SUSTAINABLE MANAGEMENT OPTIONS AND BENEFICIAL USES FOR CONTAMINATED SEDIMENTS AND DREDGED MATERIAL

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ABSTRACT

Remediation and management of contaminated sediments and dredged material is often difficult due to the complex mixtures of chemical substances that usually impact the sediments. The selection of the best option that leads to the sustainable management of the sediments of a site is not an easy task, and it should be based on the integration of technical, economic, social and environmental criteria. In this paper, an overview of the main alternatives for the management of contaminated sediments and dredged material is presented. This work highlights the wide variety of techniques and treatments that can be applied, including no action (i.e. monitored natural recovery), in-situ capping, in-situ treatments, ex-situ treatments or confined disposal. The utilisation of contaminated dredged material for a beneficial use, directly or after treatment, should be considered as its preferred final destination. Therefore, special emphasis has been given in this study to the presentation of possible beneficial uses of dredged material. The results of our research group, which are summarised in this paper, about the use of contaminated sediments as alternative raw materials in the manufacture of traditional clay-based ceramics, can serve as an example of beneficial uses that lead to marketable products.

KEYWORDS: sediment, dredged material, remediation, treatment, beneficial use, ceramics.

INTRODUCTION

Sediment quality assessment should be carried out through the application of tiered decision-making frameworks which involve measurement of multiple lines of evidence (e.g. chemistry, acute or chronic toxicity, in situ benthic community alteration, biomagnification) in sequential steps [1]. Most tiered assessment frameworks begin with an evaluation of chemical levels in sediments. If the concentrations of contaminants are below sediment quality guideline (SQG) values [2], the sediments are perceived to pose negligible environmental risks, so the management options can be more flexible and less stringent [3]. However, when the decision reached after the application of the sequential tiers of a framework is that the sediments could pose an environmental risk, the need to remediate the contaminated sediments in a sustainable way arises, and management actions are therefore required.

Management of contaminated sediments should be based on a risk-based assessment strategy [4]. Furthermore, factors like costs [5], technical feasibility, short- and long-term effects and social repercussion should be considered as well [6].

Figure 1 shows a flow diagram for the management of sediments and/or dredged material, in which all the main options have been included. These options have been set in order of priority [7, 8] according to a sustainable management strategy. When dredging is necessary and sediments are not contaminated, aquatic relocation must be selected, since it is the better option owing to its low environmental impact. In other cases, other management options, among the ones shown in Figure 2, should be chosen, taking into account that the preferred final destination for dredged material should be its reutilisation for a beneficial use.

The aim of this paper is to present an overview of the main alternatives, techniques and treatments (outlined in Figure 2) that can be applied in the management of contaminated sediments and dredged material, emphasising the possible beneficial uses for dredged material. In this sense, this paper finally summarises the experimental results obtained by our research group about the use of contaminated sediments as alternative raw material in the manufacturing process of traditional clay based ceramic materials.

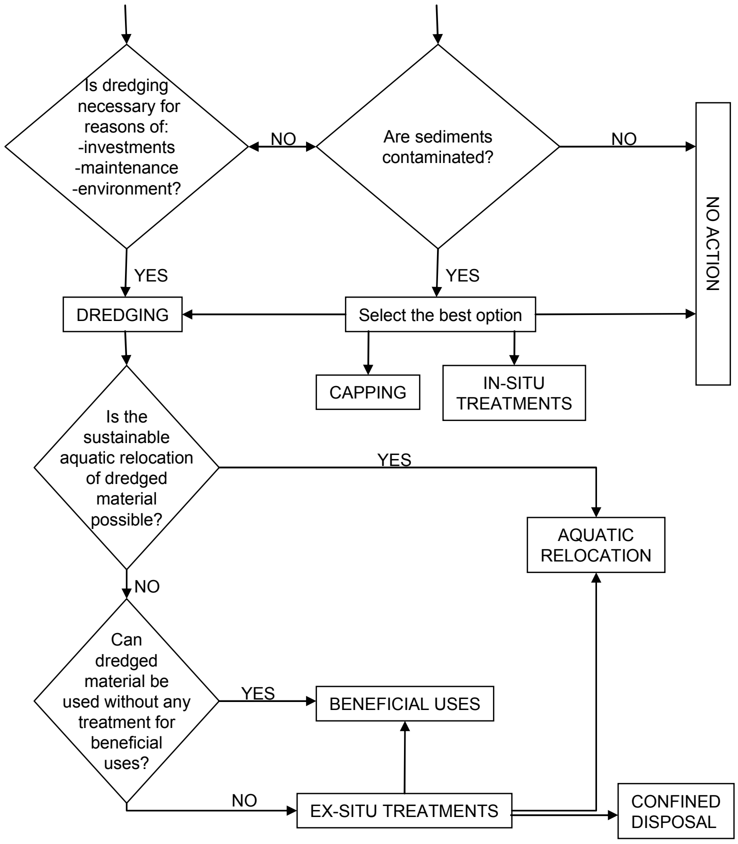

FIGURE 1 - Flow diagram for the management of sediments and/or dredged material.

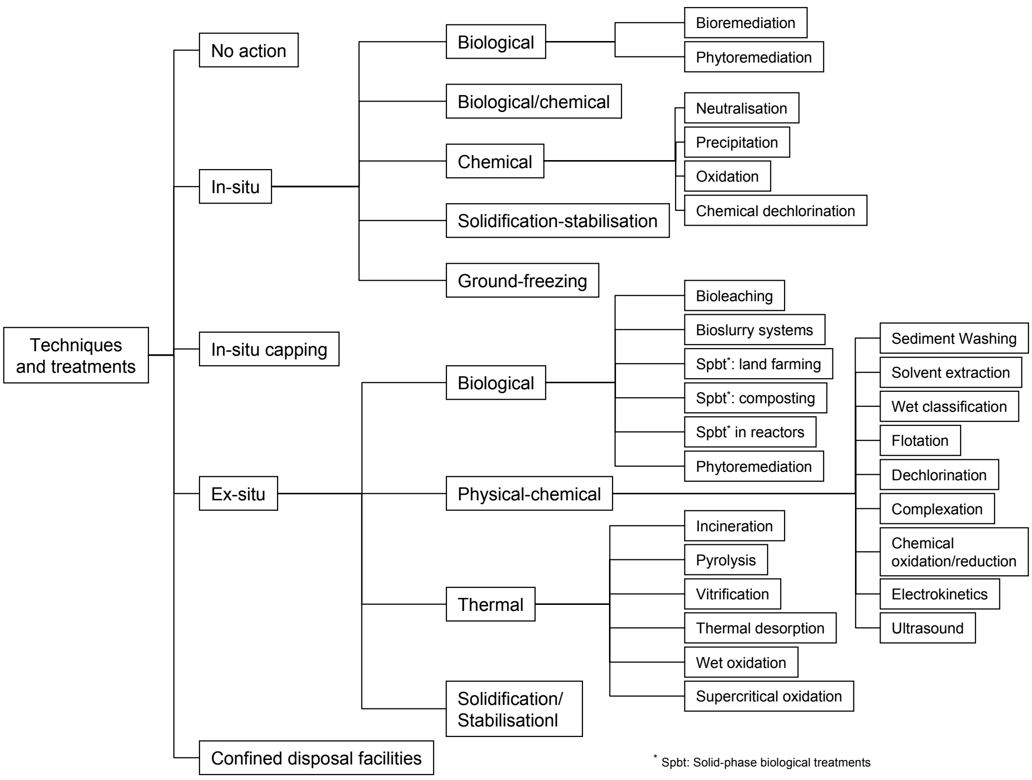

FIGURE 2 - Synthesis of techniques and treatments for the management of contaminated sediments or dredged material.

MANAGEMENT OPTIONS

No action: monitored natural recovery

Natural recovery of contaminated sediments consists in a combination of simultaneous physical, chemical and biological processes which bury, retain or destroy contaminants, reducing their concentration and their associated risks [9, 10]. The natural processes which lead to these reductions are physical (sedimentation, advection, diffusion, dilution, dispersion, bioturbation, volatilization); biological (biodegradation, biotransformation, phytoremediation, biological stabilisation); and chemical (oxidation/reduction and sorption) [4, 11, 12].

Some factors like contaminants present in the sediments, the extension of the contaminated area and its hydrodynamic, the time required or its economic feasibility [6, 10] have to be taken into account when natural recovery is considered as a solution. However, the option of no action with the hope of the natural remediation of the site is only recommended when risks are low and, consequently, the application of aggressive techniques does not make sense because they may cause more environmental impact than contaminants do. For more details, [4] gives the conditions which a site should have and make it suitable for its natural recovery.

The advantages of no action are its low implementation cost, its non-invasive nature and the no need of contaminant removal, whereas the disadvantages are the persistence of contaminants in the site, the dispersion of contaminants which may cause their transport to other areas, the wrong feeling of the society of a lack of concern from stakeholders and the possible slowness of remediation [4, 10, 13]. However, this slowness can be minimised if a thin-layer placement of clean sediment is applied, but that should not be confused with in-situ capping since thin-layers are not designed to provide long-term isolation of contaminants.

Monitorisation and control of natural processes are essential because results may be different from expected, predictions of future effects of these processes are required, the cessation of pollutant sources has to be corroborated (if other case, natural recovery is not recommended) and the uncertainties inherent in these processes have to be evaluated [11].

In-situ capping

In-situ capping refers to the placement of a subaqueous covering or cap of clean material (usually sand) over contaminated sediment that remains in place, in order to prevent contact between sediment contaminants and the overlying surface water and benthic communities [4,14]. Several authors [8, 13, 15, 16] only consider capping as a technique of confinement, but not as a treatment.

In-situ capping must fulfill three primary functions: physical isolation of contaminated sediments from the benthic environment; stabilisation of contaminated material to prevent resuspension and transport to other sites; and reduction of the flux of dissolved contaminants into the water column [17]. Capping projects should involve an evaluation of: site conditions (physical environment, sediment characteristics, waterways uses and infrastructure, habitat alteration); material used, thickness and number of caps; technique and equipment selection; monitorisation and management program; and costs [17-19].

Apart from being a technique of low-cost and low-technology [20], the main advantages of capping are the promptness with which the exposure of biota to contaminants is reduced and the less risk associated with dispersion and volatilization of contaminants during construction comparing capping with dredging or excavation. On the other hand, the most important disadvantage is the contaminant exposure or dispersion in case cap alters or contaminants move through the cap significantly [4], although natural attenuation processes continue acting after sediments are covered [14]. The conditions which make it possible that in-situ capping is a recommended solution are summarised in [4, 21].

In-situ treatments

In-situ treatments involve the biological, chemical or physical remediation of contaminated sediment in place [4], that is, sediments are treated without dredging [22]. The application of in-situ treatments to remediate contaminated sediments is an option that, although it has been considered as potentially viable from the technical point of view and beneficial in economic terms, has not been developed widely, with few projects of real cases carried out. This is due mainly to the difficulty to control the treatments conditions and due to the generation of secondary pollutants and negative environmental effects.

In-situ biological treatments

They are based on the use of living beings to eliminate (total or partially) contaminants of the sediments by transforming them to harmless, metabolic products or less toxic ones. Among the different biological treatments which are applied nowadays, two should be outstood: bioremediation and phytoremediation.

Bioremediation (also called bioreclamation) is defined as the process in which microbiological processes are used to degrade or transform contaminants to less toxic or non-toxic forms [4]. The microorganisms that decontaminate sediments can be inoculated (bioaugmentation) or biostimulated (e.g. air supply); it is reported that biostimulation alone and the coupling of both techniques have been demonstrated to produce the best results [23]. Both aerobic and anaerobic conditions are possible, but anaerobic degradation is slower and can treat less compounds.

Several references [15, 21, 24] point out several limitations of bioremediation, such as its restrictive application only to organic compounds, the difficulty to control the reaction and possible undesirable secondary effects, the lack of oxygen due to the high BOD (biological oxygen demand)

and its unsatisfactory efficacy in unfavourable environmental conditions (pH, temperature, absence of energy-rich electron or high concentrations of certain pollutants [23]). However, recent studies may make it possible to mitigate these limitations [25].

Phytoremediation (or phytoreclamation) is a biological treatment based on the use of plants to transfer, stabilise, concentrate and/or destroy organic and inorganic contaminants. Several mechanisms of phytoremediation might be distinguished [22, 26, 27]: phytostabilisation, in which plants limit sediment mobility, and consequently, immobilise contaminants and reduce their bioavailability; phytoextraction (by trees, grasses or crops), in which heavy metals are accumulated in plants; rhizofiltration, which uses plant roots to absorb pollutants; phytodegradation, in which plants degrade contaminants within their tissue; phytostimulation, which uses symbiotic soil microbes to degrade pollutants (mainly, polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs)) on the plant roots; and phytovolatilisation, with which contaminants are transformed into volatile compounds and released into the atmosphere.

Nevertheless, there are many restrictions to the application of phytoremediation [24]: the difficulty to control the process and its secondary effects, the limited knowledge about long-term treatment, the depth of the sediment conditioned by the used plants and the lack of effectiveness if the concentrations of contaminants are so high that they are toxic to plants.

In-situ chemical treatments

These are the treatments which use chemical products to treat in-situ the contaminants in sediments. Different chemical treatments-neutralisation, precipitation, oxidation and chemical dechlorination- are summarised in Table 1 [21].

The main disadvantage of these techniques is that they can produce secondary contaminants due to the toxic agents of the treatment or as a result of the induced reactions. For this reason, in-situ chemical treatments are only applicable if the contaminated area can be isolated during the process, which represents an obstacle. Another important problem is the difficulty to guarantee the complete mixing of reactants with contaminated sediment.

In-situ biological/chemical treatments

These treatments consist in the injection of microorganisms and/or chemical products to the sediments in order to start or intensify the degradation of toxic contaminants. As a result of the slowness with which microorganisms degrade contaminants naturally, microorganisms and/or chemical products, such as oxidising agents or nutrients, are added to stimulate this degradation. Examples of the application of these treatments have been developed in Salem (Massachusetts, USA), where calcium nitrate was injected to stimulate PAH biodegradation [15], and in coastal sediments of the Cantabrian Sea (Spain) contaminated with hydrocarbons by the Prestige fuel oil spill [28].

In-situ solidification/stabilisation treatments

These techniques involve the addition of binders (cement, lime) to encapsulate contaminated sediments and/or transform them into less toxic, soluble or removable compounds. The physical/chemical changes in sediments pursue a reduction of the contaminant spread due to leaching, erosion or dispersion [29]. Solidification/stabilisation is mainly used to treat metals because there are few destructive techniques available for metals, while organic contaminants are less stable and biodegradable [13]. However, the addition of activated carbon can be a potentially attractive method for in-situ treatment of organic contaminants, since recent results suggest that adding activated carbon reduces PCBs and PAHs availability to the aqueous phase [30, 31]. The use of traditional binders could raise the sediment pH and change the metals speciation, turning some metals into more soluble forms [15].

TABLE 1 - In-situ chemical treatments.

Treatments	Used reactants	Contaminants to treat	Potential problems
Neutralisation	-Weak acids and bases -$CaCO_3$, Na_2CO_3 or $NaHCO_3$	-Acids and bases	-Toxicity to benthic organisms sensitive to pH. -The use of $Fe_2(SO_4)_3$ under aerobic conditions may produce iron hydroxides which may remove heavy metals from water, and the gills of fish of the bottom of the sea can get blocked.
Precipitation	-Calcium, iron and sodium sulfides	-Inorganic cations and anions	-Possible formation of H_2S gas; this probability increases when sulfide and metal reactivities decrease. -Effective only under anaerobic conditions, because aerobic conditions may produce an oxidation which leads to more soluble forms.
Oxidation	-O_2 and/or O_3 and H_2O_2	-Organic compounds, but it is not suitable for very chlorinated or nitroaromatic compounds	-Oxidation may cause more mobile degradation products. -Both O_3 and H_2O_2 can react with organic compounds of the water column or of the sediments which are not an objective of the treatment, so its effectiveness comes down. -Compounds adsorbed in sediments may be difficult to oxidise
Chemical dechlorination	-Polyethylenglycol and KOH	-Very chlorinated organic compounds (e.g. PCBs, dioxins...)	-Degradation depends on temperature, and this process might be too slow at room temperature.

The inability of these processes to eliminate or destroy contaminants, erosion and the difficulty to adjust solidification mixture compositions are their most relevant disadvantages [15, 32]. Anyway, solidification/stabilisation treatments are usually applied as ex-situ owing to the last limitation exposed.

Ground freezing

"Ground freezing" is an in-situ treatment which avoids the dispersion of contaminants in sediments due to their freezing. The process consists in introducing refrigeration probes, which are chilled by a portable refrigeration unit, into the sediments during short time intervals [21]. Therefore, ice crystals grow until their coalescence and they make a wall of frozen sediment.

The main limitations are its high costs due to energetic requirements and the slowness of freezing processes since each probe is only able to freeze a small area (less than 0,5m in diameter). However, the future feasibility of its application can be for dredging, because this technique minimise contaminant dispersion during the process [33].

Ex-situ treatments

Ex-situ treatments are applied to remediate contaminated sediments which have been removed from the aquatic environments and constitute dredged material. Generally, more than one technique is usually used to decontaminate dredged material, applying several technologies in consecutive steps, which is called train treatments [8].

Dredging for cleanup purposes has been considered for some time as a primary means for managing contaminated sediment [17]. However, dredging has an important impact in the environment, because it disturbs benthic community and may affect structure, and species diversity and richness [34].

A great number of ex-situ technologies require a pretreatment. There are two main reasons which justify the need of a pretreatment: conditioning the material to meet the chemical and physical requirements for treatment or disposal; and reducing the volume and/or weight of sediment that requires transport, treatment or restricted disposal. Dewatering and physical or size separation are the most frequent processes used in pretreatment [4, 24, 32].

The residues produced in each technique must be treated and properly managed since they can contain part of the contaminants present in the dredged material. Its technical, economic and regulatory requirements should be taken into account owing to its possible impact in the global considerations of the general treatment [4]. Ex-situ treatments, classified into biological, physical-chemical, thermal and solidification/stabilisation categories, are described below briefly:

Biological treatments

The majority of contaminants remediated by living beings in these techniques are organics, whereas heavy metals can only be treated using bioleaching and phytoremediation. The main ideas of these treatments are stated below:

Bioleaching allows the remediation of sediments which contain heavy metals by means of their mobilisation by several types of acidophilic bacteria (among which the genus *Thiobacillus* excels) that oxidise reduced sulphur compounds to sulphuric acid [35-38]. An external reduced sulphur source has to be added in case no sulphides are present in the sediment. Bioleaching can be performed either in sediment-slurry systems or by heap leaching [22]. However, freshly dredged sediment is nearly impermeable, unsuitable for solid-bed bioleaching, and therefore needs preliminary conditioning, which is accelerated by the presence of reed canary grass (*Phalaris arundinacea*) [39].

Bioslurry systems are controlled in a reactor where contaminated sediments are treated in the form of slurry with low content of solids. Sediments are mixed with water in a predetermined concentration which depends on contaminants levels, biodegradation velocity and physical nature of sediments. It is suitable for many types of organic contaminants: VOCs, PAHs, PCBs and pesticides [40]. Microorganisms which biodegrade contaminants are already present in the sediment or can be added during the biodegradation processes. Dewatering is required after the technique and the wastewater resultant should be treated.

Land farming (a solid-phase biological treatment) is a method in which partially dewatered sediment is spread out in a relatively thin layer (15-30cm) on an especially built site [22]. The addition of nutrients and the plough of the layer to maintain an adequate aeration are used to stimulate the microbiological activity. The process can be developed in a greenhouse: despite its higher cost, it is more effective (three months in a greenhouse are equivalent to a year) [24]. As all solid-phase biological treatments, land farming can be applied to organic contaminants which may be biodegraded under aerobic conditions; however, it does not remediate inorganics and it has difficulties to treat high molecular weight PAHs and highly chlorinated PCBs [40].

Composting (a solid-phase biological treatment) is a microbiological treatment system in which partially dewatered sediment, polluted with organic contaminants, is intensively mixed with bulky natural organic materials (which improves the permeability of the pile and supplies nutrients necessary for the microbiological aerobic biodegradation) and then placed in an aerated stockpile [22]. However, this also leads to some disadvantages, like the rise in volume due to the organic material added or the vast area which is usually required to develop this technique [40].

Solid-phase biological treatments in reactors is also known as composting in reactor. Contaminated sediments are placed into bioreactors, with ventilation systems and leachate collection, during two weeks. It is the most expensive process of the solid-phase biological treatments [40].

Phytoremediation is a bioremediation process on the application of special plants to remove or destroy pollutants (such as heavy metals or PCBs) in sediments [22]. The modifications of this process have already been described in in-situ treatments. Furthermore, phytoremediation is able to stimulate the dewatering of sediment because of its extraction and evaporation. In order to assess the success of the treatment, three points must be considered: physical and chemical characteristics of the sediment, plant exposure effects and contaminant reduction effectiveness [26]. Depending on the pollutant that is going to be treated, specific plants are recommended [41]. Phytoremediation should be applied more than once or combined, sequentially or concurrently, with other treatments (such as bioremediation or chemoremediation) in case two or more contaminants are present in the sediments.

Physical-chemical treatments

Physical treatments are designed to reduce the volume of the contaminated material by means of separating the most contaminated fractions of a sediment from the remainder. Thus, only the polluted fraction will be treated or disposed in a CDF (confined disposal facility), whereas the less contaminated, or clean, fraction may be suitable for a beneficial use [42]. On the other hand, chemical techniques tend to immobilise or transform contaminants into less hazardous compounds. These treatments are summarised below:

Sediment washing involves the extraction of contaminants using only water or water in combination with additives (surfactants, acids, bases, chelating agents) as solvents [32], and, in this way, transferring the contaminants from the sediments to the wash solution [37]. A clean fraction of coarse particles (sand and gravel) is separated from another fraction of fine particles (clay and silt) which contains the contaminants concentrated [21]. Sediment washing is suitable to treat organics (PCBs, PAHs, hydrocarbons) as well as heavy metals (weaker bound metals in the form of hydroxides, oxides and carbonates [37]). However, this technique does not destroy contaminants: it only reduces the volume of contaminated material, which should be managed afterwards. It is limited to dredged material which is permeable, with little humus and with particles no greater than 0.5cm [21, 40]. Sediment washing may also generate large volumes of contaminated wastewater which must be treated prior to discharge [4]. Several steps of washing are necessary to achieve a high efficacy.

Solvent extraction is similar to sediment washing, with the exception of using an organic solvent (hexane, methanol, kerosene) instead of water. The process has two steps: the mixing of dredged material and solvents, and the separation of the sediment particles. The solvent (extracting agent) is treated to remove and concentrate contaminants and thus, make its reuse (in a new extraction) possible [22]. It is adequate for dredged material that contains organics (PCBs, PAHs, VOCs, hydrocarbons) but not inorganics. Volume reductions of 98-99% have been obtained, but a control of the solvent must be done due to its toxicity to organisms [40].

Supercritical extraction (SCE) can be included as a subcategory of solvent extraction. This technology basically employs green supercritical fluids (SCFs) like carbon dioxide (SCCD) and water (SCW) for the extraction of VOCs and persistent organic pollutants (POPs). A pretreatment is usually required to remove debris and to adjust pH and chemical composition before the placement of the dredged material in a pressure vessel, where contaminants are extracted with a recirculated stream of SCFs [43].

Wet classification entails a process of two main steps: the mixing of sediments to disintegrate agglomerates of dredged material particles, and mechanical separation (by means of a hydrocyclone, centrifuge or up flow column) between polluted and non-polluted particles. It is applicable when contaminants are specifically bound to an easily-separable sediment fraction [22].

Flotation is based on the separation owing to density differences and surface properties of solids (their tendency to surround themselves with air -and float- or with water –and sink). Both inorganic and organic contaminants are suitable to be treated. A conditioner agent might be necessary and flotation is not adequate if clay contents are high [24].

Dechlorination consists in the heating (110-340ºC) and mixing of the contaminated sediments with chemical reagents which contain potassium or sodium hydroxide (KOH and NaOH) in a batch reactor for several hours [24, 32]. Halogenated contaminants decompose into less toxic glycolethers and water-soluble chloride compounds. It treats aromatic organic contaminants (especially PCBs, chlorobenzenes and dioxins), but it can produce more toxic compound if dehalogenation is incomplete or inefficient. It is not effective for metals, PAHs, or if there are high levels of contaminants, more than 20% of water content, or pH is lower than 2 [21, 40].

Complexation or chelation involves the addition of chemical agents with complexing properties (EDTA, NTA or ammonium acetate) to sediments so that water-soluble ion complexes are formed and consequently, metals are immobilised [24]. pH is one of the most important parameters that affects the treatment process [32]. Despite its low cost, it is not effective for organic compounds or sediments with high contents of clay and/or silt [40].

Chemical oxidation/reduction uses chemical additives to convert organic contaminants into immobile or less toxic forms. Some contaminants (like PCBs or dioxins) also require ultraviolet radiation. Chemicals such as O_3, H_2O_2, peroxone (a combination of O_3 and H_2O_2), $KMnO_4$, $Ca(NO_3)_2$ and O_2 are the most frequent oxidising agents employed. The use of O_3, H_2O_2 and peroxone has come to be known as advanced oxidation processes [32]. It is useful for organic compounds, excepting some PAHs and highly halogenated contaminants. The main disadvantages of this technique are its high cost, the risk of an incomplete oxidation which may produce more toxic contaminants and the necessary dewatering (before and after the treatment) [40].

Electrokinetic processes involve passing a low intensity electric current between a cathode and an anode imbedded in the contaminated sediments. Ions and small charged particle, besides water, are transported between the electrodes, where they are concentrated and removed. An electric gradient initiates movement by electromigration (charged chemicals movement), electro-osmosis (movement of fluid), electrophoresis (charged particle movement) and electrolysis (chemical reactions due to electric field) [37]. The temperature of the sediment layer increases slowly during the electrokinetic remediation, which can be beneficial in case that also organic biodegradable pollutants have to be removed from the sediment [22]. Some examples to remove heavy metals [44, 45] and organics [46] using this technique have been referenced.

Ultrasound is a recent technology which tries to decontaminate dredged material using ultrasound coupled with vacuum pressure. Meegoda and Perera [47] and Meegoda and Veerawat [48] managed to remove successfully both inorganic (chromium) and organic (p-terphenyl) contaminants from dredged sediments obtained from NY/NJ harbour by means of two processes designed to separate and to treat both coarse and fine fractions of the sediments; however, they determined that the clay fraction is not able to be treated by this technology in the case of chromium.

Thermal treatments

Incineration consists in the destruction of contaminants due to their combustion using high temperatures (800-1200ºC), in presence of oxygen. Two steps usually form this process: 1) combustion at high temperature in a rotary furnace; 2) post combustion in a specific chamber [24]. Incineration is especially adequate to treat sediments with large amounts of mineral oil (in this way, the energy requirement is relatively low) or sediments contaminated with toxic organic compounds such as PAHs, PCBs, pesticides, dioxins or chlorinated hydrocarbons. Heavy metals (excepting mercury) are in general immobilised in the solid matrix [22]. The main disadvantages of incineration are its high cost because of energy consumption and the need of treating the gases (in which dioxins and furans are present); ash production as a waste, which contains high concentrations of heavy metals and should be managed/ treated; and its low water content requirements, which may imply a pre-treatment step [40].

Pyrolysis destroys organic material using high temperatures, in absence of oxygen. This permits the separation of the material into an organic fraction (gas) and a carbonised inorganic one (salts, metals, particles). It is used to treat high organic concentrations (e.g. semivolatiles and pesticides) which are not suitable for a conventional incineration [40]. A pyrolysis system consists in a main combustion chamber (540-760ºC) and the resultant gaseous products may be destroyed into a secondary chamber (1200ºC) or collected (e.g. on a carbon bed). The most outstanding limitation of pyrolysis is that its costs are higher than the costs of incineration [32].

Vitrification is a thermal process in which the sediment is heated to such a temperature (usually more than 1600ºC) that the inorganic compounds in the sediment will melt and the organic pollutants will be completely destroyed (although an after-burning process is often required) [22]. In this way, sediments are converted into a vitreous mass (slag), which can be suitable as building material or possess, with the proper modifications, cementations properties. It can treat all type of contaminants, so it is considered one of the most effective techniques to treat dredged material. Although vitrification is one of the most expensive technologies [37], it can compete when a treatment chain is required, because vitrification can act as a stand alone technology [40]; the possibility of selling a useful glass product can also contribute to make this process economically viable [37]. However, vitrification is not feasible for sediments with high levels of electrically conducting metals and molten products may require months to years to cool [32].

Thermal desorption heats partially-dewatered-sediment in a closed reactor system to such a temperature (100-500ºC) that organic pollutants (volatile, VOCs, and semivolatile organic compounds, SVOCs) will vaporise from the sediment together with water [22]. In general, it is applied to treat contaminants with a boiling point below 500ºC, especially VOCs and SVOCs, but also some volatile metals. However, contaminants are transferred to gaseous phase but they are not destroyed, so this phase must be treated before its emission. For instance, VOCs can be destroyed in a secondary combustion chamber at a higher temperature, recovered by condensation or adsorbed into activated carbon [21].

Wet oxidation consists in a process which occurs in the water phase at high temperatures and high pressures but below the supercritical temperature and pressure of water. The type of pollutants (mainly petroleum hydrocarbons, phenolic compounds and PAHs) and conditions of the process, like temperature or the residence time in the reactor, determine the degree of oxidation [22].

Supercritical oxidation is a modification of the previous technique in which temperature and pressure are above the critical point of water. At these conditions, oxygen and toxic organic compound solubility in the water phase is higher, so a major degree of oxidation is obtained [22], including sediments highly contaminated with PCBs [49].

Solidification/Stabilisation (S/S) treatments

S/S alters the physical and/or chemical characteristics of the sediment through the addition of binders (cements, limes, pozzolans, thermoplastics, clays or silicates). Physical alteration implies the reduction of the accessibility of the contaminants to water because of their immobilisation by cement matrix, whereas chemical stabilisation minimises the solubility of metals primarily through the control of pH and alkalinity [4]. It is widely applied to treat metals, such as arsenic, lead, mercury, cadmium, copper or zinc, owing to the few destructive techniques available for these contaminants [37]. Nevertheless, chemical stabilisation of organic compounds might also be feasible [4]. The most important disadvantages are the significant raise of volume and the possible volatilisation of organics due to the heat generated during the reaction [40].

Confined disposal facilities (CDFs)

CDFs are engineered structures enclosed by dikes and designed to provide the required storage volume for dredged material and to meet the required suspended solids in effluent released from the facility [50].

CDFs should be the end of the management cycle only for these contaminated sediments or dredged material for which other options that are in previous steps in the management hierarchy (like beneficial uses, after a possible treatment) are not viable. They have been widely used for navigational dredging projects and some combined navigational/environmental dredging projects but are less common for environmental dredging sites [4]. Many CDF field experiences are summarised in [50]. Nevertheless, as land development and acquisition costs continue to rise, there is a growing shortage of CDF storage capacity. Several options can be considered to increase capacity, including restricted disposal (storage of only the most contaminated sediments), dredged material dewatering and densification, and, more recently, reclamation and reuse of materials from the CDF [51].

The design of CDFs has to take into account the losses of contaminants and try to eliminate them [4, 9, 21]. These losses may occur by leaching through the bottom of the CDF, filtration through dikes or walls, volatilisation to the atmosphere (if CDF is not closed), or the possible consumption of contaminants by plants and animals which live near the CDF.

BENEFICIAL USES OF DREDGED MATERIAL

Nowadays, it is believed that considering dredged material as a natural resource and not as a waste may solve some problems of dredged material management. In this way, not only is space in confined disposal facilities (CDF) saved but beneficial uses of dredged material might also allow the economic feasibility of some processes.

It is necessary to evaluate several aspects before developing a project to valorise dredged material [52]. Firstly, assessing contaminant status of materials is essential to know if they are or not suitable. Moderate and slightly contaminated sediments can be appropriate for many beneficial uses (it is probable that a pre-treatment or a clean material coating will be needed), whereas it is unlikely that highly contaminated materials are adequate for the vast majority of applications, with few exceptions such as construction products (ceramics, aggregates…). However, these highly contaminated sediments can be recovered if they are treated properly. In general, suitability for a specific use depends on the impact on sensitive natural resources and the compatibility between the properties of the dredged material and those of the intended beneficial use [53]. Guidance on the nature and types of physical, engineering, chemical and biological characterization tests appropriate for determining the potential for beneficial uses of dredged material in aquatic, wetland and upland environments can be found in [54]; practical cases of the application of these characterisation tests are described in [55]. These tests should be done periodically when dredged material comes from ongoing activities and their characteristics might change. Moreover, site selection must be considered and its choice is not independent from the use selected [52].

After use and site selection, it is important that there are no technical (like water depth or access), legal or environmental impediments. In many cases, an Environmental Impact Assessment (EIA) is mandatory for projects where beneficial use of dredged material is planned [56]. Finally, a complete project should develop a commercialisation plan and examine the costs (compared to disposal of dredged material) and the benefits of using dredged material, not only economic but also environmental or social ones.

Beneficial uses are not only possible for "new" dredged material, but they can also be applied to recover the sediments stored in CDFs. In this way, [51, 57, 58] state the relevant aspects to valorise material contained in CDFs.

In 1992, PIANC edited a guide about beneficial uses of dredged material [59] in which three categories of uses are identified: engineered uses, agricultural and product uses, and environmental enhancement. This classification has also been included in the guidelines of the OSPAR Convention for the management of dredged material [7]. Another classification was proposed in 1996 [60]: construction, coast protection, agriculture, horticulture and forestry, amenity, habitat and capping. A summary of the main uses found at bibliography [8, 24, 52, 61] grouped in the three categories proposed by PIANC is stated below and shown in Figure 3:

a) Engineered uses

i) Land creation: filling, raising and protecting an area which is otherwise periodically or permanently submerged.

ii) Land improvement: increasing the quality of the land which is not suitable for its planned use or because of its insufficient height which may produce floods.

iii) Fills and replacements: use as filling material in abandoned mines, obsolete canals, etc. Substitution of low quality soils or improvement of their properties so that they can fulfill requirements to be used in construction [62].

iv) Submerged berm creation: use of the dredged material to build submerged berms or embankments, which are utilised to modify shoreline in order to improve beach stability, to alter wave direction and to change the speed or the direction of local sediment transport. Swell modifications with berms can also improve recreational uses like swimming, surfing, etc.

v) Shore protection: it includes beach nourishment and dike or berm construction.

vi) Beach nourishment: creation of new beaches for recreational purposes or replacement of material which is swept by swell and it is not recovered naturally.

vii) Capping: use of dredged material as a coating in in-situ capping techniques.

b) Agricultural and product uses

i) Construction materials: use of sediments as substitutes of raw materials in the manufacturing of construction materials such as bricks, tiles, bituminous mixtures, mortar or cement [63, 64].

ii) Aquaculture: use of dredged material in the construction of containment structures or ponds of aquaculture.

iii) Topsoil: dredged material is an excellent topsoil to improve soils with agricultural purposes. For food products, clean material is needed, whereas for other agricultural uses, permitted contamination levels depend on the type of cultivation and its final use [65, 66].

c) Environmental enhancement

i) Creation or recovery of habitats for fauna: creation of nesting islands for water birds-dredged material may provide substratum for nests [67].

ii) Fisheries improvements: the proper placing of dredged material can improve fishing resources-mounds of dredged material may be useful as a shelter for fish and, moreover, plants may grow on them, offering a better habitat recovery for fish.

iii) Marsh recovery: it may be applied as a barrier against wind to permit plant growth or to restore shores from erosion [68].

Figure 3 shows a scheme of the beneficial uses of dredged material.

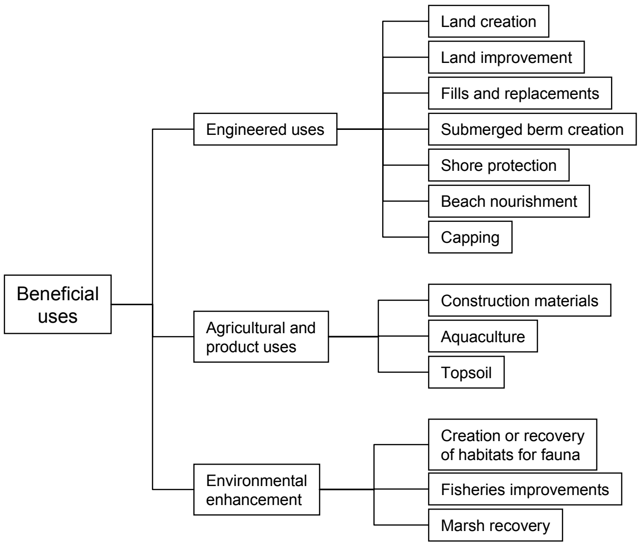

FIGURE 3 - Scheme of the beneficial uses of dredged material.

Beneficial use of dredged material is the aim of many processes or technologies. Table 2 shows some cases of technologies or processes, developed at full, commercial or demonstration scale, which valorise dredged material.

As already stated, the type of material determines the beneficial uses dredged sediments have. In this way, rock is a valuable construction material and whether or not it can be used economically depends on its quantity and size. Gravel and sand are generally considered the most valuable material for use that a dredging project can provide due to their many engineered uses and frequently, without the need to sort (or pre-wash) the material prior to being used [8]. Clay and silt are the most common acquired from dredging: consolidated clay can find more engineered uses than soft clay, whereas silt in particular is more suitable for agricultural/horticultural purposes and all forms of habitat creation and/or enhancement [53].

Table 3, which is based on [8, 52], indicates possible applications of dredged material depending on the type of predominant material: rocks, gravel and sand, consolidated clay, silt or soft clay, and mixture of previous.

TABLE 2 - Examples of technologies or processes (tested at full, commercial or demonstration scale) applied to dredged material from which beneficial uses are obtained.

Technology	Marketed/ Manufactured by	Application sites	Fundamentals of the technology	Results over contaminants	Beneficial uses
Cement-Lock® [69-72]	Gas Technology Institute/ Endesco Services, Inc. of Des Plaines, IL	New York, New Jersey and Michigan (USA)	Rotary kiln natural gas-fired thermo-chemical process	Organics thermally oxidised; heavy metals immobilised in cement matrix	Glassy granular product ("Ecomelt") that can be used as a pozzolan in commercial-grade blended cement for general construction, soil stabilisation and solidification
METHA (MEchanical Treatment and dewatering of HArbour-sediments) [69,73]	Industrial METHA plant facility	Hamburg (Germany)	Two-step physical separation. The products of separation (sand, fine sand and silt) are then dewatered	Concentration of the contaminants in the different fractions.	Use of the separated dewatered silt as a sealing material. Back-filling of harbour basins. Use as clay replacement in the production of bricks (in Hanseaten-Stein facility) or in dike works
Hanseaten-Stein brick production [69,73,74]	Hanseaten-Stein Ziegelei GmbH	Hamburg and Bremen (Germany)	Mixing of the sediments with crushed bricks and natural clay. Drying of the mixture and ceramisation process	Organics thermally oxidised; metals converted to stable immobilised compounds or volatilised	Commercial/residential construction: bricks for use in the building industry
Plasma Vitrification [69-71,75]	Global Plasma Systems Corporation, Washington, DC, in partnership with Westinghouse Plasma Corporation	Port of New York/ New Jersey (USA)	High temperature plasma arc vitrification process	Organics thermally oxidised; heavy metals immobilised in glassy matrix	Glass aggregate that can be used as a raw material in the manufacture of architectural tile, glass fibre, blasting grit, aggregate and glass cullet
Novosol® process [76]	Solvay Company	Dunkirk (France)	Chemical inertisation followed by thermal process	Heavy metals chemically inertised; organics thermally eliminated	Introduction of treated sediment aggregate in cement-based materials
Flowable Fill [69]	Pohlman Materials Recovery, Cary, IL	New York (USA)	Non thermal, mixing process using chemical additives	Contaminant chemical stabilisation and incorporation in physical matrix	Replacement for compacted fill
BioGenesis Sediment Washing [69-71,77,78]	BioGenesis Enterprises, Inc. and Roy F. Weston, Inc.	Port of New York/ New Jersey (USA). Venice (Italy)	Combination of mechanical and chemical processes using high-pressure water jets and chemical additives	Organics are oxidised; metals are removed	Manufactured soil by addition of bulking materials for fill, cover or topsoil applications (in NY/NJ). Raw product (decontaminated silt and clay) that can be used for brick manufacturing (in Venice)
Solidification/ Stabilisation [69]	OENJ Cherokee Corporation, Bayonne, NJ	Elizabeth (USA)	Non thermal, mixing process using chemical additives	Incorporated in physical matrix product	Compacted fill and capping
Rotary kiln-JCI/Upcycle [71,79]	Jay Cashman, Inc. of Boston, MA and Upcycle Aggregates of New Providence, NJ	Port of New York/ New Jersey (USA)	Belt press dewatering. High-temperature rotary kiln thermal process	Organics thermally destroyed; metals immobilised in the mineral matrix	Feedstock for the manufacture of lightweight aggregate
HREG process [71,80]	Harbor Resource Environmental Group, Inc. (formerly NUI Environmental Group)	Port of New York/ New Jersey (USA)	Chemical oxidation with $KMnO_4$ and superionised water. Dewatering and addition of cement	Organics oxidised; metals stabilised in the matrix	Manufactured soil that can be used in construction and remedial projects.

TABLE 3 - Beneficial uses of dredged sediments depending on material.

	Beneficial use options		
Dredged material sediment type	**Engineered uses**	**Agricultural/Product uses**	**Environmental enhancements**
Rock	a,b,c,d,e	h	k,l
Gravel and sand	a,b,c,d,e,f,g	h	k,l
Consolidated clay	a,b,c,d,g	h,i	k,l,m
Silt/Soft clay	a,b	h,i,j	k,l,m
Mixture	a,b,c,e,g	h,i,j	k,l,m

a:land creation; b:land improvement; c:berm creation; d:shore protection; e:replacement fill; f:beach nourishment; g:capping; h:construction materials; i:aquaculture; j:topsoil; k:wildlife habitats; l:fisheries improvement; m:wetland restoration

UTILISATION OF CONTAMINATED SEDIMENTS FROM CANTABRIAN ESTUARIES FOR CERAMIC MATERIALS: A CASE STUDY

A summary of the previous results obtained by our research group about the use of contaminated sediments, from the Santander Bay and Suances Estuary (Spain), as alternative raw materials in the manufacture of traditional clay based ceramic materials is presented.

Sediments from Cantabrian estuaries (Northern Spain) have been previously characterised by different chemical and ecotoxicological parameters showing that sediments contain significant concentrations of heavy metals and organic pollutants derived from intensive industrial, agricultural and urban activities [81-83]. The most polluted sediment samples S1, S2 and S3 have been studied in order to evaluate landfill disposal and introduction into ceramic matrices as alternatives to sediment management [84]. Physico-chemical and environmental characterisation of these sediments has been carried out (Figure 4). The evolution of the physical parameters, density, porosity, specific surface and water content with particle size, showed that the studied sediments can be used as clay replacement in ceramic processes. Availability and mobility concentrations of inorganic parameters obtained by leaching tests suggest that only a small part of the total concentrations are bioavailable for all samples except for zinc, which has very high availability. From the mobility results, it can be concluded that all sediments can be disposed of in a landfill for non hazardous waste, but should be pretreated if their disposal is to be in a landfill for inert waste, due to their high lead mobility.

The work of Romero et al. [85] has proved that it is possible to obtain dense sintered compacts from sediments S1, S2 and S3. Water absorption and mechanical properties showed that the specimens fired at their respective optimum sintering temperature met the requirement values established by different European Standards for high density traditional bricks, covering ceramic tiles as well as for the more demanding criteria to high density clinker bricks and pavement ceramic tiles. Based on these preliminary analyses, the investigation conclude that the studied marine sediments can be used as secondary raw materials for the production of ceramic bricks, floor and wall tiles by powder technology [85]. The ceramic bodies obtained from the contaminated marine sediments S1, S2 and S3 have been fully characterised in terms of both phase evolution during firing and microstructure at the optimum sintering temperature. The mineralogical evolution examined revealed that quartz, which is a main crystalline phase in the original marine sediments, remains as a main phase in the sintered bodies. In addition, a glassy phase and new crystalline phases appear as result of different chemical reactions during firing. Scanning electron microscopy (SEM) showed a homogeneous microstructure composed by pores, crystals and a ceramic matrix [86].

As shown in Figure 4, the sediments S1, S2 and S3 with physico-chemical characteristics similar to clay, have been used as clay replacements in bricks, obtaining a final marketable product. The sintering process leads to a ceramic material with similar density and porosity, but smaller specific surface area than the commercial bricks which could have an effect on the technical properties as water absorption as well as on the leaching characteristics of the final product [87]. The technical parameter, water absorption, fulfils the proposed limit for Spanish standards (15%) in spite of the quantity of sediment in the brick that increases this technical value.

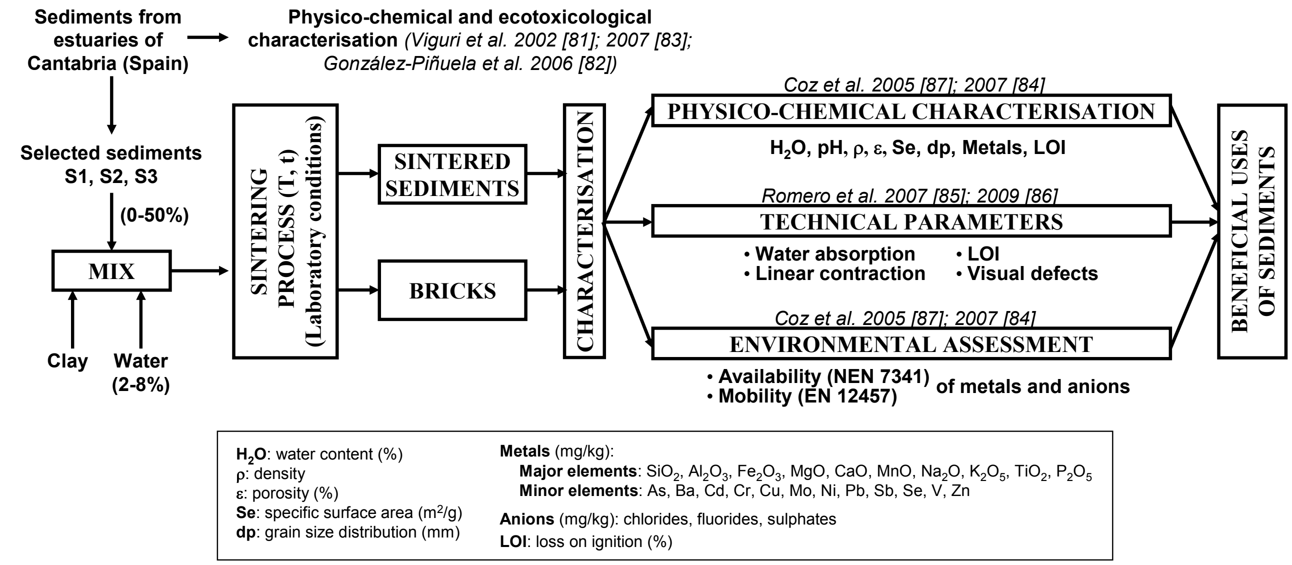

FIGURE 4 - Framework of the research activities to the use of contaminated sediments in the ceramics production.

The availability and mobility of the anions increase with the percentage of sediment used in the brick, showing linear expressions between both parameters in all cases. The results of mobility are due to the solubility and not the availability of the corresponding anion. A comparison of the L/S=2 and L/S=10 ratio results and the regulated limits by the Council of the EU for inert waste have been performed. The arsenic limits are not fulfilled for the commercial brick (0% of sediment) and lead exceeds the limits in both the sediment and the bricks with high percentages of sediment (> 10%).

On the other hand, the effect of water content (between 2-8%) and sediment content (between 0-50%) on the technical characteristics density, porosity, LOI, water absorption and linear contraction, of fired clay products, have been studied. The results show linear correlations between additions and technical properties. The regulated values [88] to water absorption (max. 15%) and linear contraction (max. 8%) are fulfilled in all experiments; only the limit value of 15% LOI in bricks is exceeded at sediments additions higher than 30%.

The physico-chemical and environmental properties of the studied sediments, their sintering behaviour, and the technical and environmental properties of the final bricks obtained with sediment additions, lead to obtain a marketable product. This strategy gives the opportunity to use waste materials containing various organic or inorganic contaminants as admixtures together with common raw material to overcome environmental problems.

CONCLUSIONS

This work reports a wide variety of techniques and treatments that can be applied for the remediation of contaminated sediments and dredged material. The selection of the best option that leads to the sustainable management of the sediments of a site is not an easy task, and it should be based on the integration of technical, economic, social and environmental criteria. The involvement of stakeholders is of crucial importance to achieve successful results in the decision-making process of management and treatment alternatives. The development of sound methodological tools and clear environmental criteria is necessary as well to make these decision-making processes easier.

In any case, the utilisation for beneficial uses should be the preferred option for contaminated dredged material whose relocation in the aquatic environment is not possible, while confined disposal should be considered as the last option in the hierarchical sequence of alternatives (i.e. only for those contaminated sediments for which other options are not viable). Although the direct use of dredged sediments with high levels of pollutants is not normally appropriate, there are several examples of the application of treatments that finally allow the beneficial and commercial use of the material. In this sense, successful results such as the ones obtained by our research group about the use of contaminated sediments as substitute of raw materials in the ceramics production can promote a wider application of beneficial use options for dredged sediments.

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