mineral oils, converting them into non-hazardous compounds (Feng

548 *et al.*, 2022). Bioremediation can be achieved due to naturally occurring indigenous microbes  
549 by introducing nutrients in the form of water-soluble, slow-release, and oleophilic fertilizers  
550 and oxygen (biostimulation) in the contaminated sediments or can also be achieved by the  
551 addition of alien microorganisms (bioaugmentation viz., external microorganisms, enzymes,  
552 nutrients, etc.) to the *DS* (Maletić *et al.*, 2019). As the microorganisms available in sediments  
553 plays a major role in the biodegradation of the contaminants, this process is known as natural  
554 attenuation (Maletić *et al.*, 2019). However, the duration required for contaminant degradation  
555 is noticeably high, but this treatment presents a low impact on carbon footprint (Crocetti *et al.*,  
556 2022). Phytoremediation comprises remediation by plants and the root colonizing microbes to  
557 degrade the toxic compounds to non-toxic metabolites and is effective for the immobilisation  
558 of Zn, Fe, Mn, Cd, etc. (Peng *et al.*, 2009). Unfortunately, bioremediation is less predictable  
559 than other processes (Maletić *et al.*, 2019). Therefore, more extensive research should be  
560 conducted considering different plants, microorganisms, enzymes, nutrients, and  
561 environmental conditions to establish the suitability and effectiveness of this method and  
562 proper guidelines and regulations.

563 The permissible limits of heavy metals and *PAHs* have been presented in *Table S2*,  
564 which might be achieved with the methodologies discussed above in *Section 4* of this paper.  
565 The regulatory conditions that necessitate to be achieved by allowing proper treatment would  
566 be helpful in promoting *DS* use as raw material for on-shore and off-shore applications.  
567 Furthermore, establishing of various sediment remediation techniques on the basis of their type  
568 and concentration of contaminants is a future scope of work.

## 569 **5 Utilization strategies for Dredged sediments**

570 The *DS* finds its utilization in soil filling, coastal nourishment, construction purposes,  
571 horticulture, forestry, agriculture, etc., a few of which have been discussed elaborately in the

572 following sections. Here, it should be noted that the *DS* should comply with pollutant-specific  
573 regulations before utilization.

### 574 **5.1 Agricultural applications**

575 The contaminated *DS* can be used for horticulture, forestry, and agricultural  
576 applications (Crocetti *et al.*, 2022; Rakshith and Singh, 2017). The presence of micro-nutrients  
577 (viz., Cu, Fe, Mn, Zn, etc.) and macro-nutrients (viz., C, Ca, K, N, P, Mg, etc.) induces inherent  
578 fertility in the *DS* (Renella, 2021). Furthermore, freshwater *DS* application to agricultural land  
579 increases the *OM* content, cation exchange capacity of the soil mass, improves soil structure,  
580 enhances water retention, and thus increases overall soil microbiological, chemical, and  
581 physical fertility, apart from improvement in sorption properties and nutrient concentrations  
582 (Leue and Lang, 2012; Renella, 2021). Kazberuk *et al.* (2021) performed a pot experiment on  
583 white mustard as a test plant to evaluate the possibility of using bottom sediments from  
584 reservoirs and rivers contaminated with heavy metals (viz., Zn, Cu, Cd, Pb) and its impact on  
585 soil and plants. The obtained results motivate further research as the bottom sediment added  
586 soil yield was higher than the control soil. Thus, there is a dire need for field-scale experiments  
587 to understand the behavior of various crops, contaminant transmission in food chain, etc., with  
588 the addition of *DS* having different concentrations of *OM*, organic carbon, nutrients, heavy  
589 metals, etc.

### 590 **5.2 Infrastructure development**

591 The transformation of *DS* into geomaterials is an attractive way to relieve the shortage of  
592 high-quality raw materials for various applications and projects, such as constructing coastal  
593 highways and manufacturing pavements (Couvidat *et al.*, 2016), road construction (Hussan *et*  
594 *al.*, 2023), producing cleaner pervious concrete (Beddaa *et al.*, 2023), constructing plant-  
595 growing substrate to cultivate lettuce (Ferrans *et al.*, 2022), fill material (Wang *et al.*, 2018),  
596 bricks (Wang *et al.*, 2015), and breakwaters. The treatments techniques can be different and

597 vary depending on the target to be reached for the sediment reuse; for example: (i) chemically  
598 immobilize the contaminants, reducing the leachability and bioavailability, and (ii)  
599 mechanically stabilize the material for its reuse as new construction material. The reuse of  
600 contaminated *DS* would facilitate the recycling of dredged materials from local sources and  
601 save natural soil resources and transportation costs for construction, in line with the philosophy  
602 of the circular economy (Todaro *et al.*, 2016; Wang *et al.*, 2012). However, only 5% of the  
603 materials generated from recycling operations are currently used in public works (Wang *et al.*,  
604 2014). These data indicate that in the context of sustainable development, it is still necessary  
605 to further study solutions for recycling sediments as renewable geomaterials. CemShell-based  
606 solutions can provide the physics-based methodology for achieving the change of focus in  
607 relation to the management of *DS* and mussel shells (Paleologos *et al.*, 2022). In particular, the  
608 leaching test is one important aspect in the environmental assessment of the reuse options of  
609 treated sediments. The selection of an appropriate test or combination of tests (e.g., batch tests  
610 or column tests) is vital for predicting the long-term contaminants' release into the environment.  
611 Several authors have shown that to successfully transform a *DS* (i.e., a waste) into a geo-  
612 material, a multi-level testing program (e.g., with geotechnical and leaching tests) is required  
613 to investigate the effective recovery of treated sediments (Barjoveanu *et al.*, 2018; Todaro *et*  
614 *al.*, 2020).

615 In this context, it should be noted that the *DS* primarily consists of mineral, organic and  
616 liquid phases. It is well established that the presence of *OM* in sediments affects their  
617 engineering properties adversely (Benaissa *et al.*, 2016; Hamouche and Zentar, 2020a). It is  
618 worth mentioning here that the valorization of *DS* should be conducted considering the  
619 environmental, economic, geotechnical, and mechanical feasibility and sustainability. Also, it  
620 has been reported that the permissible limit of *OM* in road pavement material should be  $\approx 2\%$  -  
621  $4\%$ , while that for embankment material should be  $\approx 5\%$  -  $7\%$  (Hamouche and Zentar, 2020a).



622 The concern with the presence of *OM* is its decomposition with time, leading to an increase in  
623 the porosity and, thus, an increase in the compressibility, which is a controlling parameter for  
624 most of the infrastructure developments. Furthermore, *DS* utilization for infrastructure  
625 development, viz., road pavement, embankment, etc., could be an interesting and sustainable  
626 solution that covers sustainable development goals (*SDGs*), viz., *SDG-8*, *SDG-9*, *SDG-11*,  
627 *SDG-12*, *SDG-13*, *SDG-14* and *SDG-17* (Suedel *et al.*, 2022). Unfortunately, one of the major  
628 barriers is the perception of different stakeholders towards *DS* as a waste material that needs to  
629 be changed.

## 630 **6. Technology Readiness Level (*TRL*) and Circular Economy**

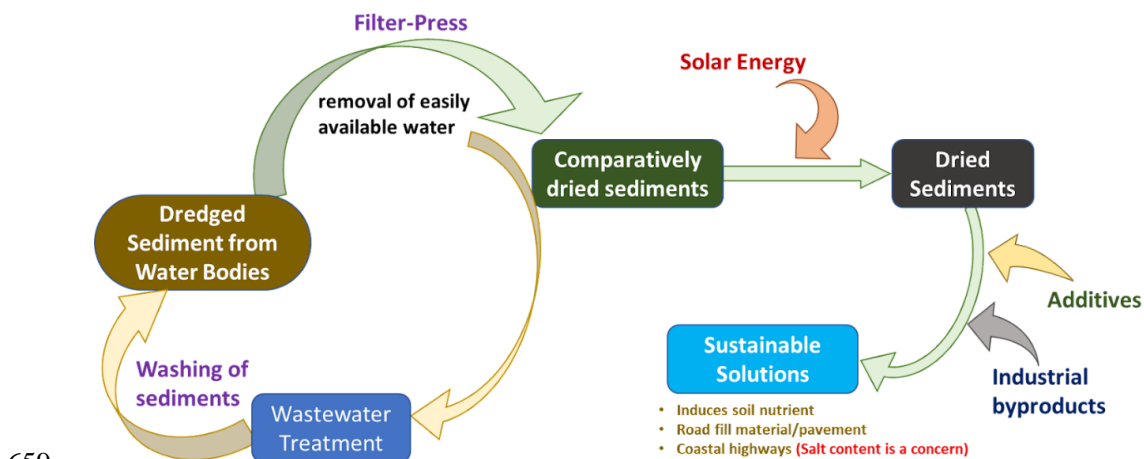
631 The technical maturity of any process/technique is assessed through the technology  
632 readiness level, *TRL*, a point-based framework system from concept to commercial use.  
633 Unfortunately, most configurations and processes used for the contaminated *DS* focusing  
634 circular economy objective is at low *TRL* and have only been tested in lab conditions (Crocetti  
635 *et al.*, 2022). Therefore, there is an urgent need to establish effective methodologies and  
636 upscaling the processes to pilot and field scale.

637 It should be noted that the major barrier behind the bulk utilization of *DS* include (i)  
638 policy/legal challenges at the local, national, and international levels, (ii) no guidelines against  
639 dumping of *DS* in the deep sea, which is catastrophic for marine flora and fauna and waste of  
640 money and material, (iii) lack of government initiative and policy framework in respect of  
641 promoting mission zero waste, and (iv) delay in approval from environmental agencies. Thus,  
642 there is an urgent requirement for the introduction of codes, standards, and guidelines for the  
643 utilization of *DS* as a secondary product that can reduce the use of raw materials and fill the  
644 gap created due to the scarcity of natural aggregates promoting circular economy and  
645 sustainable development.

646 Furthermore, the possible applications for *DS* utilization discussed should be modified,  
 647 bringing utilization of industrial by-products (*IBPs*) and other waste material replacing  
 648 conventional additives/modifiers, viz., cement, lime, sand, etc. (Singh et al., 2023; Singh and  
 649 Singh, 2023). It should be noted that this approach of using *IBPs* and waste will improve the  
 650 production chain, making it more environmentally friendly, sustainable, cost and energy  
 651 effective, apart from reducing carbon, water, and land footprints. Keeping in view of sediment  
 652 management and considering sustainable development, *Figure 2* has been developed. From the  
 653 figure, it is clear that sustainable products can be generated by proper management of  
 654 sediments, and thus better *TRL*'s can be achieved. In this context, industries having *IBPs*,  
 655 government bodies/policymakers, researchers, and ports should come forward to make  
 656 sustainable management of *DS* a new reality.

657

658



660 **Figure 2.** Sediment management considering sustainable development and circular economy

## 661 **7. Prospects and recommendations**

662 Based on the critical synthesis of the literature that deals with the (i) contamination  
663 assessment, (ii) testing and characterization methodology, (iii) remediation strategies, (iv)  
664 utilization strategies, and (v) *TRLs* and circular economy of the dredged sediments, the  
665 following generalized prospects and recommendations can be drawn:

666 (i) The major contaminants in the *DS* are heavy metals, *PAHs* and *PCBs*, which are  
667 case- and site-specific, and their concentration changes with prevailing  
668 environmental conditions and nearby contamination sources. In this context, the  
669 source, concentration level, and effect of emerging contaminants in the *DS* need to  
670 be further deepened case-by-case, which would help understand the primary and  
671 secondary sources of contaminants and address the most suitable remediation  
672 strategies.

673 (ii) The standard characterization and testing methodologies can fail when dealing with  
674 *DS* due to contaminants in the soil matrix and various pore fluid chemical  
675 compositions. In some cases, non-standard approaches need to be used to catch the  
676 complexity of the sediment matrix and fully understand the coupled related chemo-  
677 mechanical effects.

678 (iii) Based on a comparative procedure, the proposed framework appears transparent  
679 and interdisciplinary and represents a reliable way to select the most sustainable  
680 remediation alternative. The long-term performance is yet a matter of study for  
681 several remedial options (such as reactive capping and S/S). Moreover, evidence is  
682 provided about the need for further testing of these technologies at the real scale  
683 and to carry out a cost-benefit analysis considering the life-cycle impact analysis.

684 (iv) The possible utilization schemes of *DS* in agricultural applications and  
685 infrastructural development has been discussed. Also, the efforts tried by earlier  
686 researchers have been mentioned elaborately.

687 (v) The technology readiness level and circular economy perspective of *DS* utilization  
688 have been shown to open broad perspectives for scientists and policymakers to  
689 contribute and establish guidelines. Furthermore, such scientific advances will  
690 likely prompt a paradigm shift towards more sustainable industrial development.

691 Keeping this in view, the utilization of *DS* considering a circular economy perspective and  
692 *SDGs* needs to be established, which will further solve the issue being created due to the  
693 scarcity of natural aggregates and raw materials. Also, remediation techniques keeping in  
694 reference to type and concentration of the contaminants needs to be established. Furthermore,  
695 the local government should promote *DS* utilization by subsidizing transport facilities, creating  
696 a flexible licensing system for *DS* processing, policies to design life cycle assessments, and  
697 compulsory use of *DS* as secondary material.

698

### 699 **Acknowledgments**

700 Part of the research has brought about the filing of patent application (number IT  
701 102021000025103, title: “Stabilisation method of marine clays by shells and cements”) by  
702 POLIBA and ETH Zurich.

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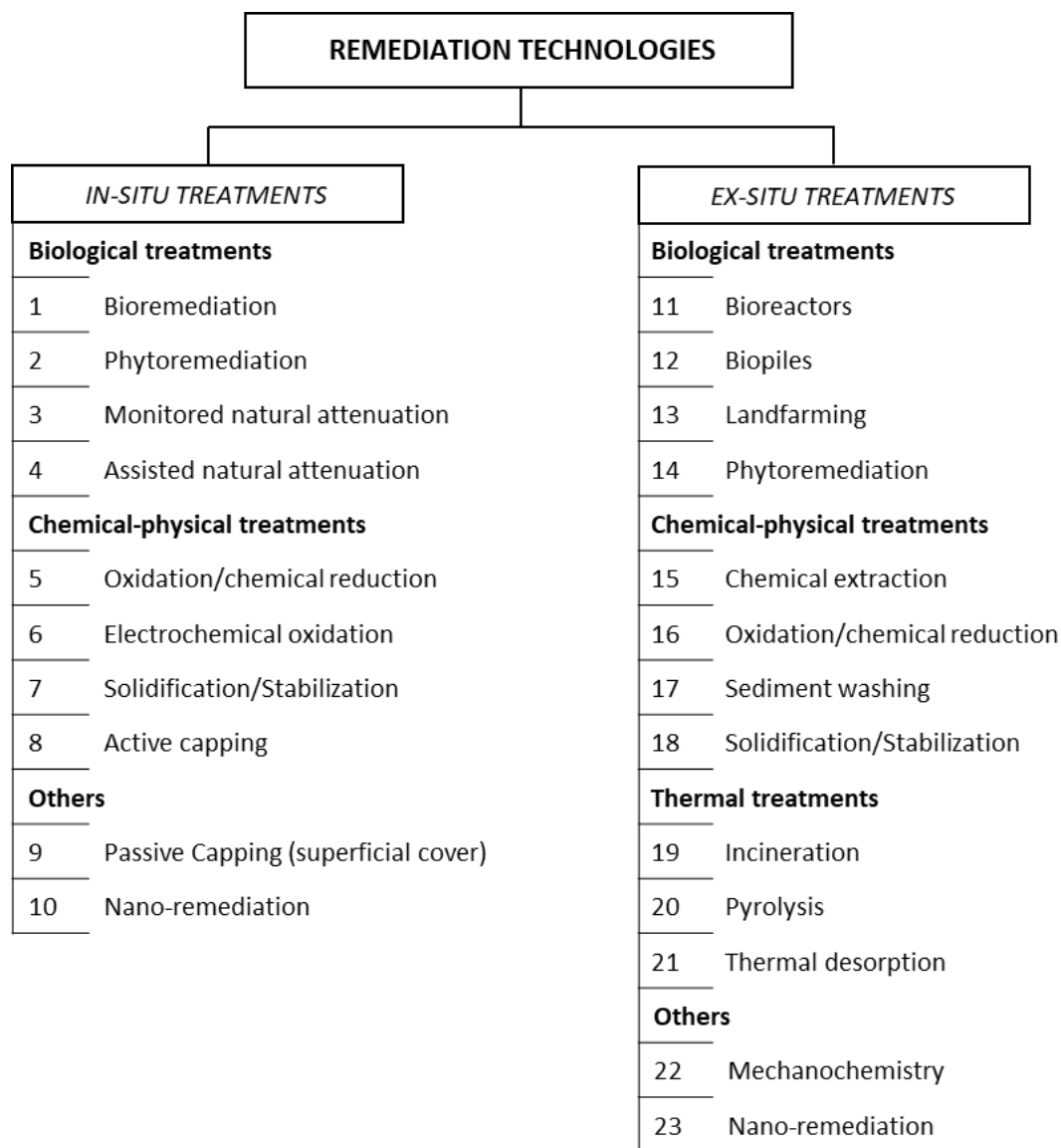
Table 1. Summary of the studies conducted on the concentrations of ECs in DS

Reference	Study area	Source of DS	ECs detected with concentration	Possible source of ECs	Detection techniques
Torres <i>et al.</i> (2009)	Port of Santos, Brazil	Marine sediments (18 samples from dredged areas and disposal sites, 4 samples from hopper dredge)	PAH (27.86 to 679.35 $\mu\text{g}/\text{kg}$ ); PCB (0.17 to 12.33 $\mu\text{g}/\text{kg}$ )	Emissions and activities of steel plant and industrial complex	Gas Chromatography/Mass Spectroscopy (GC/MS)
Rocha <i>et al.</i> (2011)	Porto region, Portugal	4 river estuary and 2 marine beach sediments	PAH [Estuary (98.40 to 156.50 $\mu\text{g}/\text{kg}$ dw); Marine sediment (52.00 to 54.80 $\mu\text{g}/\text{kg}$ dw)]	-	GC/MS
Tavakoly Sany <i>et al.</i> (2014)	Klang strait, Malaysia	Coastal sediment	16 compounds of PAHs (994.02 $\pm$ 918.10 $\mu\text{g}/\text{kg}$ dw)	Contamination due to cargo transport, petrogenic spillage and pyrogenetic combustion	GC/MS
Kafilzadeh (2015)	Soltan Abad river, Iran	River sediment (4 sampling locations at a depth of 5 cm from the bed)	16 compounds of PAHs (180.30 to 504.00 $\mu\text{g}/\text{kg}$ )	Pyrogenic combustion and petrogenic spillage	Gas Chromatography/Flame Ionisation Detection (GC/FID)
Couvidat <i>et al.</i> (2018)	Port in the south of France	Harbour sea bed (Top 50-80 cm)	16 compounds of PAHs (62.18-62.40 mg/kg) 7 compounds of PCBs (0.96-0.97 mg/kg) 3 compounds of Organotin compounds (65.50 mg/kg)	Extensive anthropogenic activity for centuries and contamination due to industrial activity	GC/MS and low-resolution MS
Shilla and Routh (2018)	Rufiji Estuary, Tanzania	River sediment (top 1-2 cm sediment was scrapped on South, middle and north parts of Rufiji Delta)	19 compounds of PAHs (128 to 377 $\mu\text{g}/\text{kg}$ )	Petrogenic spillage and pyrogenic combustion of coal and biomass (mainly grass and wood)	GC/MS
Norén <i>et al.</i> (2020)	Two ports, three marina and one waterway leading to the marina in Sweden	Marine environment	Tributyltin: ports (150230 mg/kg); marina (50 $\pm$ 50 and 310 $\pm$ 240 mg/kg); waterway (70 $\pm$ 60 mg/kg)	Pollutants released by recreational and public transport boats, cargo vessels. Effluents from Cu production, wastewater treatment, battery production industries and shipyards.	-

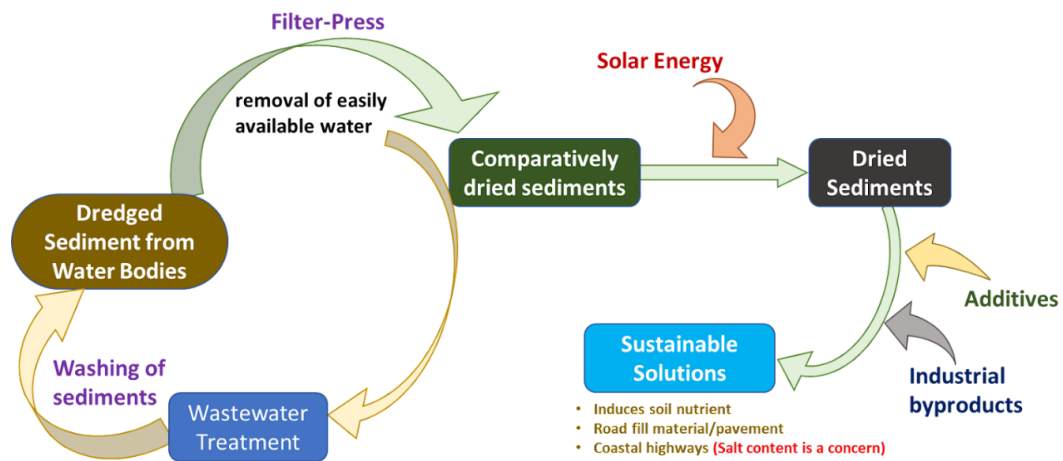
	Apatites	Metals	Alirol <i>et al.</i> (2012); Wang <i>et al.</i> (2016); Zhang <i>et al.</i> (2016)
	Bauxite	Metals	Taneez <i>et al.</i> (2018) <i>Environmental Geotechnics</i>
	Biochars	Organics/inorganic	Bianco <i>et al.</i> (2021); Janssen Beckingham (2013); Ting <i>et al.</i>
	Organoclays	Organics/inorganic	Erten <i>et al.</i> (2012); Olsta (201 Pagnozzi <i>et al.</i> (2020)
	Zeolites	Metals	Gu <i>et al.</i> (2019); Kang and Pa (2015)
Degrading	Bioremediation agents	Organics	Atashgahi <i>et al.</i> (2014); Sun <i>e</i> (2010); Wang <i>et al.</i> (2014)
	Zero-Valent Iron, ZVI	Organics	Chapman <i>et al.</i> (2020); Hu <i>et</i> (2020)

Levaener (2000)	Le Havre)			heavy metals
Colin (2003)	Sediment (Rouen harbour)	Cement	Environmental Geotechnics	Improvement of physical mechanical, and environmental properties
Scordia <i>et al.</i> (2008)	Sediment contaminated by heavy metals and organic matter (channel linking Charleroi to Brussels)	Roc Sol (commercial product) and lime		- Increase in bearing capacity - Increase in compressive strength and Brazilian tension - Decrease in expansion behaviour
Silitonga <i>et al.</i> (2010)	Sediment contaminated by heavy metals (port En-Bessin)	Cement and silica fume		- Increase in compressive strength - Decrease in leaching of heavy metals
Wang <i>et al.</i> (2012)	Marine sediment (Dunkirk)	Lime and cement		- Increase in unconfined compressive strength and tensile strength
Zentar <i>et al.</i> (2012)	Marine sediment (Dunkirk)	Cement and fly-ash		- Increase in tensile strength and compressive strength - Restrain of the swelling potential
Kogbara (2014)	Contaminated sediment	Cement and blends of cement-fly ash, cement-slag, lime-slag, lime-fly ash		- Increase in compressive strength. - Decrease in leaching heavy metals
Radenović <i>et al.</i> (2019)	Highly contaminated sediment, dominantly by heavy metals (Great Bačka canal)	Kaolinite, quicklime and cement		- Decrease in leaching heavy metals
Mastoi <i>et al.</i> (2022)	Sediment (Nanhu lake located in the Chinese city of Wuhan)	Cement		- Increase in compressive strength

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**Figure 1.** Sediments remediation technologies for in-situ and ex-situ treatments



**Figure 2.** Sediment management considering sustainable development and circular economy