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

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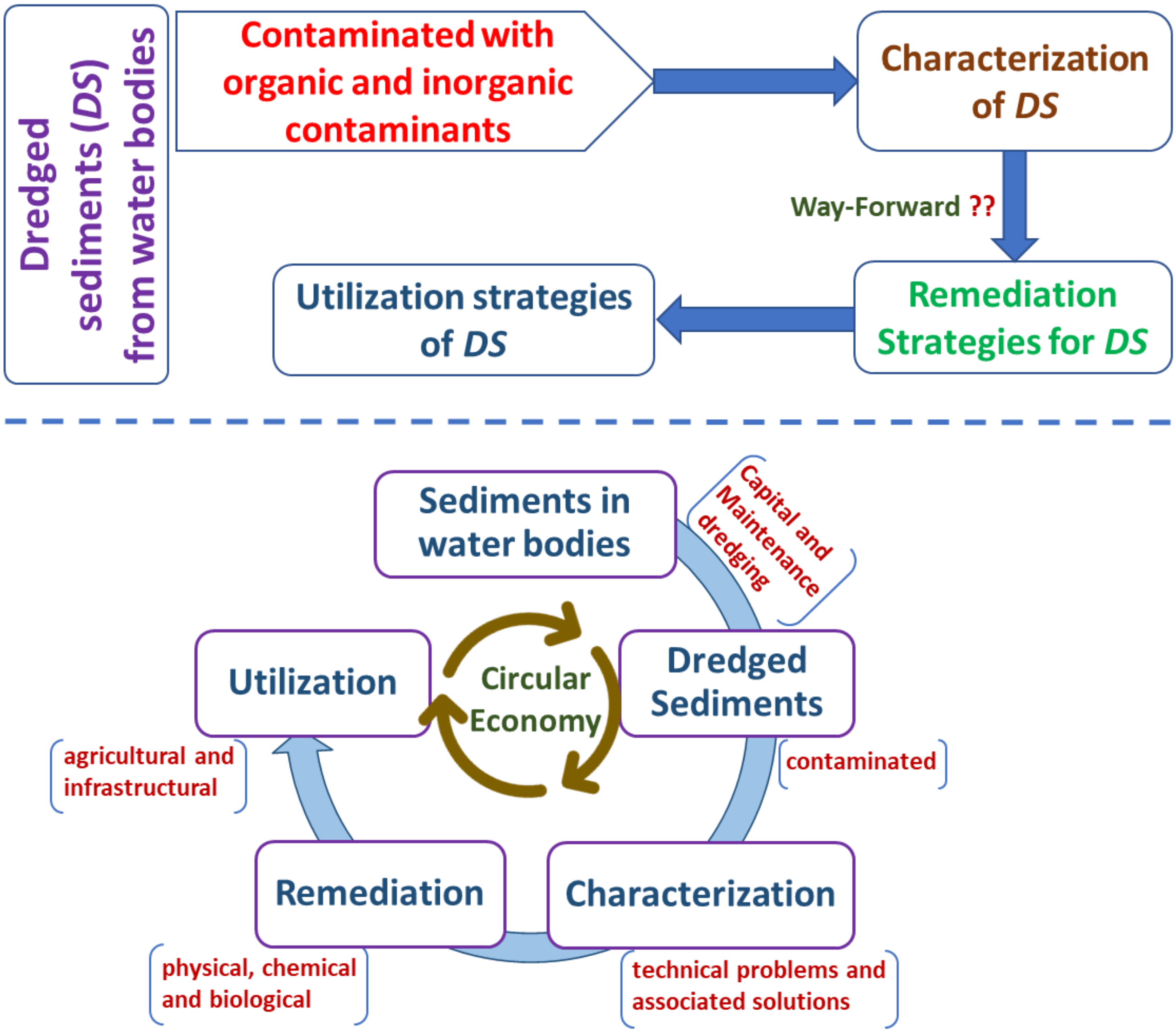
Abstract

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

Keywords: *sustainable development goals; dredged sediments; contamination; toxicity; remediation; utilization, UN SDG 8, UN SDG 9, UN SDG 11, UN SDG 12, UN SDG 13, UN SDG 14, UN SDG 17.*

Graphical Abstract



Highlights

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

1 Introduction

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

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

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

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

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

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

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

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

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

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

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

Reference	**Study area**	**Source of *DS***	***EC*s detected with concentration**	**Possible source of *EC*s**	**Detection techniques**
Torres *et al.* (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 µg/kg); *PCB* (0.17 to 12.33 µg/kg)	Emissions and activities of steel plant and industrial complex	Gas Chromatography/Mass Spectroscopy (*GC/MS*)
Rocha *et al.* (2011)	Porto region, Portugal	4 river estuary and 2 marine beach sediments	*PAH* [Estuary (98.40 to 156.50 µg/kg dw); Marine sediment (52.00 to 54.80 µg/kg dw)]	-	*GC/MS*
Tavakoly Sany *et al.* (2014)	Klang strait, Malaysia	Coastal sediment	16 compounds of *PAH*s (994.02±918.10 µg/ 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 *PAH*s (180.30 to 504.00 µg/kg)	Pyrogenic combustion and petrogenic spillage	Gas Chromatography/Flame Ionisation Detection (*GC/FID*)
Couvidat *et al.* (2018)	Port in the south of France	Harbour sea bed (Top 50-80 cm)	16 compounds of *PAH*s (62.18-62.40 mg/kg) 7 compounds of *PCB*s (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 *PAH*s (128 to 377 µg/kg)	Petrogenic spillage and pyrogenic combustion of coal and biomass (mainly grass and wood)	*GC/MS*
Norén *et al.* (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.	-

3 Geotechnical characterization: technical problems and adopted solutions

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

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

For example, in situ capping is a remediation option that can be selected and designed only after a site characterisation which includes geotechnical considerations (Vitone *et al.*, 2016). Usually, contaminated sediments are predominantly fine-grained and often have high water content and compressibility, and low shear strength. Cap stability and settlement due to consolidation are geotechnical issues that may be important for cap effectiveness. After placement of a cap, consolidation of both the underlying contaminated sediment and the cap layer usually occurs (Reible *et al.*, 2014). The consolidation of the cap is typically small. On the other hand, the consolidation of the underlying contaminated sediments may be significant, especially when dealing with soft soils, and expresses porewater pressure from the contaminated layer up into the cap. Moreover, the fluid expelled during the consolidation process should be evaluated for the investigation of a contaminated marine site. In fact, contaminant migration can change sediment properties (e.g., consistency limits), influencing capping design (Erten *et al.*, 2011). The impact of these processes depends on the sediment geotechnical properties that should be known for an efficient design of capping (Reible, 2014).

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

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

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

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

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

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

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

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

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

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

3.2 Effect of pore water salinity

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

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

3.3 Use of statistical techniques for integrated sediment characterization

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

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

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

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

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

4 Remediation strategies for contaminated sediments

4.1 Technologies overview

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

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

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

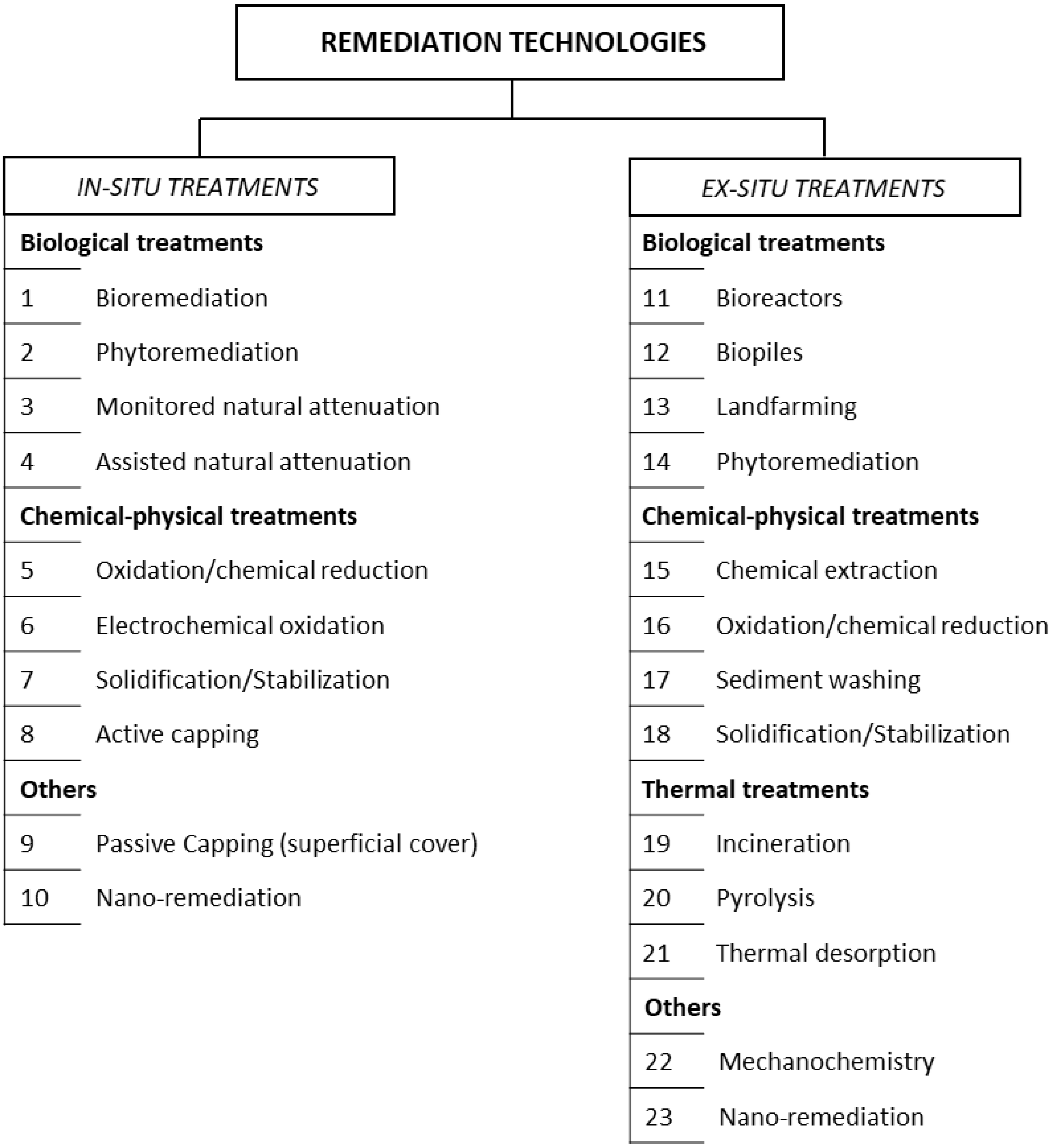


Figure 1. Sediments remediation technologies for in-situ and ex-situ treatments

4.2 In situ capping

In situ remedial alternatives generally involve: (i) natural attenuation, which is based on the assumption that, although sediments pose some risk, it is low enough that natural processes can reduce risk over time in a reasonably safe manner (De Gisi *et al.*, 2017b); (ii) in situ containment and treatment via capping, in which contaminated sediment is physically and chemically isolated from aquatic ecosystems or the contaminants in sediment are sequestered and degraded (Todaro *et al.*, 2018).

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

Sand capping has been widely investigated, being mostly utilized for large availability and ease of placement of sand (Jiao *et al.*, 2020). In Bortone *et al.* (2018) a sand cap was investigated to reduce the exposure of the aquatic ecosystem to *PCB*-contaminated sediments in Lake Hartwell, US. Specifically, it was demonstrated that *PCB* transport was extremely dependent on the cap characteristics, and that the cap thickness could be reduced at 20 cm-thick by using a high sorbent cap. In the study by Meric *et al.* (2014), a 7.5 cm thick sand cap reduced the bioavailability of *PCB*s by a factor of 100 compared to the no-capping scenario, but it did not influence the bioavailability of naphthalene.

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

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 *et al.* (2017)
	Apatites	Metals	Knox *et al.* (2012); Xing *et al.* (2016); Zhang *et al.* (2016)
	Bauxite	Metals	Taneez *et al.* (2018)
	Biochars	Organics/inorganic	Bianco *et al.* (2021); Janssen and Beckingham (2013); Ting *et al.* (2020)
	Organoclays	Organics/inorganic	Erten *et al.* (2012); Olsta (2010); Pagnozzi *et al.* (2020)
	Zeolites	Metals	Gu *et al.* (2019); Kang and Park (2015)
Degrading	Bioremediation agents	Organics	Atashgahi *et al.* (2014); Sun *et al.* (2010); Wang *et al.* (2014)
	Zero-Valent Iron, *ZVI*	Organics	Chapman *et al.* (2020); Hu *et al.* (2020)

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

4.3 Stabilisation/Solidification (*S/S*)

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

4.3.1 Traditional chemical reagents

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

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

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

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 *et al.* (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 *et al.* (2010)	Sediment contaminated by heavy metals (port En-Bessin)	Cement and silica fume	-Increase in compressive strength - Decrease in leaching of heavy metals	
Wang *et al.* (2012)	Marine sediment (Dunkirk)	Lime and cement	- Increase in unconfined compressive strength and tensile strength	
Zentar *et al.* (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
Rađenović *et al.* (2019)	Highly contaminated sediment, dominantly by heavy metals (Great Bačka canal)	Kaolinite, quicklime and cement	- Decrease in leaching of heavy metals	
Mastoi *et al.* (2022)	Sediment (Nanhu lake located in the Chinese city of Wuhan)	Cement	- Increase in compressive strength	Civil construction materials

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

4.3.2 Sustainable solutions

Despite cement being largely used in several *S/S* treatments, it is responsible for 5–8% of global anthropogenic CO_2 emissions and accounts for 12–15% of total industry energy use (Ali *et al.*, 2011; Scrivener and Kirkpatrick, 2008). This finding has prompted new research to investigate more environmentally friendly and sustainable materials for *S/S* applications. For example, in recent years, research has focused on proposing a partial or total replacement of traditional cement with natural additives to treat contaminated sediments (Patmont *et al.*, 2015; Lofrano *et al.*, 2017). For example, a variety of waste shells, including eggshells, mussel shells, and oyster shells, were analysed by researchers to verify their efficacy to immobilise pollutants (in particular heavy metals) in contaminated soils (Islam *et al.*, 2017; Liu *et al.*, 2018).

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

4.4 Electrochemical remediation

The electrochemical remediation includes the passage of electric current between the cathode and anode rods inserted in the slurry of the *DS* (Pedersen *et al.*, 2015). The positively charged particles start moving towards the cathode, while the negatively charged particles move towards the anode due to the influence of generated electric field (Pal and Hogland, 2022; Pedersen *et al.*, 2015). However, as the fine-grained sediments have more affinity to adsorb metal ions on their surface, electrochemical remediation finds its utility for their remediation (Pal and Hogland, 2022; Peng *et al.*, 2009). The basic mechanisms involved in this remediation include (i) electro-osmosis, (ii) electromigration, (iii) electrolysis, and (iv) electrophoresis, which can remove even soluble metal ions and ions bounded with sediment oxides, hydroxides, carbonates, nitrates, and cyanide (Peng *et al.*, 2009).

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

4.5 Biological remediation

The biological processes (read bioremediation) include the action of microorganisms or plants (viz., phytoremediation) for the remediation of contaminated *DS* by oxidation of the *PAH*s, hydrocarbons, and mineral oils, converting them into non-hazardous compounds (Feng *et al.*, 2022). Bioremediation can be achieved due to naturally occurring indigenous microbes by introducing nutrients in the form of water-soluble, slow-release, and oleophilic fertilizers and oxygen (biostimulation) in the contaminated sediments or can also be achieved by the addition of alien microorganisms (bioaugmentation viz., external microorganisms, enzymes, nutrients, etc.) to the *DS* (Maletić *et al.*, 2019). As the microorganisms available in sediments plays a major role in the biodegradation of the contaminants, this process is known as natural attenuation (Maletić *et al.*, 2019). However, the duration required for contaminant degradation is noticeably high, but this treatment presents a low impact on carbon footprint (Crocetti *et al.*, 2022). Phytoremediation comprises remediation by plants and the root colonizing microbes to degrade the toxic compounds to non-toxic metabolites and is effective for the immobilisation of Zn, Fe, Mn, Cd, etc. (Peng *et al.*, 2009). Unfortunately, bioremediation is less predictable than other processes (Maletić *et al.*, 2019). Therefore, more extensive research should be conducted considering different plants, microorganisms, enzymes, nutrients, and environmental conditions to establish the suitability and effectiveness of this method and proper guidelines and regulations.

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

5 Utilization strategies for Dredged sediments

The *DS* finds its utilization in soil filling, coastal nourishment, construction purposes, horticulture, forestry, agriculture, etc., a few of which have been discussed elaborately in the following sections. Here, it should be noted that the *DS* should comply with pollutant-specific regulations before utilization.

5.1 Agricultural applications

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

5.2 Infrastructure development

The transformation of *DS* into geomaterials is an attractive way to relieve the shortage of high-quality raw materials for various applications and projects, such as constructing coastal highways and manufacturing pavements (Couvidat *et al.*, 2016), road construction (Hussan et al., 2023), producing cleaner pervious concrete (Beddaa et al., 2023), constructing plant-growing substrate to cultivate lettuce (Ferrans et al., 2022), fill material (Wang *et al.*, 2018), bricks (Wang *et al.*, 2015), and breakwaters. The treatments techniques can be different and vary depending on the target to be reached for the sediment reuse; for example: (i) chemically immobilize the contaminants, reducing the leachability and bioavailability, and (ii) mechanically stabilize the material for its reuse as new construction material. The reuse of contaminated *DS* would facilitate the recycling of dredged materials from local sources and save natural soil resources and transportation costs for construction, in line with the philosophy of the circular economy (Todaro *et al.*, 2016; Wang *et al.*, 2012). However, only 5% of the materials generated from recycling operations are currently used in public works (Wang *et al.*, 2014). These data indicate that in the context of sustainable development, it is still necessary to further study solutions for recycling sediments as renewable geomaterials. CemShell-based solutions can provide the physics-based methodology for achieving the change of focus in relation to the management of *DS* and mussel shells (Paleologos *et al.*, 2022). In particular, the leaching test is one important aspect in the environmental assessment of the reuse options of treated sediments. The selection of an appropriate test or combination of tests (e.g., batch tests or column tests) is vital for predicting the long-term contaminants' release into the environment. Several authors have shown that to successfully transform a *DS* (i.e., a waste) into a geo-material, a multi-level testing program (e.g., with geotechnical and leaching tests) is required to investigate the effective recovery of treated sediments (Barjoveanu *et al*., 2018; Todaro *et al.*, 2020).

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

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

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

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

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

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

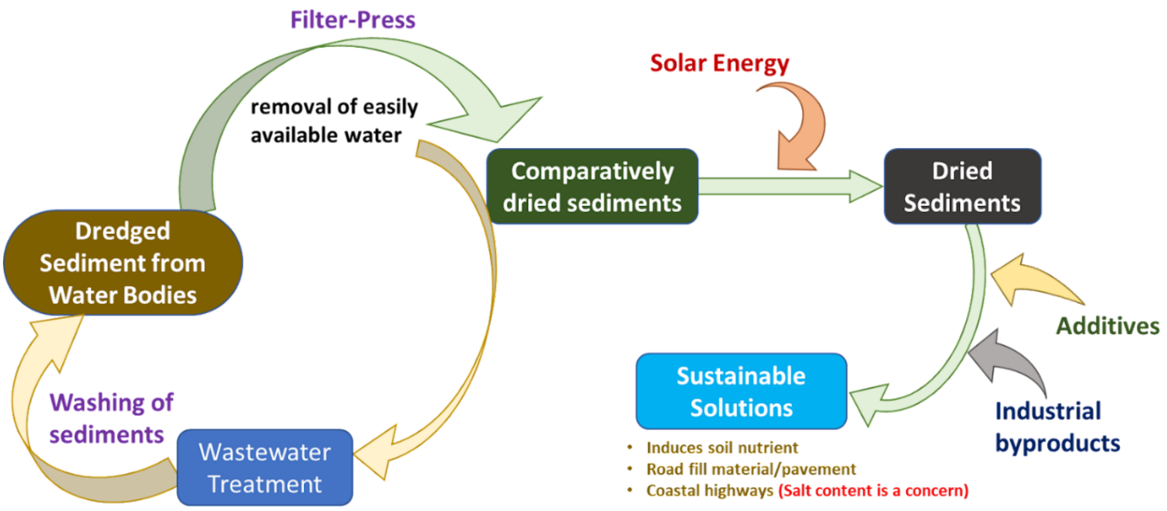


Figure 2. Sediment management considering sustainable development and circular economy

7. Prospects and recommendations

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

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

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

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

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

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

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

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