

Introduction to the treatment of polluted sediments

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Abstract

This paper overviews the large variety of remediation options for polluted sediments that are currently available. These include physico-chemical and biological techniques that can be applied in situ or ex site.

1. Introduction

Polluted sediments are worldwide a serious environmental risk for human beings and ecosystems. Large volumes of polluted sediments have to be dredged for nautical or environmental reasons. When contaminated sediments are dredged major costs are involved for proper disposal or remediation of the sediment material. Due to these high costs the management of polluted sediments is a serious issue. Currently the management approach for these polluted sediments is strongly based on a risk based assessment strategy (Brils & Deckere 2003). Besides the reduction in environmental risks the various remediation options can offer to abate the problems caused by these sediments also their sustainability, technical and economical feasibility and social acceptability, have to be assessed. In this respect also the potential beneficial use of the sediment have to be taken into account. In this risk based assessment strategy remediation technologies and technological remediation approaches play a key role.

The aim of this paper is to give a very brief overview and short description of available remediation technologies and remediation options. Besides a brief assessment will be given regarding development stage, experience with practical application and indication of costs. Finally a

short discussion will be given of expected future developments regarding sediment remediation.

2. Characterisation of polluted sediments and sediment sites

Important pollutants in sediments are polyaromatic hydrocarbons, mineral oil compounds, compounds of the heavy metals cadmium, copper, chromium, lead, mercury, nickel, zinc, arsenic, tributyl tin compounds, PCB's, organochlorine pesticides, organophosphorous pesticides, dioxins, and endocrine disruptors (Apitz et al. 1999). The majority of these pollutants have a low solubility in water. It can be expected that soluble pollutants have already dissolved in the water phase.

In general heavy metal compounds such as copper, mercury and nickel are primarily bound to the organic matter of the sediment or present as sulphide precipitate. Lead is primarily bound to iron and manganese hydroxides. Organic pollutants are in general bound to organic substances in the sediment. However, a strong deviation of this general picture is also possible depending on the source of the pollution. Pollutants can also be present in the sediment as particles consisting of pure pollutant. Especially in case of a remediation approach focused on a complete or partial clean up of the sediment the

physical state of the various pollutants is a crucial factor.

Compared with polluted soil, which very often contains only one type or a few types of pollutants, polluted sediments very often comprise a mixture of strongly different pollutants such as organic pollutants and a variety of heavy metal compounds. A characteristic aspect of sediment is also the large fraction of water in the sediment. Due to the presence of this water phase remediation costs can be substantially.

For the development of an optimal remediation strategy it is also very important to have a granular size characterisation of the sediment particles. Not only the size distribution of these particles is in that respect relevant but also the composition of the various granular fractions in terms of concentration of pollutants in these fractions and the organic matter content of these fractions.

Because current remediation strategies for polluted sediments are primarily based on risk assessment a very important characteristic factor of pollutants in sediment is the bioavailability of these pollutants. Unfortunately currently there is no uniform and consistent definition and interpretation of the concept of bioavailability. This also holds for the measurement of the bioavailability. In literature a variety of definitions of bioavailability is found as well as underlying factors, which determine the bioavailability. The focus of bioavailability therefore varies strongly. This focus can be related to:

- The toxic effect of pollutants or intermediates originating from the degradation of pollutants on humans and organisms belonging to the sediment (or soil) ecosystem,
- The biodegradation rate of pollutants by micro-organisms or the conversion or uptake of pollutants by plants,
- The release of pollutants from the sediment (or soil) matrix and in case of soil the transport by ground water flow in the polluted site itself but also to the non-polluted surroundings, including the groundwater. In this definition the bioavailability is not directly associated with toxic effects on sediment (or soil) biota or on biodegradation but is only a measure for the dispersion potential of pollutants.

For several reasons these definitions hamper practical application. The factor time is not clearly specified, while it is known that the bioavailability can change strongly with time due to

changes in the interaction between sediment (or soil) matrix and pollutants and/or sediment (or soil) structure and composition. Using three types of (bio)available fractions can partly solve the aspect of the factor time: the actually available fraction, the potentially available fraction, and the non-available fraction. The potentially available fraction includes the actually available fraction but it cannot exceed the total concentration as measured analytically. Chemical or microbiological degradation of a pollutant, especially of chlorinated organic pollutants, may result in intermediates that are even more toxic or more mobile than the original pollutants. In such a case decrease of the bioavailable fraction will lead to increase of toxic effects. The toxic effect of pollutants on sediment organisms varies strongly with the type of organisms.

Real measuring of bioavailability in an accurate and consistent way is much time consuming and therefore in general, expensive. This limits its application on a practical scale. This is the reason that a considerable amount of research is focused on the development of simple, less time-consuming physical/chemical laboratory methods for the assessment of bioavailability. The basic idea behind these physical/chemical methods is that with these methods the easily available pollutants, corresponding with the bioavailable pollutants, can be removed from the sediment (or soil) matrix, while the non-easily available pollutants, corresponding with the non-bioavailable pollutants, are not removed. Of course, also here the intensity of the physical-chemical treatment process, and the duration of the process, influences to a certain extent the fraction of the pollutants removed.

Finally a proper and optimal remediation strategy for contaminated sediments requires also factors dealing with the amount of sediments, characterisation of the polluted site, and characterisation of potential disposal sites for the sediments.

3. Remediation options

A large variety of remediation options for polluted sediments is available (Rulkens et al. 1998; Fletcher & Burt 1999; Tuchman et al. 1999; Schumacher et al. 1999; Thomas & Gidarakos 2001; Anonymous 2002; Saporano et al. 2003). Table 1 gives a summary of these options

Table 1. Brief overview and assessment of the remediation options

Option	Development stage	Practical experience	Cost indication
<i>Thermal treatment</i>			
Incineration	+	+	-
Thermal desorption	+	0	0
Vitrification/thermal immobilisation	0	0	-
Wet oxidation	0	0	-
Supercritical oxidation	-	-	-
<i>Physical/chemical (ex-situ) treatment</i>			
Stabilisation and solidification	+	0	0
Wet classification	+	+	0
Solvent extraction	+	0	0
Chemical extraction/chemical leaching	0	0	0
Electrokinetic remediation	+	+	0
<i>Biological (ex-situ) treatment</i>			
Bioleaching	0	0	0
Aerobic composting	+	0	+
Bioslurry systems	+	-	-
Anaerobic treatment	0	-	?
Landfarming	+	+	+
Phytoremediation	0	0	?
<i>In situ treatment</i>			
Aerobic treatment	-	-	?
Anaerobic treatment	-	-	?
Addition of stabilisation agents	-	-	?
Use of aquatic plants	-	-	?
Natural attenuation	-	-	+
<i>Disposal</i>			
Confined fully submerged aquatic disposal	+	+	+
Confined partially submerged aquatic disposal	+	+	+
Controlled landfill	+	+	+

Development stage: - (initial stage), 0 (further development necessary), + (in practical stage).

Practical experience: - (no), 0 (limited), + (substantial).

Cost indication: - (high), 0 (moderate), + (low), ? (no indication possible).

together with a brief evaluation of these options. In general these options can be subdivided in options dealing with the removal of pollutants by thermal treatment, the physical removal of pollutants from the sediment, the chemical or (micro) biological destruction of sediment pollutants, the disposal of sediments in a landfill or in confined or non-confined disposal facilities. Also natural attenuation is possible. Another subdivision regarding the remediation of sediments that can be made is treatment or disposal of dredged sediment (*ex-situ* treatment) or treatment of the sediment without dredging (*in-situ* treatment). In the following the options mentioned in Table 1 are briefly described.

3.1. Thermal treatment

3.1.1. Incineration

In this process the sediment is fed to an incinerator where the organic pollutants are completely destroyed by an oxidation process at high temperatures. A separate dewatering step is necessary as pre-treatment step to reduce energy costs of the incineration process. The incineration process is especially suitable to treat sediments with large amounts of mineral oil (because then the energy requirement is relatively low) or sediments contaminated with toxic organic compounds such as polyaromatic hydrocarbons, PCB's, pesticides, dioxins or chlorinated hydrocarbons.

If dealing with chlorinated toxic organics an after burner operating at special conditions is a requirement to prevent emissions of toxics to the atmosphere. Heavy metals are in general immobilised in the sediment matrix. Exceptions are mercury containing compounds. Oxo-anions forming elements like As, Mo and V will often form easily leachability compounds.

3.1.2. *Thermal desorption*

In this process the (partially dewatered) sediment is heated in a closed reactor system to such a temperature that organic pollutants will vaporise from the sediment together with water. The vaporised pollutants can be removed and concentrated from the gas phase by absorption or condensation. In this process the pollutants are not destroyed but are concentrated in a small volume. This residue can be treated subsequently. The process is suitable to remove pollutants that have a relatively high volatility and stability at elevated temperature such as low molecular hydrocarbons and low molecular poly aromatic hydrocarbons. In case of unstable volatile pollutants such as chlorinated hydrocarbons special equipment and conditions can be necessary to prevent formation of dioxins and furans. The required temperature for thermal desorption depends on the type of pollutants to be removed and varies between 100 and 500°C.

3.1.3. *Vitrification/thermal immobilisation*

Vitrification or thermal immobilisation is a thermal process in which the sediment is heated to such a temperature that the inorganic compounds in the sediment will melt and the organic pollutants are completely destroyed. Complete destruction of the volatilised organic compounds and its destruction products often requires an after-burning process operating at appropriate conditions. Inorganic pollutants such as heavy metals are immobilised completely in the slag that is obtained after cooling of the melt. Depending on the process conditions and type of sediment the slag can be suitable as building material (Ulbricht 2001). By adding appropriate additives and applying appropriate process conditions a slag can be obtained with pozzolanic material, possessing cementations properties (Gardner et al. 2001).

3.1.4. *Wet oxidation*

Wet oxidation is an oxidation process that occurs in the water phase at high temperatures and high pressures but below the supercritical temperature and pressure of water. It is suitable to oxidise pollutants such as petroleum hydrocarbons, phenolic compounds, and polyaromatic hydrocarbons. The degree of oxidation strongly depends on the type of pollutants and on the applied process conditions such as temperature and residence time of the contaminated sediment in the reactor.

3.1.5. *Supercritical oxidation*

Supercritical oxidation is a special modification of the wet oxidation process. Temperature and pressure are above the critical point of water. At supercritical conditions the water phase has special properties such as a substantially higher solubility of oxygen and many toxic organic compounds in the water phase and a much higher oxidation rate and oxidation efficiency of contaminants. Also very persistent contaminants such as PCB's can be oxidised completely. The extreme conditions of this process require special equipment.

3.2. *Physical–chemical (ex-situ) treatment*

3.2.1. *Stabilisation and solidification*

Stabilisation and solidification processes are aimed to immobilise the pollutants in the sediment at ambient temperature by mixing the partially dewatered sediment with specific bulk materials such as cement, fly ashes, thermo plastics, etc (Mensingher et al. 2001). Often also chemical reagents are added to this mixture. The immobilisation process occurs by chemical reaction or by entrapment or by both. The result is a product with a strongly reduced leachability of the pollutants. Dependent on its properties the obtained product has to be disposed in a landfill or can be used as a construction material.

3.2.2. *Wet classification*

Wet classification is a treatment process for contaminated sediments focused on a separation of polluted sediment particles from non-polluted sediment particles. The process consists of two main steps: the first step is an intensive mixing of sediment aimed to disintegrate agglomerates of

sediment particles. This pre-treatment step is followed by a mechanical separation step by means of a hydrocyclone, centrifuge, flotation process, or up flow column. In this step polluted particles are separated from the non-polluted particles and concentrated. Often the obtained water phase requires an appropriate treatment process. Wet classification is applicable to sediments in which pollutants are specifically bound to a certain sediment fraction that easily can be separated from the other fractions.

3.2.3. *Solvent extraction*

Solvent extraction is a physical process in which the pollutants are removed from the sediment by extraction with an organic solvent that is intensively mixed with the sediment and that has a high solubility for the pollutants (Bruning & Rulkens 2001). Two types of solvents can be used: solvents which can completely dissolve in water and solvents that are not or only very slightly soluble in a water phase and in which water is also not or only very slightly soluble. The mixing step of solvent and sediment is followed by a separation step of the sediment particles. The solvent (extracting agent) is treated in order to remove and concentrate the pollutants. The treated solvent can then be reused again in the extraction step. The process can be applied to remove toxic organic pollutants from sediment.

3.2.4. *Chemical extraction/chemical leaching*

Chemical extraction, also called chemical leaching, is a clean-up process aimed at the transfer of heavy metals from the polluted sediment into solution in a liquid phase, which has been brought into contact with the polluted sediment (Rulkens et al. 1995; Massecheleyn et al. 1996; Kiefer & Höll 1998; Thöming et al. 1998). Several types of chemicals can be used in chemical extraction or chemical leaching. One type are the inorganic acids such as hydrochloric acid, sulphuric acid and nitric acid. The main purpose of these acids is to lower the pH in order to improve the release of heavy metals ions from the sediments. Also organic acids such as acetic acid, tartaric acids, and citric acid can be used. These acids are also intended to decrease the pH to release the heavy metals from the sediment structure but they also can form water-soluble metal-ion complexes. Chelating agents such as

ethylenediaminetetraacetic acid (EDTA), diethylenetriaminepentaacetic acid (DTPA) or nitrilotriacetic acid (NTA) can also be applied. These chemicals are primarily focused on the formation of strong metal-ion complexes.

3.2.5. *Electrokinetic remediation*

Electrokinetic remediation is a process in which an electrical field is applied within a sediment layer by means of electrodes placed in the sediment layer (Doering et al. 2001; Granade & Gent 2001). The electrodes are connected to a low voltage electrical power generator. The electrical current causes mobilisation and electromigration of heavy metals to the electrodes. At these electrodes the heavy metals are concentrated and removed. Besides electromigration also electroosmosis and electrophoresis take place, causing transport of water and also of mobile organic pollutants. However, the electrokinetic remediation process is primarily focused on the removal of heavy metals during the electrokinetic process. The temperature of the sediment layer increases slowly during the electrokinetic remediation. This can be beneficial in case that also organic biodegradable pollutants have to be removed from the sediment.

3.3. *Biological (ex-situ) treatment*

3.3.1. *Bioleaching*

Heavy metals and heavy metal compounds in sediment can be mobilised by several types of acidophylic bacteria that oxidise reduced sulphur compounds to sulphuric acid. This offers the possibility to clean up heavy metal containing sediments by bioleaching (Rulkens et al. 1998; Tichy et al. 1998). The most important bacteria that oxidise reduced sulphur compounds include the genus *Thiobacillus*. These bacteria live under aerobic conditions, are autotrophic, and show optimal growth at pH 1–4. The performance of the bioleaching process depends on the speciation of the heavy metals. If the heavy metals are present as a sulphide (e.g. in anoxic sediment), then *Thiobacilli* will oxidise the metal sulphide precipitate directly. If a low pH is reached, then the metals will become mobile and can be extracted from the contaminated sediment. If however no sulphides are present in the sediment, an external reduced sulphur source has to be added.

Bioleaching can be performed either in sediment-slurry systems or by heap leaching.

3.3.2. *Aerobic composting*

Composting is a microbiological treatment system in which partially dewatered sediment, polluted with organic contaminants, is intensively mixed with bulky natural organic materials and then placed in an aerated stockpile. The natural organic material improves the permeability of the pile and supplies nutrients necessary for the microbiological aerobic biodegradation or transformation of organic pollutants in the sediment. Aeration of the pile can occur by mechanical means or by natural ventilation. The process can in general handle sediments polluted with relatively easily biodegradable contaminants but also petroleum hydrocarbons and low molecular polyaromatic hydrocarbons can be biodegraded.

3.3.3. *Bioslurry systems*

A bioslurry system consists of a reactor in which the sediment slurry is intensively mixed and aerated in order to stimulate the growth of microorganisms that can biodegrade the contaminants present in the sediment. These microorganisms are already present in the sediment or can be added during the biodegradation processes. Also part of the natural organic substances in the sediment can be converted releasing pollutants that are strongly bound to the organic matrix of the sediment. The process is more or less comparable with an activated sludge system for treating municipal wastewater. Bioslurry reactor systems are suitable for treating sediments consisting of fine particles and contaminated with organic pollutants sensitive to aerobic biodegradation (Fletcher & Burt 1999).

3.3.4. *Anaerobic treatment*

Polychlorinated organic pollutants in sediments such as trichloro-ethylene tetrachloro-ethylene polychlorinated biphenyls (PCB's) and dioxins are in general not biodegradable at aerobic conditions. However at anaerobic conditions many of these pollutants are biodegradable to compounds with a lower number of chlorine atoms (Gruden et al. 2001). The anaerobic microbiological biodegradation process has to be followed by an aerobic microbiological biodegradation process to get a further dechlorination of the pollutants.

3.3.5. *Landfarming*

Landfarming is a treatment method in which partially dewatered sediment is spread out in a relatively thin layer on a specially constructed site. The microbiological activity in this layer, responsible for aerobic biodegradation of the pollutants, is stimulated by regular ploughing of the layer, in order to maintain an adequate aeration, and by the addition of nutrients (Vermeulen et al. 2003a). Further improvement can be obtained by covering the site to prevent the site from getting too wet and to elevate the temperature of the site in order to stimulate the microbiological degradation process (Harmsen & Bouwman 2001). Also mechanical aeration or artificial heating of the sediment layer can enhance the biodegradation process. Very often landfarming is applied to sediment that has previously been dewatered to a certain extent by a simple natural dewatering process. Landfarming of this type of sediment is aimed at both dewatering and ripening of the sediment and aerobic biodegradation of the organic pollutants. Ripening is a natural process in which the sediment is slowly physically/chemically transformed in soil (Vermeulen et al. 2003b).

3.3.6. *Phytoremediation*

Phytoremediation is a bioremediation process focused on the application of special plants to remove or destroy pollutants in sediments (Lee et al. 2001; Hueseman et al. 2003). There are several modifications of this process. Phytodegradation of organic pollutants occurs by stimulating the biodegradation of pollutants such as polyaromatic hydrocarbons and PCB's by root associated microorganisms. Another modification of phytoremediation is the extraction of heavy metals from the sediment and the accumulation of these heavy metals in the plant. A third modification of the process is phytostabilisation of pollutants by plants. In this process plants stabilise the sediment resulting in an immobilisation of pollutants in the sediment. Phytoremediation can also stimulate the dewatering of sediment by extraction and evaporation of water, resulting in a faster ripening process.

3.4. *In-situ treatment*

Several options are under development for remediation *in-situ* (Suman Raj et al. 2003). These

options are in general focused on an anaerobic or aerobic biodegradation of the pollutants. Addition of electron acceptors or electron donors can be helpful. Another process is the use of aquatic plants to oxygenate the sediment. It is also possible to reduce the mobility of the pollutants in the sediment by the addition of proper chemicals or binding materials.

3.5. Natural attenuation

Natural attenuation involves the natural degradation or immobilisation of pollutants in the sediment without application of an external intervention. Degradation (or immobilisation) occurs by biological, physical or chemical transformation (Ickes et al. 2003). In general natural attenuation is a very slow process and to follow this process an intensive and long lasting monitoring process is necessary. It is of course possible to stimulate natural attenuation by external intervention.

3.6. Disposal

Several options for the disposal of dredged polluted sediments exist. Most important options are confined fully submerged aquatic disposal, near shore partially submerged disposal or disposal of (dewatered) sediment in a controlled landfill.

4. Discussion and conclusions

In the previous paragraphs a brief overview is given of possible remediation options for polluted sediments. These remediation options vary from individual technologies focused on clean-up of polluted sediments to more integrated remediation approaches on management level. It is evident that these technologies not only strongly vary with respect to the type of technology but also with respect to its development stage, field of application, practical experience and costs. Among others the field of application strongly depends on the type and physical state of the pollutants in the sediment, the granular composition of the sediment and the physical/chemical composition of the sediment. It has also to be noted that the application of these methods is not always allowed.

As already mentioned the management approach for polluted sediment is currently strongly based on a risk reduction approach. In this approach the various relevant management options for sediment are assessed and compared in order to underpin an optimal decision regarding the selection of the option that will be applied. In this strategy clean-up technologies and other remediation technologies are key factors not only because the chosen option has to be technologically feasible but also because this option has to be economically feasible, environmentally sustainable, and acceptable. In this context it will be evident that there is an ongoing challenge to develop remediation technologies that are more cost effective and more sustainable. The latter means that the focus on remediation has also to be on the reduction of energy in the remediation process and the beneficial use of the sediment matrix. Regarding the development of clean-up technology it has to be noted that research in this area not only results in an increased insight in the way to mobilise or destruct pollutants from sediments in order to remove these pollutants from the sediment but increases also the insight in factors that are responsible for the bioavailability of pollutants. The same also holds for the spin off that can be obtained in the research into immobilisation technologies for pollutants in sediments. This increased insight in bioavailability factors is therefore in a direct way very helpful in the risk based management strategy of polluted sediments.

Overlooking the entire problem of polluted sediment management some major research challenges can be indicated:

- Further development of clean-up technologies that can deal with organic pollutants, inorganic pollutants or combinations of these pollutants. Also the improvement of the theoretical insight is an issue of high importance.
- Development of *in-situ* treatment technologies focused on *in-situ* aerobic or anaerobic biodegradation, chemical oxidation or chemical reduction. Addition of appropriate chemicals, bioaugmentation, biostimulation or use of specific water plants are options in that respect.
- Treatment of sediments confined in sub water disposal sites. Options may be oxygenation and stimulating aerobic biodegradation, anaerobic biodegradation and use of specific

adsorbents for the pollutants, which after a set adsorption period can selectively be removed from the sediment.

- Acceleration of the ripening and dewatering process of contaminated sediments that are spread out in a thin layer over a prepared soil area. A better theoretical insight is necessary for understanding the ripening and dewatering processes of sediment.
- Improvement of the theoretical basis of mobilisation processes of pollutants from sediment particles, release of mobilised pollutants from these particles, and processes dealing with immobilisation.
- Broadening the theoretical basis for decision tools, such as measuring and interpretation of the bioavailability, is very relevant.
- Because the current management strategy for polluted sediments often results in a remediation approach that is highly space consuming, and because this space is not always available, links have to be made with other social and industrial activities that require large space area such as road construction, development of industrial area, etc.
- More focus on the beneficial use of the sediment material is required. It has to be noted that not all sediments are real waste in the end.

It is evident that it is impossible in this brief introduction to highlight and evaluate the entire pallet of sediment remediation processes. This overview is aimed as a tool that may be helpful in the further orientation in this issue.

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