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CONTAMINATED MARINE SEDIMENTS: WASTE OR RESOURCE? AN OVERVIEW OF TREATMENT TECHNOLOGIES*

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Abstract

The continuous stream of sediments dredged, from harbors and waterways, is a considerable environmental issue recognized worldwide. Every year about 200 million of m^3 of sediments are dredged only in Europe, over half contaminated and expensive to dispose of. In a vision of sustainability and of circular economy, the proper management of such sediments plays an important role. Therefore, the aim of this study was to critically revise the remediation technologies currently adopted for the suitable reuse, recycling and recovery of contaminated marine sediments (CMS). First of all, the description of the common technologies for the complete removal and/or immobilization of pollutants was realized. Subsequently, potentially technical solutions for marine sediments reuse were discussed. Such re-use or recycling is in line with the European Waste Hierarchy and generates positive environmental impacts. Finally, the research proposes a new approach to the "sediment issue", an eco-sustainable management where the contaminated marine sediments are viewed as a useful resource.

Keywords: beneficial reuse, contaminated marine sediments, treatment technologies

1. Introduction

Sediments are dredged, from harbors and waterways, for maintenance of navigation, environmental remediation, or both. Every year about 200 million of m3 of sediments are dredged only in Europe, over half contaminated and expensive to dispose of (SedNet, 2011). In fact, contaminated dredged sediments are considered as waste materials and only less than 5% of these is being treated for ultimate beneficial re-use, at present.

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Until the early 90s, in most cases the dredged sediments were disposed of at deeper seas or deposited on land (the less expensive method). Fortunately, this tendency has been changing in recent years. Conventions for the protection of the marine environment and some new European regulations concerning waste have been introduced to set guidelines for a proper disposal of dredged material into the sea and to avoid the traditional perception of these contaminated materials as waste, considering them as a commercially exploitable resource. In fact, the Protocol of the London Convention (IMO, 2009), for disposal at sea, and the European Waste Framework Directives (EU Directive, 2008), for disposal on land, demand sustainable management of sediments and waste minimization.

There is a growing impetus for considering dredged sediments as a resource rather than a waste. However, whether a beneficial use is possible, the Contaminated Marine Sediments (CMS) should be treated. In fact, the re-use of contaminated materials is possible when these possesses technical characteristics appropriate for its specific utilization and are environmentally acceptable in accordance to end-of-waste criteria.

This report proposes a critically review of remediation technologies currently adopted for sustainable management of CMS. Recent developments in this area were assessed through a literature search, and promising technologies were identified.

2. Contaminated marine sediments treatment technologies

Different treatment technologies are well known by worldwide experiences of sustainable management to CMS. In this section, some of these technologies are presented, with special attention to new emerging technologies employed in relevant case studies to attend the reuse goals (technical characteristics and environmentally acceptable).

2.1. Landfarming

Landfarming treatment is a full-scale technology in which the soil, climate, and biological activity interact to degrade, transform, and immobilize the contaminants.

Contaminated sediments are spread over lined beds in the upper soil zone or in biotreatment cells, and regularly turned over or tilled to aerate the mixture. The conditions of the sediments are controlled to optimize the rate of contaminant degradation. These include: moisture content (by irrigation or spraying), aeration (by tilling soil with predetermined frequency, the soil is mixed and aerated), pH (buffered near to neutral pH through adding crushed limestone or agricultural lime) and the addition of other amendments such as soil bulking agents and nutrients.

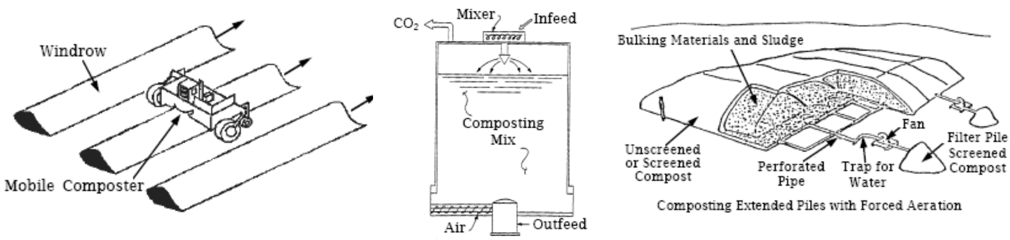
A landfarming treatment site should be managed correctly to prevent both on-site and off-site problems with ground water, surface water, air, or food chain contamination. Sufficient monitoring and environmental protections are required (NSW EPA, 2014).

2.2. Composting

Composting is a controlled biological process by which organic contaminants are broken down by indigenous microorganisms (under both aerobic and anaerobic conditions) to innocuous by-products, such as carbon dioxide and water. Maximum degradation efficiency is achieved through maintaining of thermophilic conditions (54 to 65 °C) and monitoring of water, oxygen, and nutrients.

Available composting techniques include: (i) aerated pile, where compost is formed into piles and aerated with blowers; (ii) windrowing, here compost is heaped in long piles known as windrows and periodically mixed with mobile equipment, such as a digger; (iii) closed reactor designs where compost is put into a reactor vessel where it is mixed and aerated (US ACE, 2014). These are shown in Fig. 1.

Volatilization of contaminants may be a concern during composting and may require controls, such as enclosures or air management system.



a) **b)** **c)**

Fig. 1. Composting techniques: (a) Windrow, (b) Closed reactor and (c) Aerated Static Pile. (US ACE, 2014)

2.3. Solidification/Stabilization

Chemical fixation and solidification, also commonly referred to as solidification/stabilization treatment (S/S), is a widely-used treatment process for the management and disposal of a broad range of waste materials, even those classified as hazardous.

The S/S techniques is based on adding chemical compounds to dredged material with two objectives: (i) chemical immobilization of the contaminants to reduce the leachability and bioavailability; (ii) stabilization to use the product as a construction material. This technique does not remove the contaminants from the dredged material, but they are transformed into a less mobile, and therefore less harmful species. In general, the target contaminant group for s/s treatment is mainly inorganic (Bortone and Palumbo, 2007).

The simplest form of treatment is with Portland cement. Further materials can be added, such as calcium aluminates, fly ashes, bentonite or other clays, phosphates, lime, oil residue and silicate fume. The additive used depends on the type of contaminants, water content and characteristics of the dredged material.

In the last years, innovative binders and mixtures (other than, or in combination with, cement) have been tested in pilot sites and in full scale, showing a significantly reduced of leachability of organics from stabilized contaminated sediment.

2.4. Sediments washing

Sediments washing is an effective technique to remove (by dissolving or suspending them in a water-based solution) a wide range of organic and inorganic contaminants from sediments.

The process involves some steps: a preliminary treatment for separation of metal parts (by magnetic belts), separation of the clay and silt particles from the sand fraction (using hydrocyclones and upstream classification), unit of gravel washing (coarse-fraction is cleaned by scrubbing and counter flow washing), sludge treatment (the sludge-fraction is further dewatered in a filter press) and process water treatment.

Surfactants, organic solvents, acids and bases or chelating agents may be used with water to effect separation of some contaminants. For example, the use of surfactants may be successful for removing organic compounds from sandy sediments.

One of the most promising sediment washing treatment using the BioGenesis technology (Fig. 2). The basic treatment train involve the separating the particles using high energy, mixing with oxidants, and finally separating the solids into organic and mineral fractions. The completely disaggregated mineral fractions are then mixed with suitable clean organic amendments, to create clean manufactured topsoil with the necessary nutrients added to promote plant growth. The organic and ultra-fine fractions, as well as the effluent, must be disposed of or treated off site.

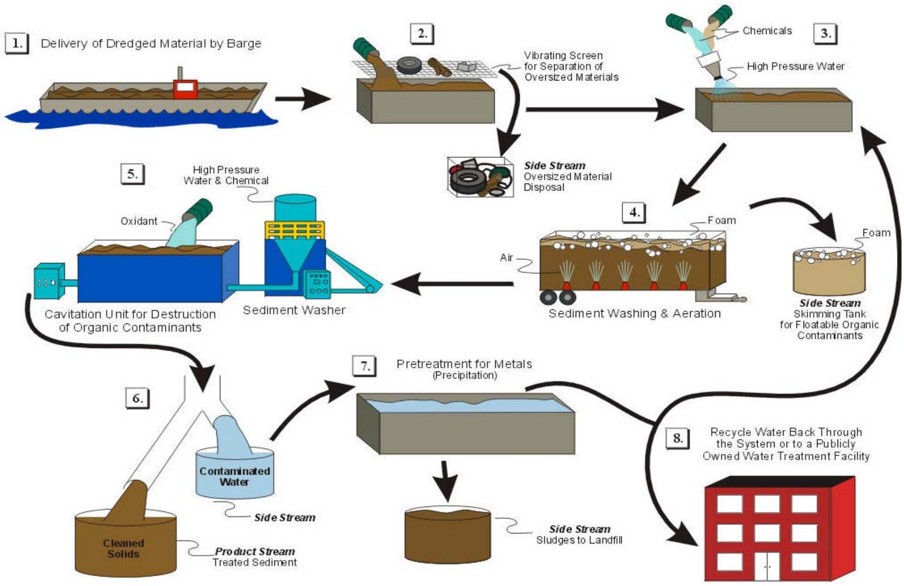


Fig. 2. BioGenesis sediment washing process (Stern et al., 2007)

2.4. Thermal desorption

Thermal desorption can be applied to treat sediments containing contaminants that can be volatized at temperatures below 650 °C (e.g. mineral oil, mono-aromates, PAHs, PCBs, cyanides, chlorinated solvents and TBT).

The most promising thermal processes utilize rotary kiln technology, which operates at temperatures of over 2,000 °C. The contaminants present in the material that enters the drum are volatilized at the working temperature and transferred into the gas phase. The processed and cleaned material is leaving the installation through a discharge system with water nozzles to cool it down and preventing dust formation. The gases leaving the rotary drum are treated by a system that includes a multicyclone separator, an oxidizer, an air-to air cooling chamber, baghouse and optional an acid gas scrubber (Bortone and Palumbo, 2007).

In general pretreatment is necessary. Dewatering methods should be used to reduce the moisture content of sediment before it enters a rotary kiln. Some thermal processes may require the separation of the sand fraction and washing with fresh water of sediments to remove salinity (US ACE, 2014).

2.5. Vitrification

Vitrification is an emerging technology that uses electricity to heat and destroy organic compounds and immobilize inorganic contaminants on sediments.

A typical unit consists of a reaction chamber divided into two sections: the upper section, from which it is introduces the feed material, containing gases and pyrolysis products, while the lower section contains a two-layer molten zone for the metal and siliceous components of the sediments.

Materials are vitrified by high electrical currents. Electrodes are inserts into sediments and a large current is applied, resulting in rapid heating of the solids and melted of the siliceous components. The product is a solid, glass-like material that is very resistant to leaching. Temperatures of about 1600 °C are typically achieved (US ACE, 2014).

In general, the technology requires extensive pre-processing of the dredged material. Salts must be removed from the sediment before of the vitrification, since some of them are volatilized during the melting process and provide problems corrosion of the system. Furthermore, the pretreatment process includes dewatering of the sediment to remove water prior to injection into the melting chamber, avoiding increasing costs with electrical power.

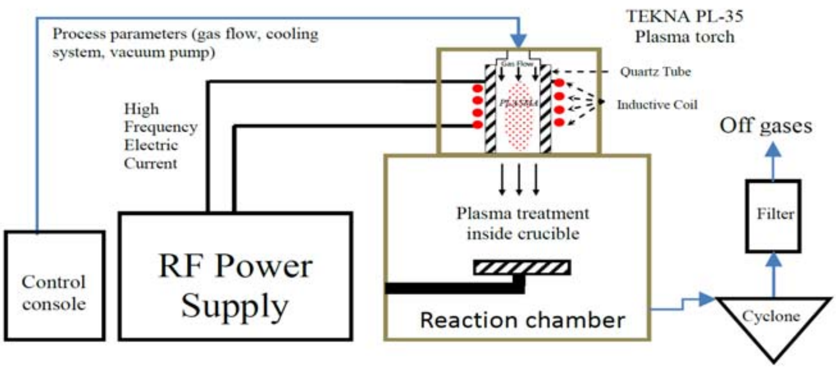


Fig. 3. Schematic illustration of the inductive plasma reactor system (Colombo et al., 2012)

3. Results and discussion

This paragraph provides information regarding potential alternatives of management of CMS using decontamination technologies. Relevant case studies, result from a literatures survey about the treatment technologies currently adopted for the sustainable reuse, recycling and recovery of CMS, are reported in Table 1.

The obtained results showed how, biological treatments have been used only in treating of marine sediments contaminated by organic compounds. For all biological treatments, there is as a rule of thumb: the greater the molecular weight (and the more rings with a PAH), the slower the degradation rate. Therefore, increases the time to complete remediation. For intensive land-farming is necessary a period of at least 1-2 years. Instead, passive landfarming may last several decades. As for other biological treatments, removal efficiency is dependent upon many factors, the most important is inorganic contaminant concentration.

Landfarming is considered as a promising technology (Harmsen et al., 2007) to remediate PAHs and mineral oil contaminated sediments, because provides an excellent opportunity to produce non-fossil fuel. Trees Fast-growing (willow and poplar) can be grown and harvested with regular cycle to produce usable wood biomass for a range of applications. This practice is called Short Rotation Coppice (SRC) and maximizes the social, economic and environmental benefits of reuse and remediation of sediments (Paulson et al., 2003). Several full-scale operations have been utilized.

Table 1. Summary of relevant case studies. Re-use is the use of material in same form (though cleaning or separation for re-use is acceptable); Recycling is the use of material in a different form after processing. Can be beneficial use, but materials changed (bricks, aggregate, etc.); Recovery of energy, biomass or other materials

Treatment	*Target contaminants*	*Scale*	*Processing*	*Product*	*References*
Solidification/Stabilization	Metals	Laboratory	Recycling	Cemented mortars	Couvidat et al., 2016
Solidification/Stabilization	Metals	Laboratory	Recycling	Fill material and blocks	Wang et al., 2015
Solidification/Stabilization	Metals	Laboratory	Recycling	Fill materia	Tang et al., 2015
Thermal pretreatment and S/S	Metals	Laboratory	Recycling	Fill material and bearing	Wang et al., 2015
Solidification/Stabilization	Metals	Laboratory and pilot	Recycling	Granular material	Achour et al., 2014
Sediment Washing + Vitrification	Hydrocarbons	Pilot	Recovery	Fill material and silicon	Magagnini et al. 2013
Phytotreatment processes	Hydrocarbons	Pilot	Recycling	Recycled land	Masciandaro et al., 2012
S/S + Thermal Desorption	Hydrocarbons	Laboratory	Recycling	Fine aggregates	Careghini et al., 2010
S/S + Thermal Desorption	Mercury and hydrocarbons	Laboratory	Recycling	Fine aggregates	Bonomo et al., 2009
Vitrification	Metals and hydrocarbons	Laboratory	Recovery	Silicon	Colombo et al., 2009
Landfarming	PAHs and mineral oil	Pilot	Recovery	Biomass for bioenergy	Harmsen et al., 2007
Stabilisation + Thermal Desorption	Metals	Laboratory	Recycling	Cement-based materials	Agostini et al., 2007
Stabilisation + Thermal Desorption	Metals	Laboratory	Recycling	Clay bricks	Zoubeir et al., 2007
Sediment washing	PAHs	Pilot	Re-use	Manufactured soil	Stern et al., 2007
Composting	PAHs and PCBs	Pilot	Re-use	Recycled land	US ACE, 2014
Composting	PAHs and PCBs	Pilot	Re-use	Recycled land	US ACE, 2014
Solidification/Stabilization	Metal	Pilot	Recycling	Fine aggregates	Wiley et al., 2002

The most studies shown in Table 1 are referred to S/S treatments. The fact that S/S has been so extensively utilized for management of CMS is testament to the effectiveness of the technology in terms of structural properties of dredged material and environmental compatibly. Use of S/S treatments to beneficial reuse of CMS appears to be well demonstrated for civil engineering applications. However, performance of this treatment is difficult to predict because of the complex interactions between organic contaminants and binding agents. For this reason, often S/S treatments are associated with thermal treatments.

The obtained results show as sediment washing is a physical/chemical treatment that can be applied to organic and inorganic contaminants. Contaminant reduction varies and depends on the sediment matrix, total organic carbon content, contaminant concentrations, contaminant type(s) and extent of treatment. Removal rates are typically in the 60 to 80% range for fine materials, with higher rates achievable on coarser grain fractions.

With this treatment is possible the separation of fine fraction (the most contaminated) to produce sandy material sufficiently decontaminated for beneficial reuse.

The public supports sediment washing due to low environmental impact, low emissions, beneficial products, and competitive costs at a commercial-scale level. Thermal technologies have been shown to be effective in destruction of organic contaminants and immobilization of metals. The most promising thermal treatments use technologies of the rotary kiln and the plasma torch. Though thermal technologies have proved highly successful from a strictly decontamination standpoint, from a logistical standpoint, kilns are expensive and difficult to realized (due to air pollution concerns) and prone to breakdowns that reduce reliability.

Thermal desorption plants are in operation at full scale in several countries (e.g. Belgium, The Netherlands, Germany). Proponents of rotary kiln technology suggest that costs for sediment treatment might be reduced by adding electronic waste, waste solvents/ oils, or tires to the input stream. Small-scale tests indicated that adding other waste streams does not negatively impact the quality of the final product. Unfortunately, adding other waste streams does reduce the processing rate of dredged material. Compensating for this requires additional kilns, which would further increase capital costs. Another way to lower processing costs without sacrificing processing capacity for dredged material might be to scavenge waste heat from the process and use it to generate electricity (cogeneration).

Plasma technology offers a great number of unique advantages to decontaminate marine sediments and to obtain a final recyclable product: (i) the high temperatures ($T \geq 104$ K) help to decompose all organic contaminants presented, encapsulating heavy metals and mineral phases into a stable glassy matrix, (ii) the use of electric energy to generate the plasma discharge allows to decreasing the gas flow produced, facilitating control and treatment of emission and giving the possibility of generating saleable products (Colombo et al., 2009)

4. Conclusion

Among the various technologies stated in this review, S/S remains the most widely used technology for beneficial reuse of CMS. However, this technique has certain drawbacks such as its limited application for organic contaminants.

Although thermal treatments are superior from decontamination standpoint, these techniques are not preferred. Compared with other conventional treatments, there is basic apprehension about construction of thermal plants. Equipment for pollution controls must be state-of-the-art to insure regulatory compliance and instill public acceptance/confidence.

Future research is needed to study the combined effect of two or more technologies or to develop an efficient innovative technology that is eco-friendly, affordable, scalable and easily available.

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