

SEDIMENT TREATMENT: A DECADE OF PRACTICAL EXPERIENCE LE TRAITEMENT DES SÉDIMENTS: UNE DÉCENNIE D'EXPÉRIENCE EN PRATIQUE.

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ABSTRACT

The treatment of contaminated sediments plays a key-role in re-establishing the complete water-ecosystem. Many subaqueous "black points" are threatening our fresh water reservoirs. Nevertheless the treatment of contaminated dredged material has made significant progress during the last years. While several techniques have been developed in the past decade, practical solutions for large volumes of CDM are only available since recent times. It is the concern of many authorities involved in the sediment management sector to minimize volumes to be disposed off, while maximizing the beneficial usages. Several options are available for material that is properly conditioned: Dike and shore protection, maintenance of riversides to prevent flooding, wetland restoration, industrial site reclamation, and even horticultural usage. Sediment however must be treated and conditioned prior to this reuse. While techniques have become available two important factors have inhibited the implementation until now: budget constraints and legislation. Extensive scientific and practical research has been carried out to up-scale lab and pilot proven technology for cleansing, remediating and conditioning techniques. In addition several large-scale treatment centres have been constructed, where sediments can be treated under the environmentally most up to date circumstances.

RÉSUMÉ

Le traitement des sédiments contaminés joue un rôle clé dans le redressement de l'écosystème fragile, qu'est l'eau. De plus en plus l'approvisionnement en eau devient menacé par de nombreux sites qui sont en même temps contaminés et immergés. Néanmoins les moyens pour traiter les sédiments contaminés ont amélioré de façon significative dans les dernières années. De différentes techniques ont été développées dans les 10 années passées. Cependant il est clair que seulement depuis récemment, des solutions pratiques ne se sont présentées. C'est la tâche de tous les responsables de gérer les sédiments d'une telle manière à ce qu'un minimum ne soit mis en décharge tout en maximisant les usages bénéficiaires. Il est aussi vrai que malgré ces moyens de restauration existent aujourd'hui il faut tenir compte d'autres obstacles, tels que le cadre législatif, financière et socio-économique.

Différentes possibilités se présentent pour les boues, qui ont été proprement conditionnées: construction des digues, protection côtière, maintenance des rivières, afin d'éviter des inondations, utilisations en agriculture, voire jardinage. Il est clair que les matériaux demandent un traitement et conditionnement avant tout usage.

Des études scientifiques et pratiques ont aidé à exécuter à grande échelle les techniques labo et pilotes approuvés pour le nettoyage, la remédiation et le conditionnement. En plus des investissements importants ont été faites dans des centres de recyclage.

1. INTRODUCTION

In the course of river maintenance to prevent silt built-up the question about sediment quality, treatment and safe management is raised. In the early days, sediments were stored on land or used as soil for farming. Nowadays most sediments may no longer be regarded as clean.

The rivers and streams contain increased levels of heavy metals, PAH's, PCB's, oil and fuel and other organic and inorganic residues. Both industry and agriculture may be held responsible for the input of these pollutants. Organic and inorganic pollutants generally have accumulated in the sediment over the years. In some cases this concerns very specific compounds like Tributyl-Tin, which not only has shown disastrous effects on the aquatic biofauna. It also necessitates sophisticated and specifically monitored treatment.

Since European and national laws have become more stringent, sediments have to be treated as waste.

Therefore they may no longer be distributed freely on land or used in agriculture.

Two options are left open to deal with this waste stream: landfilling or treatment in order to recycle as much as possible.

Landfilling is the easiest solution but offers little perspective in view of the environment. Also, environmental taxes on the dumping of waste are increasing and proper management and controls for landfilling become more complicated. So most indicators are not in favour of landfilling as such. However, since no recycling activity is 100% proof, there will always be a need for landfilling, in order to dispose the non-treatable and residual fractions from different treatment methods. Landfilling however can also be turned into a beneficial operation if all parameters are well considered.

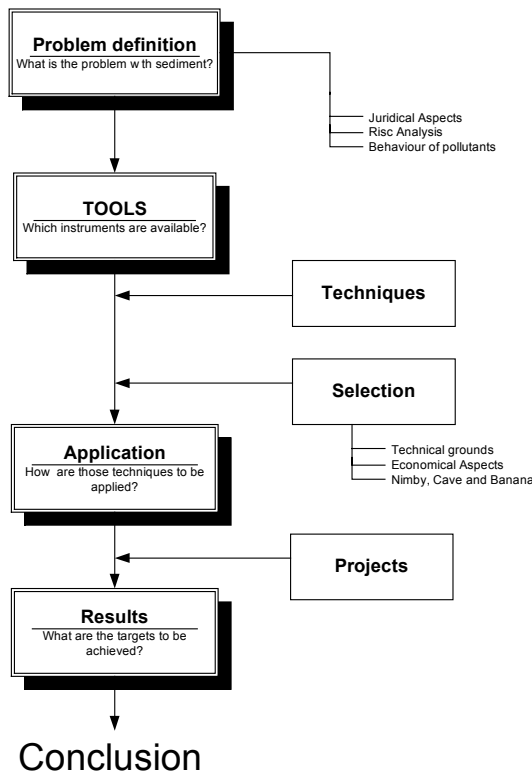


Figure 1. General Decision Framework in sediment treatment.

2. LEGISLATIVE FRAMEWORK

2.1 International legislation

At the international level the way in which dredged material is managed, is described by the OSPAR Convention (regional) and the London Convention (world). The London Convention on the prevention of marine pollution as a result of dumping waste is the equivalent at worldwide level of the OSPAR Convention. It was signed in 1972 and there are currently 78 member states. A review of the Convention began in 1993 and this review was completed in 1996 with the acceptance of the 1996 Protocol to the London Convention.

The Convention for the Protection of the Marine Environment of the North-East Atlantic (OSPAR Convention) was opened for signature at the Ministerial Meeting of the Oslo and Paris Commissions in Paris on 22 September 1992.

The Convention has been signed and ratified by all of the Contracting Parties to the Oslo or Paris Conventions (Belgium, Denmark, the Commission of the European Communities, Finland, France, Germany, Iceland, Ireland, the Netherlands, Norway, Portugal, Spain, Sweden and the

United Kingdom of Great Britain and Northern Ireland) and by Luxembourg and Switzerland.

The OSPAR Convention entered into force on 25 March 1998. It replaces the Oslo and Paris Conventions, but Decisions, Recommendations and all other agreements adopted under those Conventions will continue to be applicable, unaltered in their legal nature, unless they are terminated by new measures adopted under the 1992 OSPAR Convention.

Important in the OSPAR Convention is the general prohibition to dump waste at sea, unless for these wastes or substances that are explicitly mentioned in the Convention. Dredged material is such a waste, thus dumping at sea is not forbidden, but nevertheless it has to be regulated by licenses given by local authorities.

Dumping at sea is also only possible if the Sediment Quality Criteria (SQC's) as described in the OSPAR Convention are met.

Table 1 : Sediment Quality Criteria as described in the OSPAR Convention

	Target value	Limit value
Hg	0,3 ppm	1,5 ppm
Cd	2,5 ppm	7 ppm
Pb	70 ppm	350 ppm
Zn	160 ppm	500 ppm
Ni	70 ppm	280 ppm
As	20 ppm	100 ppm
Cr	60 ppm	220 ppm
Cu	20 ppm	100 ppm
TBT	3 ppb	7 ppb
Mineral oil	14 mg/g _{oc}	36 mg/g _{oc}
PAH's	70 µg/g _{oc}	180 µg/g _{oc}
PCB's	2 µg/g _{oc}	2 µg/g _{oc}

2.2 European legislation

Since 1 February 1993, the transfer of wastes within, to and from the European Community is described in Regulation (EEC) 259/93/EC of the Council. In this regulation different wastes are entered in three lists, a green list, an orange list and a red list. Depending on which list a waste product belongs to, specific rules are imposed when transferring these products.

The transfer of wastes from the green list is regulated by a simplified procedure.

For the transfer of wastes from the orange and red list an extensive control procedure is developed. This procedure consists of a system of prior notice to the proper authorities

of the different involved countries, so they are well aware of the type of wastes, the proposed treatment techniques and/or disposal methods and so they have the possibility to make objections to such a transfer. In case such a transfer is demanded with the aim of a beneficial use of the (treated) wastes, the possible objections are limited, contrary to these when a final disposal is intended.

Contaminated dredged material is not entered in any of the lists in annex of Regulation (EEC) 259/93/EC, which means that the procedure as if it was a red list waste needs to be followed!

Decision 2000/532/EC of the Council of 3 May 2000, gives a classification of all waste products and hazardous waste. In article 2 of this decision a series of properties are listed where waste is classified as hazardous if one or more of these properties are present. In the list of waste products in annex of this decision some wastes are considered as a hazardous waste. Dredging spoil (code 17 05 06) as such is not considered as a hazardous waste. Nevertheless if one or more properties as mentioned in article 2 are present, dredging spoil can also be classified as hazardous waste (code 17 05 05).

Finally Regulation 1999/31/EC of the Council of 26 April 1999 describes the disposal of wastes. This regulation gives a series of measures and conditions in relation to the installation of a disposal site. For example, the needed bottom protection is described in relation to the type of disposed wastes (dangerous or non-dangerous waste). Dredged spoil is considered as dangerous waste. This means that a double sealing is needed consisting of a geological barrier equivalent to a layer of more than 5 m with a permeability of less than 10^{-9} m/s and on top of that an artificial geo-membrane, e.g. High Density Poly Ethylene.

2.3 Practical experience

An example of a practical implementation of various legislations on soil and waste treatment is found in Flanders. Possible reuse of treated sediments is covered by Federal and Flemish regional regulations. In Flanders the final destination of a waste material has to follow a strict set of laws set out in the VLAREA (Flemish Regulation on Waste).

VLAREA consists of the regulations dealing with transport, treatment, storage and recycling of every imaginable waste product. Chain of custody as well as final responsibility for the waste are described.

For dredged material, re-use is only possible in one of the two following ways:

- A. Soil** use as top-soil or soil for agricultural land application
 - Pollution is very low
- B. Non-structural building material**
 - Use as filler or bulk material for dikes, roads, etc.
 - After treatment the final levels of pollutants are reduced but not as low as maximal permissible

concentrations in soil. Leachability however must always be lower than preset values.

Table 2 gives an overview of the values for the most common pollutants: Heavy metals, mineral oil and PAH's. Other parameters include BTEX and related chemical compounds, PCB's, chlorinated carbonhydroxides and insecticides. In order for a treated sediment to be allowed for re-usage residual concentrations may not exceed one single limit.

Table 2 : VLAREA-parameters for re-use of sediments (only the most common chemical compounds are displayed)

Compound	Usage as New Soil (ppm)	Usage as backfill material (ppm)
Heavy Metals		
As		250
Cd	1,2	10
Cr	78	1250
Cu	109	375
Hg	5,3	5
Pb	120	1250
Ni	55	250
Zn	330	1250
Mineral oil		
	300	1000
Polycyclic Aromatic Hydrocarbons (PAH)		
Naphtalene	0,005	20
Fenantrene	0,08	30
Fluoranthene	0,2	40
Benzo(a)antracene	0,06	35
Chrysene	0,15	40
Benzo(b)fluoranthene	0,2	55
Benzo(k)fluoranthene	0,2	55
Benzo(a)pyrene	0,1	8,5
Indenopyrene	0,1	35
Benzo(g,h,i,)perylene	0,1	35

Each project of dredging with sediment treatment is to be monitored separately. The applicant wishing to reuse the sediment must send a complete application file to the OVAM (Flemish Authority for Waste). Origin of the material, type of remediation, quantity recycled and quality are factors to be examined. Overall quality of the treated material is evaluated and used to issue a certificate for reusal of the material. This process of evaluation takes three months. The certificate regulates the transport and possible areas of re-use. It has to accompany the material at all times.

3. PROVEN TREATMENT TECHNIQUES

The first step of all treatment procedures for sludge and sediments is dewatering. The evacuation of excess water minimizes the volume of material to be handled. Furthermore the processing of the aqueous fraction is in most cases less complicated.

There are 2 main ways to dewater sediments: lagunation, which is an enhanced drainage and filtration technique and mechanical dewatering. Practical experience has shown that both techniques may be applied under different conditions.

The soil texture matrix as shown below may be helpful in the selection process. Each soil is composed of one of the three major fractions silt, clay and sand. Clay is defined as the soil fraction between 0-2 μ , silt as the fraction between 2-50 μ , and sand between 50-200 μ .

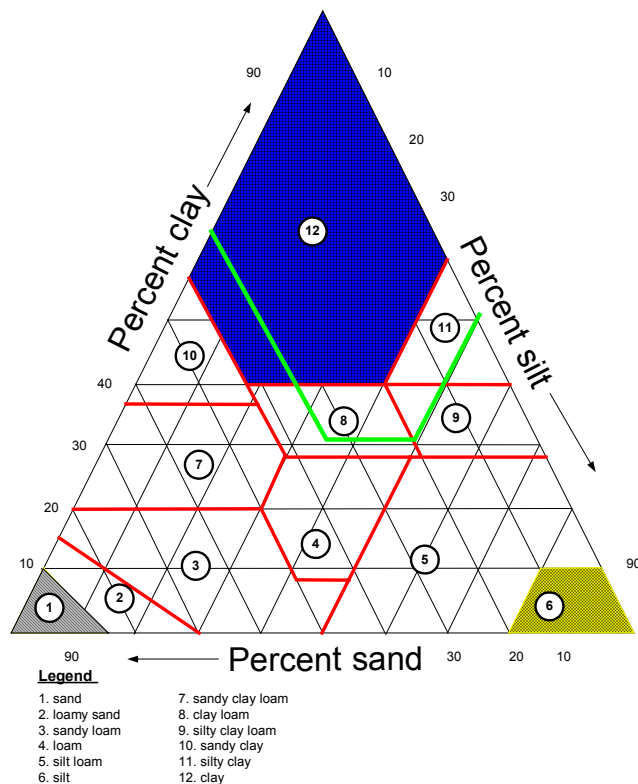


Fig 2. Soil texture matrix base for the decision of the dewatering technique.

Sand as a pure product is often dredged and reused as such. If more than 40 % sand is present in the sediments recycling techniques (hydrocycloning) are applied to recover this fraction. The remaining fraction becomes more silty or clayey.

As a general rule the more sandy or silty the material is, the more efficient the lagunation processes becomes. The pore structure enhances the removal and transport of the water in

a natural way. Sand and silty materials are more abrasive for mechanical dewatering equipment.

The more clayey the less efficient lagunation occurs. Water is physico-chemically bonded to the clay particles and interlayers. Without any action clay's will remain expanded, thus not releasing their water. Polyelectrolytes have shown to assist in the removal of this water in combination with mechanical dewatering.

Secondly the presence of pollutants must be considered. Organic compounds have more ability of biological treatment, whereas heavy metals are more likely to be treated in a chemical way, i.e. conditioning, immobilisation, extraction. Lagunation is an aërobic process which stimulates biological activity. By enhancing this process bioremediation effectively decontaminates sediments during the lagunation process.

Finally one should also consider combination of both. Mechanical dewatering followed by lagunation may also be the right choice.

3.1 Mechanical dewatering

Historically, filter presses have been showing low productions. Capacities of only up to 1 ton of dry solids an hour per cubic meter of filter press volume were achieved. In addition, the classic concept of a filter press does not allow coarse material (sand or fine gravel) to be left in the sludge, making an extensive preliminary screening and desanding necessary. Unfortunately this sand fraction would be beneficial to speed up the dewatering and increase the filter cake's geotechnical properties afterwards (Van Geldermalsen, 1995-2000).

The last years however the mechanical dewatering with filter presses has again gained interest and is further developed to be implemented on large scale (Metha-plant Hamburg, Port of Antwerp). Also mobile dewatering units, based on filter presses are recently being used in some dredging projects (Detsner, 1995).

3.2 Lagunation

In the case of large volumes of sediments to be treated it has been established that the most efficient way in terms of productivity is the lagunation. Efficiency of lagunation is governed by following (generally known) major factors:

3.2.1 Dry matter content of the initial material

At the start of the process, the drier the material is, the faster it will reach the point where it will start to behave as a soil, this is at a density level of 1,5 to 1,6.

Sediments entered at various density levels have been processed. Process-time to obtain a predefined density level was measured. Figure 3 clearly shows the increase in time needed when sediments are diluted (e.g. during dredging operations).

3.2.2 Design of the drainage system

Intensive practical studies over the last five years with increasing complexity of the build-up of the drainage layer have led to an important gain in production time. This has amongst others enabled the time interval between two successive fills in the lagunation fields to be reduced from 12 to in between 2 and 6 months. This significant improvement is caused by a major adaption of the design of the lagunation systems.

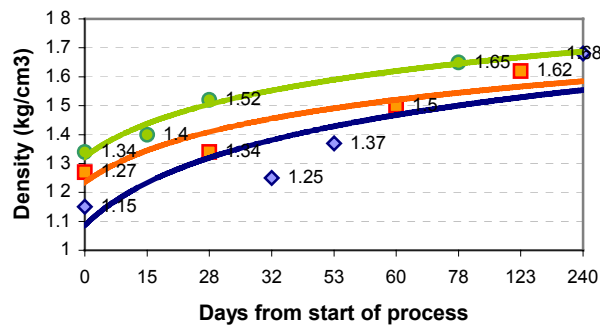


Figure 3: Evolution of the density during the lagunation process

Whereas a classical drainage system consisted of one to three layers 5 years ago, a drainage system nowadays consists of a multifiltration system of at least 10 layers.

3.2.3 Processing

The time required to obtain final soil structure is largely enhanced by the number of times the material is tilled during the process. The amount of energy used in the system will therefore account for the efficiency of the lagunation process.

3.2.4 General type of the sediment

Each type of sediment behaves differently, basically depending on the soil characteristics of the material like grain size distribution, organic matter content, presence of some chemical compounds, etc.

4. PRACTICAL EXPERIENCE

The dry matter of the sediment may vary between 20 and 35% upon arrival. The material is spread in layers up to 1 m thickness over the entire surface of the lagunation field. This maximises the surface exposed to wind and sun.

The bottom of the lagunation field is equipped with a complex drainage layer consisting of various filtering elements.

Perforated HDPE-pipes run through the drainage layer and are connected to a central water collector. This applies a

capillary suction force in the entire lagunation field, speeding up the gravitary dewatering.

The sediment is then left to dewater by evaporation and gravitary evacuation which so to say 'sucks' the water out of the pores of the material. This water may contain some of the mobilisable pollutants. It is collected in the underground reservoir from which it is pumped to a waste water treatment plant. Here the pollutants are degraded or adsorbed.

As shown in figure 3 the dewatering follows an asymptotic curve. After a primary phase during which the saturated interstitial water is evacuated the slope in dry matter curve reaches a maximum. This is due to 2 effects:

The formation of a crust on top of the material resulting from an evaporation in the top layer (sun and wind) and the consolidation of the layer located just on top of the drainage. The suction and evacuation of the water in the lower zones of the sediment creates cavities close to the filtration layer. The weight of the 1 meter of sludge compacts the layer and forms a compacted area with low water permeability.

The dewatering is again accelerated by disturbing the material, using farming equipment and excavators, in order to break the hard crust and compacted layer at the bottom.

In figure 4 the increase in dry matter content in the dredged material is shown during processing. A new increase of the dry matter content can be noticed after tilling of the material.

This process is repeated regularly until the sediment is sufficiently dry to be placed in ridges for further treatment. At this stage bioremediation starts.

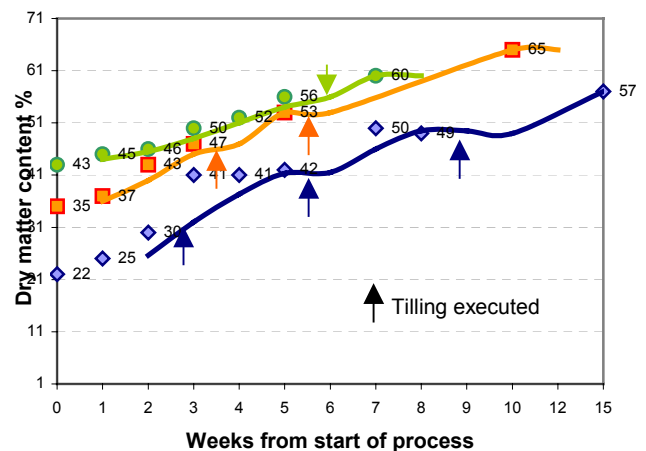


Figure 4: Evolution of the dry matter content during the lagunation process

5. BIOREMEDIATION

Bioremediation occurs at a very slow rate, since it is controlled by the level of oxygen available for the indigenous

microflora. Sediment on the bottom of rivers is in a state of near-anaerobic, which causes the black colour associated with it. This colour is the result of a chemical reaction which depletes the sediment of oxygen, e.g. sulphate to hydrogensulfide.

The anaerobic conditions in the sediment together with the small particle size (silt < 63 µm) inhibits normal indigenous microflora, therefore making natural attenuation impossible. However with the right pretreatment it is possible to render natural conditions, boosting bioremediation.

To enhance bioremediation organic fertilising materials together with soil conditioners are to be applied to the dewatered sediments.

Since the sediment was tilled during dewatering the material will start to crumble, which improves the aeration. Nutrients for the indigenous microflora are supplied to the sediment-compost. This also improves the overall grain size. Beneficial effects are further obtained by using liming techniques. Apart from the improvement of the overall grain-size distribution and formation of stable aggregates the high permeability ensures further rapid dewatering and oxydation. This is due to the fine pores created by the lime which results in high porosity even when compacted.

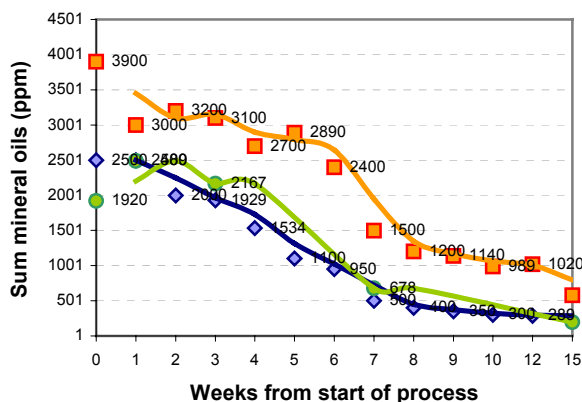


Figure 5: Evolution of mineral oil concentration during sediment remediation.

After sufficient mixing the sediment is placed in ridges for optimal bioremediation. Moisture content of the sediment is monitored to ensure proper remediation conditions. Excess rain water runs off the ridges and seeps into the drainage system. Ridges offer several advantages. They reduce the horizontal surface exposed to the rain and create a larger vertical surface for further wind evaporation, and solar contact.

Every week the ridges are turned and aerated using specially designed windrowing equipment, to supply the sediment with necessary oxygen. During this process remaining lumps of sediment/sludge are also crushed. This results in a larger surface area and hence faster bioremediation.

Figure 5 shows the effect of bioremediation on the degradation of mineral oil in sediment. Monitoring of degradation shows an increase in absolute values at the beginning. In most cases, after this shoulder a regular degradation pattern is obtained. Particular problems may arise, which delay or even stop the biodegradation process. It is then up to the operators to define what the causes of the event may be, and to take necessary measures. Sometimes, i.e. in the case when the mineral oil fraction is composed of a large amount of long-chain hydrocarbons (> C-40), bioremediation will only continue in combination with some chemical pre-treatment.

The effects of bioremediation are not limited to mineral oil. Other organic pollutants, e.g. BTEX and PAH's, are degraded due to a combined effect of stripping, desorption, oxydation, UV-light (< 400 nm) auto-oxydation and bioremediation. (De Brabandere, 1993)

For PAH's the bioremediation goes at a slower rate due to the polycyclic nature of the components. Bacteria will first degrade the easier hydrocarbons, like single chains before breaking down to the branched chains or aromatics. Extensive work on this particular area has been carried out in the past. This is described also extensively in literature.

Table 3 : Removal of TBT from sediments using an in-line oxidation process.

Experiment	TBT (µg/l)	
Start	150	300
Sample 1	16,41	30,85
2	26,1	54,61
3	19,44	37,02
4	11,54	22,61
5	12,76	26,04
Mean	17,25	34,226
% removal	88,50%	88,59%

Recently new developments have been made in tackling some of more complex organic compounds present in sediments. PCB and halogenated organic compounds seem to degrade under anaerobic conditions using some yeast strains, and eventually surfactants (Deschênes e.a.,1995). Also TBT contaminated sediments may be treated successfully. After demonstrating the fact that tributyl-tin compounds could be degraded through microbial action (Vanderhaegen, e.a., 1995), new evidence at pilot scale shows that TBT in sediments at a dry matter content of 10 % may be stripped successfully by using an in-line oxidation process. Table 3 shows some results on TBT contaminated sediments. This technique is available at 5 m³ per hour.

6. THE FATE OF HEAVY METALS

The effect of bioremediation on heavy metals could be thought as being negligible. However, the intense changes in the oxido-reduction state during bioremediation has an important effect on their behaviour.

Since these metals in sediments have been in an anaerobic environment, most of it is in the reduced form. Bringing the sediment on the surface and aerating it during the bioremediation phase leads to oxydation. The respiration is significantly governed by the sulphate/sulfide respiration of micro-organisms in the sediment. The oxidation causes a chemical shift of metals between various sinks. Some metals may therefore be mobilised and will be recovered in the drainage water during treatment.

Different sediments have been studied (in collaboration with the centre of Colloid chemistry of the Leuven University). During these experiments quantitative and qualitative speciation was carried out using the Oakley procedure (Oakley, 1980). This showed that during the bioremediation process a phase shift was obtained from the sulfide to the oxide fraction. This fraction becomes the most important sink after treatment

After treatment between 20 and 40 % of the metals remain in the sulfide fraction (Van Cauwenberg, 1998). However, the fraction extractable with EDTA is increased.

Table 4 : Effect of bioremediation on the presence of some heavy metals

Metal	ORIGINAL (ppm)	TREATED SEDIMENT (ppm)	REDUCTION (%)
Cd	2.6	1.3	49.2
Cu	130.6	36.2	72.3
Zn	625.5	243.5	61.1
Pb	153.0	53.1	65.3

This effect of mobilisation can also be reversed. It is true that a sediment under the right conditions: i.e. in the presence of sulphate, carbon donor, anaërobic conditions will immobilise heavy metals in the sulfide form. (De Brabandere, e.a. 1993-1995). This finding has been studied further quantitatively, showing that it takes about 50 days to restore the immobilisation state after oxidation (Harmsen, e.a., 2001).

This means that by correct management of the sediment treatment not only some of the most mobilisable metals will be extracted in a gentle way during the bioremediation step. It is also possible to re-immobilise the remaining fraction after treatment during safe storage of the sediment.

7. CONCLUSIONS

Lagunation and bioremediation now offer a great opportunity to deal with contaminated sediments, which until recent had to be landfilled. This also means that techniques applied in standard soil remediation now also are applicable to sediments and this on an economical viable way.

At the origin and due to its high water content, the sediment is not stable and difficult to handle.

A first priority is the dewatering in multi-layered lagunation fields, or in mechanical dewatering systems. Once a dry matter content of 65% or higher is reached, bioremediation and soil conditioning techniques are available. Based on Flemish environmental regulations it is necessary for most sediments to carry out a second treatment step after lagunation. This is carried out after improvement of the grain size of the material. Regular tilling provides the oxygen needed by the indigenous microflora to degrade the organic pollution present in the sediment.

Re-use of remediated dredged material as soil or filler is regulated by stringent directives. Analysis carried out during and at the end of the bioremediation are helpful in the monitoring of the process. The regulator performs an in depth study based on the file and may issue a proper certificate of re-use for each sediment. Such a re-use certificate for application as soil or non-structural building material opens the way to further use and recycling of the treated material.

REFERENCES

- Anonymous, VLAREA, Flemish Ruling on waste Ministry of Environment of the Flemish government, 1998
- Cauwenberg P. Speciation and behaviour of transition metals in dredged material and their potential removal by flotation. Ph. D. Kuleuven, Laboratorium voor Colloidchemie, 1998 : 161p.
- De Brabandere, J. , Treatment of contaminated dredged material and sanitation of river sediments: a comprehensive approach, Proc. CSCE-ASCE conference on Environmental Engineering, pp. 695-702, 1993.
- De Brabandere, J. and Parker, R. Quantification, remediation and management of contaminated muds, Hydrographic Soc., 1995.
- Deschênes, L., Lafrance, P., Villeneuve J.P., and Samson R. Surfactant influence on PAH Biodegradation in a Creosote-Contaminated Soil, in Microbial Processes for Bioremediation, pp.51-58, Batelle Press, ISBN1-57477-009-8 1995

Detzner, H.D., Treatment of Hamburg Harbour Sediments at the Metha III Treatment Facility, Proc. Sediment Remediation 95, Windsor, Ca, 1995.

Harmsen, J., van den Toorn, A., Bril, J. Natuurlijke immobilisatie van zware metalen in de bodem. Alterra-rapport 357, Alterra, Research Instituut voor de Groene Ruimte, Wageningen, 2001.

Higgins, I.J., The chemistry and microbiology of pollution, Ac. Press London, 1975.

Oakley SM, Delfey CE, Williamson KJ, Nelson PO. Kinetics of trace metal partitioning in model anoxic marine sediments. *Wat Res* 14: p. 1067-1072, 1980

Sebastian A., Gerlach, Marine Pollution, Springer N. York, 1981.

Van Geldermalsen, L. The remediation of the Harbour Elburg, Proc. Sediment Remediation 95, Windsor, Ca, 1995.

Vanderhaegen, B., De Brabandere, J., Dumon, G., Bioremediation of Contaminated harbour sediment II. monitoring of contaminant degradation. Proc. Setac 1995.