

# **Great Lakes Sediments: Contamination, Toxicity and Beneficial Re-Use**



(Courtesy: U.S. Army Corps of Engineers)

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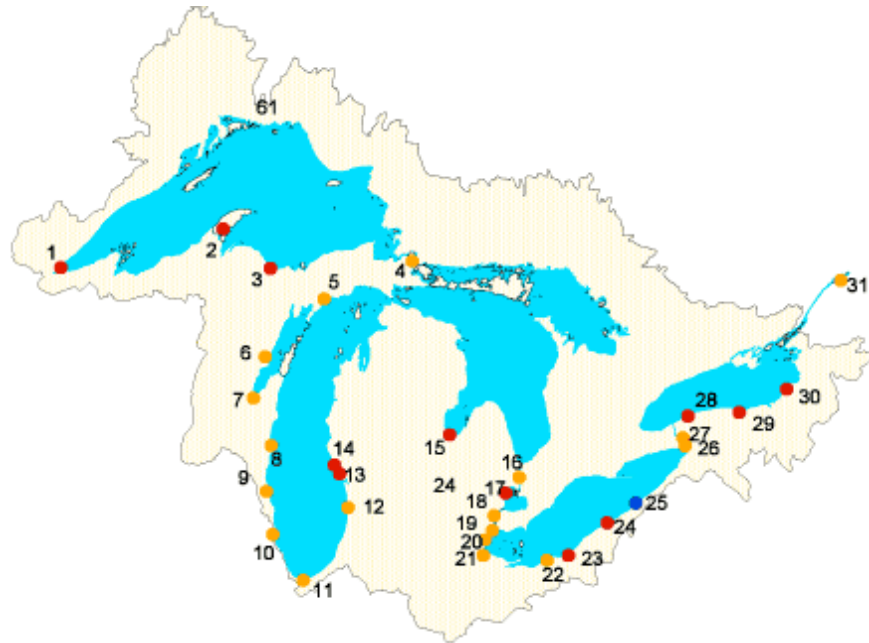
## 1. INTRODUCTION AND LEGAL FRAMEWORK

The Great Lakes are an extraordinary natural resource, holding 95% of the surface freshwater found in the United States, and represent 18% of the world's supply of surface freshwater. This wealth of freshwater sustains abundant and diverse populations of plants and animals, many recreational activities, and the five lakes are a readily available waterway system for economic activity and fisheries.

Years of point and non-point source discharges from industrial and municipal facilities, and urban and agricultural runoff to the Great lakes and its tributaries have contributed toxic substances into the ecosystem, resulting in major contamination issues. In most cases, the contamination is introduced in the tributaries which, via sediment transport and erosion mechanisms, contribute to contamination of the Great Lakes proper. Because of their vast size and volume, less than 1% of the lake waters are flushed annually, resulting in settling out and accumulation of suspended particle-associated contaminants in the water column. Hence, the sediments serve as repositories for and on-going sources of organic and inorganic contaminants, exposing and impacting aquatic organisms, wildlife and humans through the development of cancerous tumors, loss of suitable habitats and toxicity, fish consumption advisories, closed commercial fisheries, and restrictions on navigational dredging.

The programs and policies to restore and protect the chemical, physical and biological integrity of the Great Lakes have been covered under the 1972 joint binational Great Lakes Water Quality Agreement (section 118(c)(3) of the Clean Water Act) between the US and Canada. In 1987, a protocol (Annex 14 – Water Quality Act) was added to the GLWQA to jointly address concerns about persistent toxic contaminants, with specific objectives to: (i) identify the nature and extent of sediment pollution, (ii) to develop methods to evaluate both the impact of polluted sediment on the Great lakes System, and (iii) to evaluate the technological capabilities to remedy such pollution. This information is used to guide Lakewide Management Plans (LaMPs) and Remedial Action Plans (RAMs) for specific Areas of Concern (AOC), previously identified by the Great Lakes Water Quality Board of the International Joint Commission (IJC). Areas of concern are defined as “places where beneficial uses of water resources such as drinking,

swimming, fishing, and navigation are impaired by anthropogenic pollution or perturbation”; 42 out of 43 AOCs were determined to be impaired by sediment contamination. The AOCs (Figure 1) involve 2,000 miles (20%) of the shoreline considered impaired because of sediment contamination and fish consumption advisories remain in place throughout the Great Lakes and many inland lakes.



Awaiting Remediation (red)	More Remediation Needed (orange)	Natural Recovery (blue)
1. St. Louis River	4. St. Marys River	25. Presque Isle Bay
2. Torch Lake	5. Manistique River	
3. Deer Lake	6. Menominee River	
13. Muskegon Lake	7. Fox River	
14. White Lake	8. Sheboygan River	
15. Saginaw River and Bay	9. Milwaukee estuary	
16. St. Clair River	10. Waukegan Harbor	
17. Clinton River	11. Grand Calumet River	
19. Detroit River	12. Kalamazoo River	
23. Cuyahoga River	18. Rouge River	
24. Ashtabula River	20. River Raisin	
28. Eighteen Mile Creek	21. Maumee River	
29. Oswego River	22. Black River	
	26. Buffalo River	
	27. Niagara River	
	31. St. Lawrence River	

Figure 1. Geographical location of the primary areas of concern in the Great Lakes Basin

In response to the GLWQA and the Water Quality Act, the USEPA Great Lakes National Program Office (GLNPO) was authorized to carry out a five year study to address site characterization and assessment, as well as technology demonstration projects in five priority AOCs (Saginaw Bay, MI; Sheboygan Harbor, WI; Grand Calumet River, IN; Ashtabula River, OH; and Buffalo River, NY). This effort was known as the Assessment and Remediation of Contaminated Sediments (ARCS) Program, and was completed in 1994. To date, sediment remediation is not yet complete at any US AOC, due to legal, economic and technological challenges. Subsequently, the Water Resources Development Acts of 1990 (section 412), 1992 (section 405c) and 1996 (section 226) authorized the development of environmentally and economically-acceptable methods for the processing of contaminated dredged materials (for NY/NJ Harbor), several of which are currently under demonstration in the Great Lakes Basin. Not limited to sediment management, Great Lakes Strategy 2002, a plan focused on US Federal, State and Tribal government environmental protection and natural resource management activities was unveiled to address multi-stakeholder environmental protection efforts for the AOCs by integrating them in an overall basin-wide context. Its objectives relevant to sediments are to de-list three AOCs by 2005 (Waukegan Harbor, IL; Presque Isle Bay, PA; Manistique, MI), and a cumulative total of ten AOCs by 2010.

This white paper will detail some of the controlling factors and aspects of sediment contamination (sections 2 and 3), discuss the uncertainty of sediment toxicity endpoints (section 4), and detail the state-of-the-art in current (section 5) and future (section 6) sediment management strategies. A summary (section 7) and key-research needs (section 8), as they emerge in each area will be highlighted.

## 2. SEDIMENT HYDROGEOCHEMISTRY

In considering the Great Lakes sediment burden, four major contaminant migration pathways have to be considered (Figure 2): (a) tributaries which carry nutrients, conservative ions and polychlorinated biphenyls (PCBs); (b) the groundwater-surface water interface which is a dominant pathway for nutrients, pesticides (e.g. atrazine, chlordane), and chlorinated solvents as well as fuel hydrocarbons; and (c) the air-water interface which is a major source of atmospheric

contamination (e.g. PCBs, mercury) as well as a sink for volatilized contaminants from the water column, and (d) agricultural runoff (e.g. pesticides, nutrients).

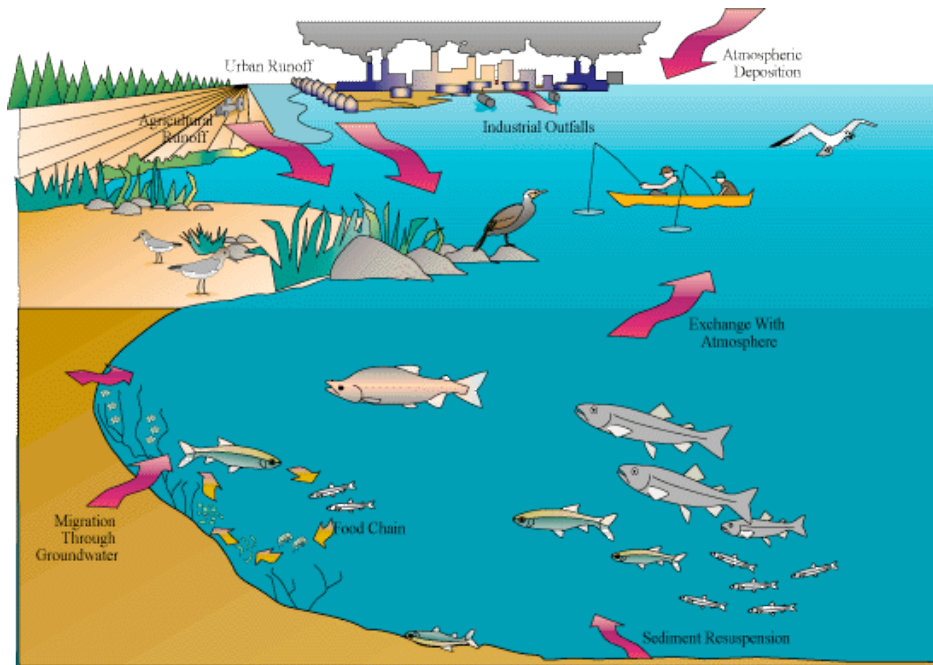


Figure 2. Contaminant migration pathways across environmental interfaces (from Atlas of the Great Lakes, 1995)

Sediment transport via tributaries to the Great Lakes proper has long been considered a primary source of contamination due to direct discharge from industrial operations based along the rivers. The extent of tributary contribution to the sediment contaminant burden is then a function of river bed stability, average water flow, and level and types of contamination. For example, in the case of PCBs, these contaminants are primarily associated with the smaller and high organic silt and clay fractions in the sediment (Moore et al., 1989). These sediments are transported initially as discrete particulates suspended in the water column, then form larger diameter aggregates as a result of physicochemical coagulation or biologically-mediated agglomeration, and settle out at velocities controlled by the flow-associated shear. Further migration is then dependent on the differing flow regimes resulting from horizontal advection and turbulent mixing. Once the flow energies decrease, the aggregates settle to the sediment water interface, forming a loosely consolidated, high water content deposit, often referred to as a “fluff layer” (NRC, 1987). Due to the porosity of this layer (often 2-4 cm thick), resuspension and transport easily ensue as the

result of prevailing flows or tidal effects. Displacement of sediment beyond the “fluff layer” requires boundary shear stresses that occur only during major storms, or shipping. The variety of factors affecting sediment erodibility present a major challenge to predict the response of a given deposit to a specified range of forces. Hence, any information on tributary contributions to the Great Lakes contaminant burden is site-specific, and order-of-magnitude range. Recent innovations such as acoustic profiling can provide high resolution characterization of surficial and sub-bottom sediments (McGee et al., 1995), and help define the thickness and distribution of disparate sediment types (Caulfield et al., 1995). In the overall quantitative mass balancing of sediment transport, resuspension and transport are computed using the output of a hydrodynamic model, and the measured characteristics of sediments (e.g. Buffalo River, Saginaw River, Fox River/Green Bay, Lake Michigan) (USEPA, 1994).

Atmospheric transport has indicated a significant regional environmental impact resulting from re-emission of the sediment burden of PCBs, toxaphene, and organohalogen pesticides into the water column and across the air-water interface. For example, air mass back-trajectory data for organochlorine insecticides and PCBs over the Great Lakes, indicated local or regional volatilization, rather than long range transport (e.g. McConnell et al., 1998). Moreover, significant temperature-dependent air-water exchange of toxaphene (polychlorinated bornanes and bornenes) in the Great Lakes was demonstrated, whereby the colder temperatures and lower sedimentation rates in Lake Superior are responsible for its higher aqueous concentrations (Swackhamer et al., 1999). Further evidence for re-emission of the PCB sediment burden via the water column to the atmosphere was obtained by Jeremiasson et al. (1994) in a mass balance study in Lake Superior.

As an example, The Lake Michigan Mass Balance study (1994-95) was commissioned to provide a coherent, ecosystem based evaluation of toxics in Lake Michigan, with the goal to develop a sound scientific base of information to guide future toxic load reduction efforts. Hence, tributary and atmospheric sources of four pollutants (PCBs, Trans-nonachlor, mercury, and atrazine) were investigated to identify and quantify sources, as well as to develop cause-effect relationships for contaminant loads and bioaccumulation. Eleven tributaries were monitored, and 20 atmospheric monitoring stations were deployed. Examples of results for PCBs and mercury indicate that

atmospheric loadings exceed tributary loadings for both pollutants by a factor of 4-5. Trans-nonachlor exhibited a net export from the Lake.

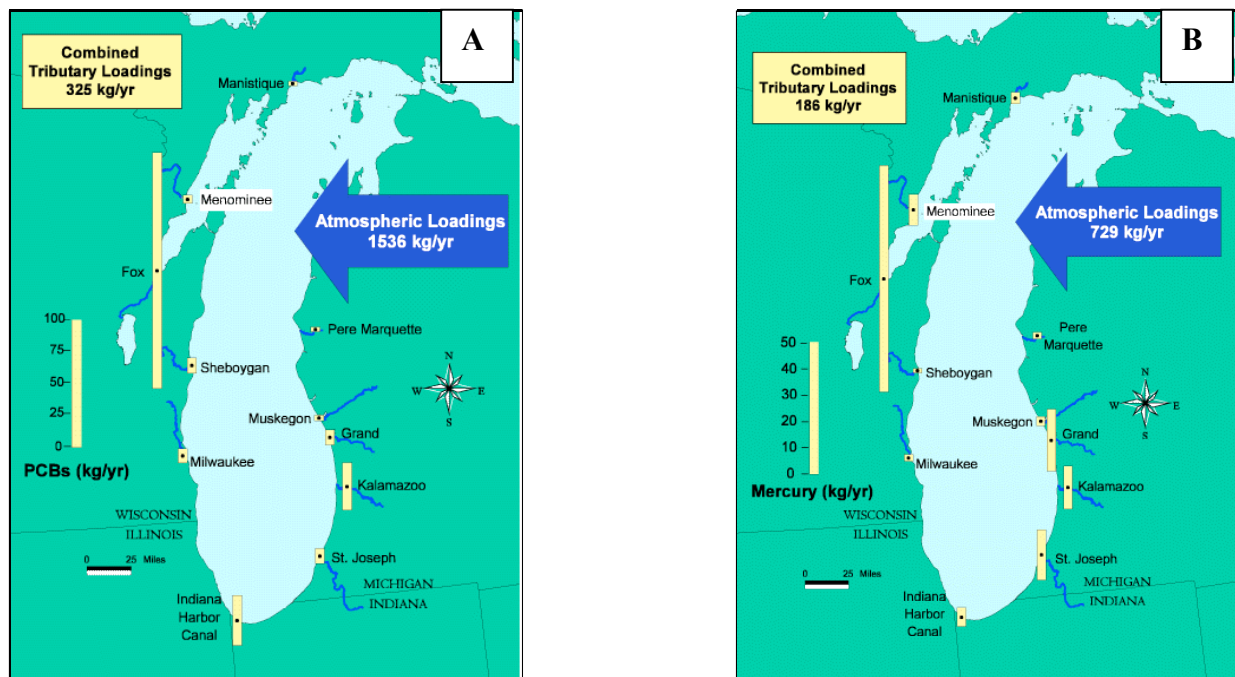


Figure 3. Sample results from the Lake Michigan mass balance study, detailing relative source contributions of PCBs (A) and mercury (B).

This study has now been expanded into the Great Lakes Environmental Database (GLEND), to integrate data entry, storage, and access for mass balance modeling efforts in the future.

### 3. SEDIMENT BIOGEOCHEMISTRY

The deposition of natural and anthropogenic organic matter, as well as heavy metals in the Great Lakes basin has resulted in a complex interaction between sediment hydrodynamics, contaminant profiles, and microbial activity.

#### 3.1. *Organic and Inorganic Contaminant Geochemistry*

The depth profiles, composition and speciation of sediment contamination carry with it a signature of human development along waterways. For example, there are typical correlations between metal accumulation in sediments and specific sources, such as discharges from smelters (Cu, Pb, Ni), metal-based industries (e.g. Zn, Cr, and Cd from electroplating), as well as

chemical manufacturing plants. Organic contaminant profiles for polychlorinated biphenyls (PCBs), polychlorinated dibenzo-p-dioxins, pesticides (e.g. atrazine), and polycyclic aromatic hydrocarbons (PAH) in sediments are often reflective of a combination of known point sources (e.g. insulators, transformers, paper plants,...) and diffuse sources (atmospheric deposition, agricultural runoff). In undisturbed core samples (evaluated by radioisotopes such as Cs<sup>132</sup>), sediment contaminant burdens (concentrations), and isomer- or congener-specific signatures are capable of revealing temporal occurrences of specific source contributions, using statistical tools in the realm of environmental forensics. Since most important point source contributions (often in Great Lakes tributaries) have been identified and closed over the last two decades, the depth concentration profiles of organic and inorganic contamination in the Great Lakes, which had steadily increased for the last 200 years and peaked 20-30 years ago, have started to decrease (Figure 4). Hence, more recent sediments are less contaminated, which renders them more amenable to beneficial re-use after dredging and disposal (section 6).

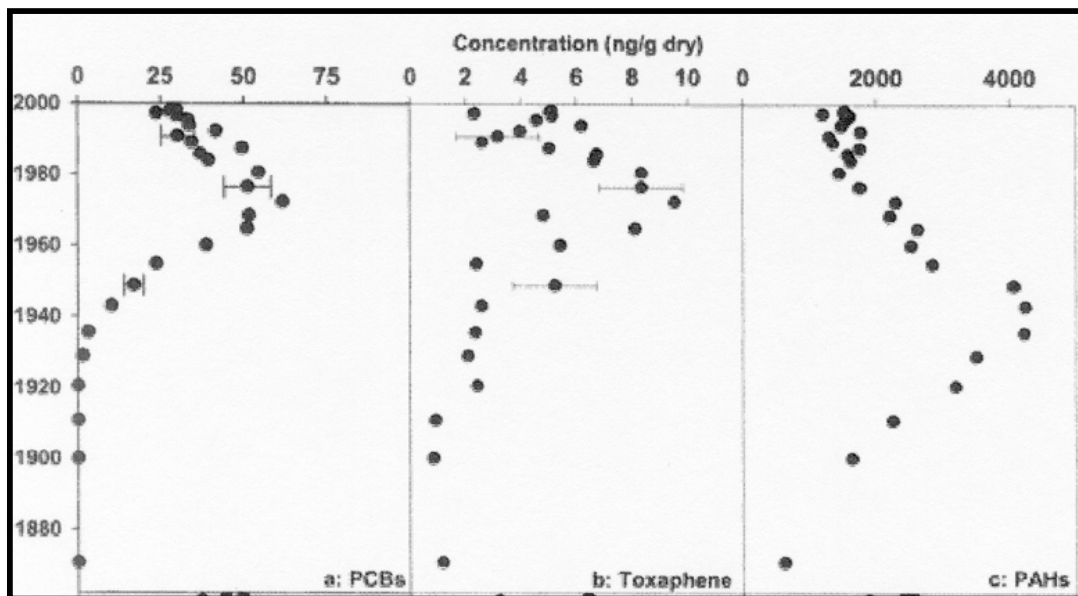


Figure 4. Observed concentrations of PCBs (a), toxaphene (b), and PAH (c) in Grand Traverse Bay sediments (the sediment accumulation rate was 0.1 g/cm.yr, PAH accumulation peaked in 1942, and PCB and toxaphene peaked in 1972 (modified from Schneider et al., 2001).

Heavy metal contamination in sediments is a major issue from a risk assessment perspective as well as from a remedial action perspective, due to disposal limitations and beneficial re-use concerns. Heavy metals (and mercury) are ubiquitous throughout the Great Lakes basin at



concentrations ranging from  $< 1$  mg/kg to  $> 100$  mg/kg. Whereas total concentrations of heavy metals in freshwater sediment environments can be determined with common analytical techniques, the issue of metal toxicity as a component of sediment risk assessment is complex, since the speciation of metals determines their bioavailability to benthic organisms and fish. Various operationally defined fractions of metals in rivers and freshwater catchments for the divalent cations Zn, Cu, Pb, Ni, and Cd: (i) exchangeable metal, (ii) surface oxide and carbonate-bound metal, (iii) Fe/Mn oxide bound metal, (iv) organic bound metal, and (v) residual metal (Forstner, 1990; Malley and Williams, 1997). Depending on the respiratory processes and availability of sulfate as an electron acceptor, a substantial fraction will be present as metal sulfides. Generally, concentrations increase with decreasing sediment particle size (Murray et al., 1999). Thus, zinc was found primarily as Fe/Mn oxides, copper was divided over the Fe/Mn oxide, the organic and the carbonated fraction, and lead in carbonates and oxides (Jackson et al., 1999). Mercury contamination exhibits a complex biogeochemical cycle (particularly in anaerobic sediments) where Hg(II): may reduce to elemental Hg(0), may become methylated, may precipitate as a sulfide. Hence, even though the total concentration may have decreased significantly during the last two decades, the bioavailable fraction (% of total) has not, and metals continue to contribute substantially to sediment toxicity (NRC, 1997; USEPA, 1994).

### 3.2. Organic Carbon Turnover and Redox Profiles

In sediments, the rates of redox zone development in sediment depth profiles are strongly correlated with sedimentation rates, due to the decreased penetration of oxygen which fuels microbial aerobic oxidation of sediment organic carbon. The particulate organic carbon sedimentation rates in north temperate lakes are on the order of 30-160 g C m<sup>2</sup>/yr (Henrichs and Reeburgh, 1987). Oligotrophic lakes, such as Lake Superior experience very low sedimentation rates (1.3 mg C m<sup>2</sup>/yr). The sources and relative quantities of natural and anthropogenic organic matter potentially exert influences on the types and rates of metabolic processes occurring there. Indeed, spatial and temporal variations in microbial processes have been observed in Great Lakes sediments (Figure 5): aerobic and denitrifying activity is confined to the top few centimeters of sediments, sulfate-reduction has been observed over 50-60 cm of sediment thickness, which is underlain by a zone of methanogenic activity (Fenchel et al., 1998; Carlton and Klug, 1990). The depth to which aerobic processes dominate depends on the depth distribution and supply rate

of dissolved oxygen from the overlying water column, and the respiratory consumption within the sediments. Up to 60-70% of natural organic matter incorporated in anoxic sediments ultimately becomes degraded via fermentation, and other anaerobic respiration mechanisms. Freshwater sediments tend to be predominantly methanogenic due to the limited input of sulfate.

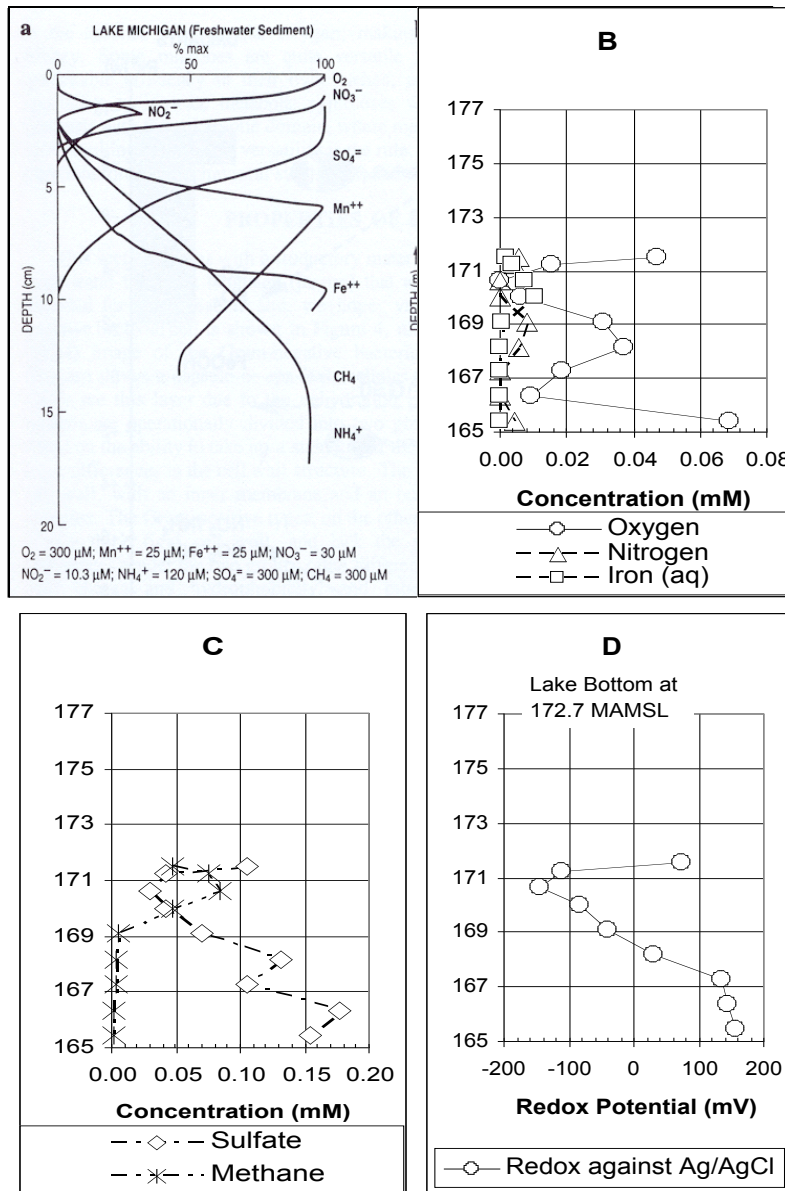


Figure 5. Modeled (A), and measured (B, C) profiles of electron acceptors and redox (D) in Lake Michigan sediments (Lendvay et al., 1998; MAMSL, meters above mean sea level).

Organic carbon respiration fluxes under these various terminal electron accepting processes are on the order of  $10^2$ - $10^3$  mmol C (as CH<sub>2</sub>O)/kg/yr (Murphy and Schramke, 1998). The organic

carbon turnover fluxes are dependent on temperature, seasonal impacts, and depth, as reflected by oxygen and microbial respiration index profiles (Carlton and Klug, 1990). Under the prevailing respiratory conditions in freshwater sediments, aerobic and anaerobic degradation of sediment-associated contaminants will occur as well, to various extents, depending on the chemical characteristics of the contaminants and the metabolic capability of the sediment microbial populations and communities (Adriaens et al., 1999; Adriaens and Barkovskii, 2002; Adriaens et al., 2002). These natural bioattenuation processes may impact sediment toxicity from contaminants through contaminant degradation, solubilization, or sequestration, depending on the pathways used, and the distribution of metabolites produced.

#### 4. SEDIMENT TOXICITY ASSESSMENT

Contaminated sediments can pose risks to public health and the environment, and sound decisions about health and ecological risks must be based on formal assessment of those risks. The most elemental form of risk assessment is intended to determine whether the concentrations likely to be encountered by organisms are higher or lower than the level identified as causing an unacceptable effect (NRC, 1997). In this context, an effects assessment is a determination of the toxic concentration and the duration of exposure necessary to cause an effect of concern in a given species. Hence, contaminated sediments are considered to be a problem only if they pose a risk above a toxicological benchmark, which can be identified through a risk assessment, and management strategies must be identified that reduce risk to the benchmark value.

As part of the ARCS Program, a comprehensive human health and ecological risk assessment framework was developed (USEPA, 1994). The goal of that study was to provide estimates of changes in potential exposure and risk that may occur either under a no-action alternative or following implementation of various remedial alternatives for contaminated sediments (selective sediment removal, capping of hot spots, source control, and dredging of an entire river). The risk assessment endpoints used for human health impacts included exposure (populations, pathways, exposure point concentrations, and intake rates) and toxicity (carcinogenic, non-carcinogenic) estimates. Ecological risk assessment requires the consideration of multiple species (wildlife, aquatic plants, benthic invertebrates and fish), other physical-chemical stressors in addition to

toxic chemicals, more complex ecological structures (populations, communities, ecosystems) and different endpoints (e.g. survival, growth, and reproduction). Furthermore, the ability of the ecosystem to recover from the stress may also be considered.

Hence, the selection of ecological assessment techniques to be applied at a given AOC in the Great Lakes Basin includes: (i) chemical analysis of samples of sediment, surface water, and organism tissues from the site; (ii) toxicity testing of sediments; (iii) community analysis based on measurements of the types and number of benthic macroinvertebrates at the site; (iv) exposure models to predict chemical concentrations and bioavailability in environmental media, and to estimate uptake by key-receptors; (v) ecological models to extrapolate from measurement endpoints to assessment endpoints in receptor groups for which community analysis is not a primary tool. These assessment techniques can then be used to evaluate remedial alternatives at contaminated sediment sites, using a comprehensive mass balance modeling approach, to describe each of the underlying mechanisms causing change in the system (Figure 6).

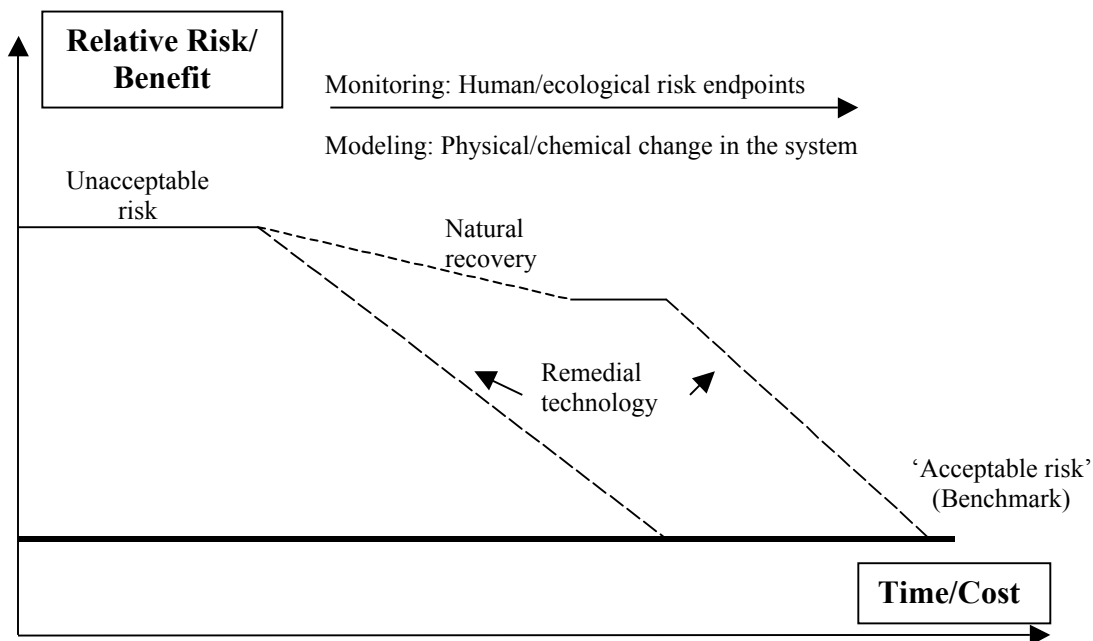


Figure 6. Risk characterization principles for contaminated sediments

#### 4.1. Bioavailability and Exposure Pathways

From a risk assessment perspective, bioavailability of sediment-associated contaminants can be defined as “the fraction of the total contaminant in the interstitial water and on the sediment

particles that is available for bioaccumulation”, whereas bioaccumulation is “the accumulation of contaminant concentration via all routes available to the organism” (Landrum and Robbins, 1992). Bioavailability is generally affected by (i) contaminant characteristics (e.g. octanol-water partition coefficient,  $K_{ow}$ ), (ii) the composition and characteristics of the sediments (e.g. organic carbon content, particle size distribution, clay type and content, cation exchange capacity, and pH), and (iii) the behavior and physiological characteristics of the organisms (e.g. organism behavior and size, mode and rates of feeding, source of water – interstitial vs. overlying – for respiration).

Bioavailability of sediment-associated contaminants is generally assessed either by comparison of sediment and organism concentrations (steady state ratios or accumulation factors), or by determining the uptake clearance (in units of g sediment/g organism.hr). Mass balance box models (sediment solids, interstitial water, unavailable contaminant), which include aqueous uptake, feeding, and excretion, as well as adsorption/desorption functions are commonly used to quantify accumulation of contaminants in the species under consideration.

#### 4.2. Sediment Toxicity Assessment

Effects-based testing is currently the primary means of sediment quality evaluation, and is a basic tool for estimating the risk of various sediment management techniques to the aquatic environment (NRC, 1997; Giesy and Hoke, 1990). Organisms used for freshwater toxicity assessment include bacteria (*Microtox*), algae (*Selenastrum, capricornutum*), cladocerans (e.g. *Daphnia*), insects (*Chironomus tentans*), and fish (e.g. *Pimephales promelas*). Several of these indicator species have been used to map toxicity in the Lower Detroit River, Western Lake Erie and Toledo Harbor, and the Trenton Channel (e.g. Giesy and Hoke, 1990). To supplement effects-based testing, the EPA has published sediment quality criteria (SQC), based on equilibrium partitioning modeling to predict porewater concentrations of non-polar organic compounds (USEPA, 1993).

It should be noted that it is not possible to make a direct comparison between both approaches, since the effects-based approach (using test species exposed to sediments) tests the effects of all contaminants and their potential interactions, whereas SQC are contaminant-specific. Recent attempts to apply contaminant (metals, PAH, chlorobenzenes, PCBs)-specific effects on the 10-

day amphipod (*Hyalella azteca*) and chironomid (*Chironomus tentans*) produced 8-52% false positives and 10-23% false negatives when exposed to contaminated sediments (Becker et al., 2002). Of all sediment quality tests (5) used, the apparent effects threshold (AET; USEPA, 1989) exhibited the greatest accuracy when compared to biomass and survival endpoints determined for both species. Nevertheless, the outcome of species-specific toxicity testing on sediment interstitial waters can then be used to create maps of vertical and horizontal sediment toxicity, and to help guide the selection of remedial technologies, or to calculate the sediment volumes which would have to be removed to improve the quality of the benthic habitat to a specified level.

#### 4.3. Remedial Options and Risk Characterization

Sediment removal, natural recovery, and disposal technologies each exhibit associated risk characteristics. For example, *non-removal technologies* (e.g. in situ capping, containment or treatment) are governed by the potential loss of contaminants in situ and thus their enhanced bioavailability to benthic macroinvertebrates and fish. For example, resuspension and advection during cap placement, and long term diffusion, advection, bioturbation, and erosion are the dominant loss mechanisms during in situ capping. Hence, information on cap integrity and sediment bed stability will be required as primary monitoring variables for this technology application. Similar processes will impact both other non-removal technologies. Particulate, dissolved and volatile contaminant releases represent the major loss mechanisms during *dredging operations*, and thus the risk associated with this activity has to be compared relative to leaving the contaminated sediments in place.

Lastly, disposal technologies have more mechanisms for contaminant loss than most other remediation components, due to volatilization, plant uptake, dispersion of dust, bioturbation, leaching and seepage (Figure 7). The potential for the various loss mechanisms should be evaluated in the laboratory or using model predictions, and appropriate design modification put in place. Particularly pathways involving movement of large volumes of water (e.g. effluent during hydraulic filling) have the greatest potential of releasing significant quantities of contaminants from confined disposal facilities (CDFs).

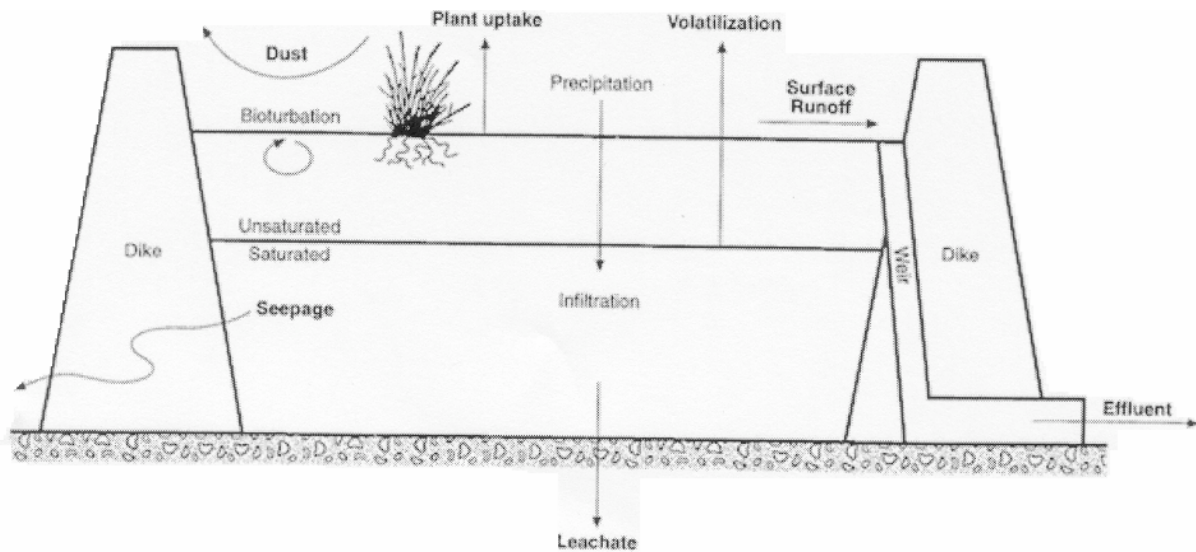


Figure 7. Loss mechanisms and pathways from a CDF.

Even though no formal guidelines are available to measure emission losses from CDFs and other remedial approaches, modeling approaches to estimate volatile losses from chemical vapor equilibrium concepts and fundamental transport phenomena, lysimeter testing protocols (surface runoff), and column settling tests (effluent losses) have been developed for this purpose.

## 5. SEDIMENT CONTAMINANT MITIGATION

During the last 30 years, navigational and remedial dredging in the Great Lakes Basin have generated in excess of 70 M. cubic yards of contaminated sediment in need of sustainable management practices. These contaminated sediments require high volume management approaches, which can be broadly classified in removal and non-removal (*in situ*) strategies. Any decision to leave sediments in place is highly dependent on an evaluation of the relative risks posed by the sediments left untreated on the bottom, the risks of performing a treatment operation on in situ sediments, and the risks associate with the removal and subsequent disposal or treatment of the contaminated dredged material (NRC, 1997). Considering the extent of Great Lakes sediment contamination, open water disposal became impossible by the early 1970s, and hence, confined disposal and treatment technologies have to be considered. Between 1993-96, open water disposal was applied with 32% of uncontaminated Great Lakes sediment, and 12% was used for beach/littoral nourishment. Currently, dredging and confined disposal is chosen in

>90% of all contaminated sediment management options, the remainder being in situ capping, and natural recovery. No full scale in situ treatment strategy is considered, and two sites apply some form of ex situ destruction or immobilization technologies. The various dredged material management alternatives applicable for Great Lakes contaminated sediments will be briefly discussed below.

### 5.1. Non-removal Technologies

Non-removal technologies are those that involve the remediation of contaminated sediments in situ (i.e. in place), and include *in situ* capping, *in situ* containment, and *in situ* treatment (Figure 8, A). These alternatives do not require sediment removal, transport, or pretreatment.

In situ capping is the placement of a cap or covering over a deposit of contaminated sediment. The cap may be constructed of clean sediments, sand, gravel, or may involve a more complex design using geotextiles, liners and multiple layers (Zeman et al., 1992; Palermo and Miller, 1995; Palermo, 1998). Capping has become one of the few accepted management techniques, despite a dearth of knowledge on long-term chemical fluxes into the overlying water column, verification methods for cap thickness, and available monitoring data on cap integrity. However, from a regulatory perspective, capping is not currently considered a permanent solution, but rather a ‘preferred remedy status’ amenable to technologies geared at toxicity reduction (NRC, 1997). Indeed, capped sediments may lend themselves for innovative technology applications, such as nutrients and gas amendments to stimulate biodegradation. A limited number of in situ capping operations have been accomplished in recent years under varying site conditions. An example relevant to the Great Lakes is a composite cap with layers of gravel and geotextile to cover PCB-contaminated sediments in the shallow water and floodway of the Sheboygan River.

In situ containment involves the complete isolation of a portion of the waterway, using physical barriers such as sheetpile, cofferdams, and stone or earthen dikes. The isolated area can then be used for the disposal of other contaminated sediments, treatment residues or other fill material, and is often modified to prevent contaminant migration (e.g. slurry walls, cap and cover). A number of Great Lakes sites have been treated using containment, such as the Crotty Street PCB site in the Saginaw River, and the Waukegan Harbor Superfund site. The latter included the

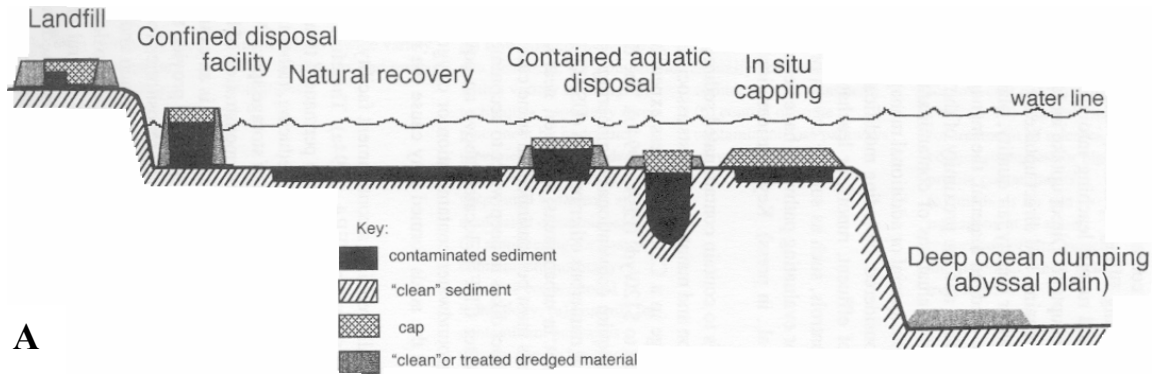


isolation of 15,000 cubic yards of PCB-contaminated sediment in a boat slip, using bentonite slurry walls, and bentonite-filled sheetpile cutoff walls.

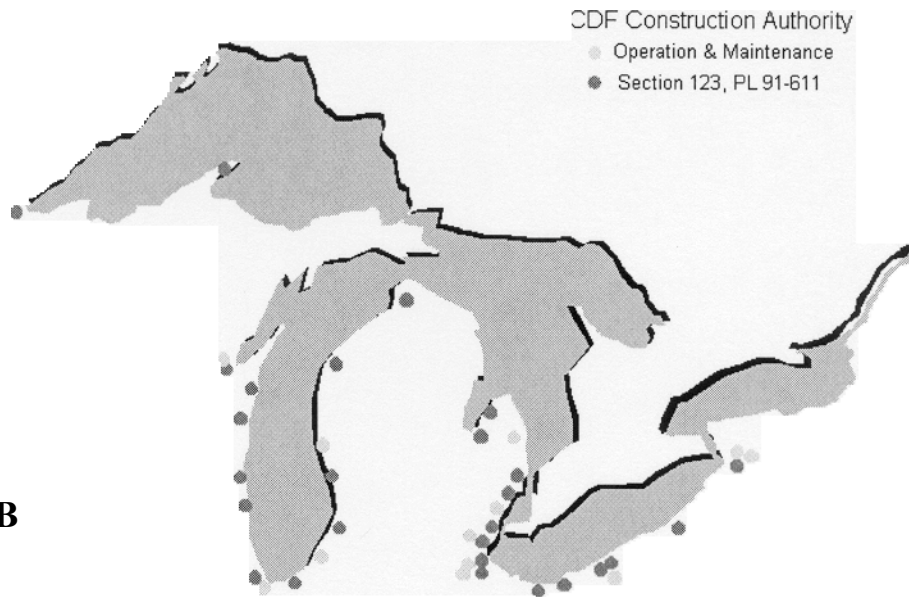
In situ treatment technologies include chemical (USEPA, 1990), biological (Murphy et al., 1993), and immobilization (Myers and Zappi, 1989) approaches as process control features for contaminant degradation. The main difficulty with in situ treatment is the determination of efficacy of a given process, considering the nonhomogenous distribution of contaminants, sediment physical properties and chemicals. Moreover, one of the most significant limitations to technology application is the impact of the process on the water column. Processes that would release contaminants, reagents or heat, or otherwise produce negative impacts on the overlaying water are not acceptable and, hence, suitable applications have to be compatible with a cap or create minimal interference with the sediment-water interface. For example, 1.4 hectares of PAH-contaminated sediments in Hamilton Harbor were injected with calcium nitrate to reduce hydrogen sulfide toxicity and stimulate biodegradation of low molecular weight compounds and PAHs (Murphy et al., 1993). In situ solidification/stabilization has been demonstrated in contaminated sediments at Manitowoc Harbor (WI), using cement/fly ash slurry injection technology. The process resulted in treated vertical columns (diameter 6 feet; 1.8 m) to a depth of 6 meters below the river bed using a steel cylinder placed 1.5 meters into the sediments. Limitations of this approach included difficulties in solidification, and management problems with liberated pore water.

### 5.2. Disposal Technologies

Disposal is the placement of contaminated dredged material into a site, structure or facility on a temporary or permanent basis, and includes open-water disposal (level bottom capping, and contained aquatic disposal), and confined disposal (Figure 8, A). Open water disposal of contaminated sediments involves a containment strategy such as capping the contaminated materials into a natural excavated depression or trench, such as those formed from sand mining in near shore areas of the Great lakes.



**A**

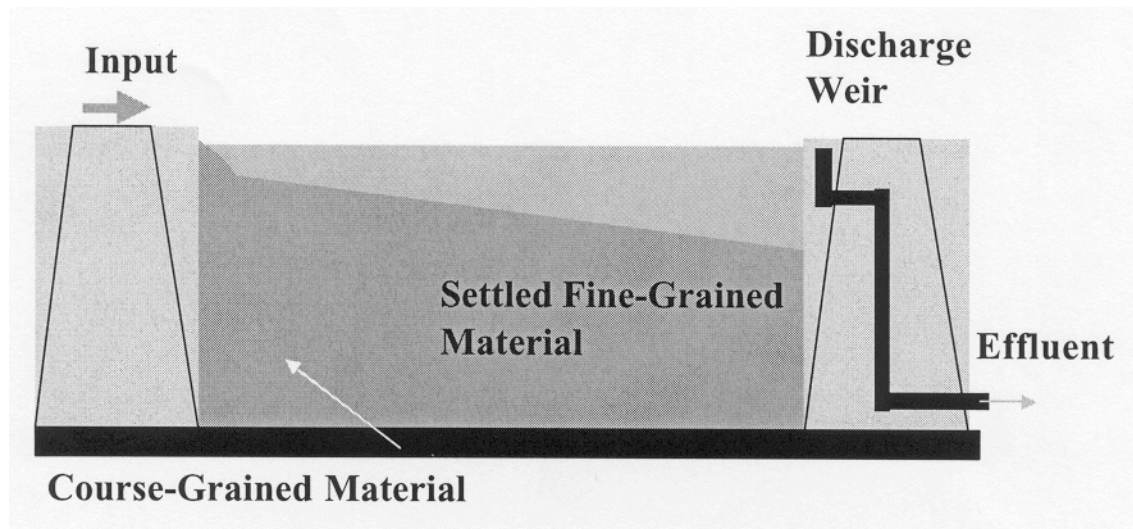


**B**

*Figure 8. Conceptual representation of containment, disposal and natural recovery technologies (A), and locations of Great Lakes confined disposal facilities (B)*

Confined disposal includes the placement of contaminated materials in commercial landfills or confined disposal facilities (CDF). In the Great Lakes Basin, 44 CDFs have been built by the U.S. Army Corps of Engineers solely for the disposal of contaminated dredged material, with the majority located in Michigan (Figure 8, B). These facilities are diked structures, designed to receive physically homogenous material that may be 10-50% solids by weight. The most commonly used management practice for contaminated sediments dredged for navigational and environmental remediation (IJC, 1997; USEPA, 2000), confined disposal takes place in upland

or in-water structures. Confined disposal facilities are designed based on an evaluation of potential pathways (Figure 7) by which contaminants associated with dredged material might impact surface water, ground water, air, plants, and animals (USACE/USEPA, 1992). Using laboratory tests, the significance of contaminant migration pathways are determined, and appropriate controls (e.g. liners, water treatment, caps) designed. In terms of treatment technology, CDFs function as settling basins (Figure 9), whereby the coarse materials (sand and gravel) settle rapidly near the point of disposal, and fine grained sediments (silts and clays) will require more time. Water is drained or discharged passively through the dikes, or via engineered release mechanisms (weirs, etc...). During the time of its operation, extensive monitoring programs have been put in place to investigate the effect of dredged materials on fauna and flora (Stafford et al., 1991), contaminant (PCB) losses from in-lake CDFs (Myers, 1991).



*Figure 9. Operational flow in a CDF*

Over the last 30 years, twenty three of the 44 Great Lakes CDFs have been filled or have less than 10% of their capacity remaining. At the same time, there is (are):

- (i) Continued demand for CDFs to manage contaminated dredged material for navigation
- (ii) Increased demand to manage contaminated material from remedial dredging
- (iii) More stringent environmental requirements for new CDFs, raising their cost
- (iv) Fewer ports and local governments are capable of sponsoring new CDFs

Several options are being considered or have been implemented to increase the capacity of CDFs: raising the dikes, increase consolidation through aggressive dewatering, particle separation (contamination is mainly associated with fine grained material), and remove material

from the CDF. The latter option has been one of the main driving forces behind innovative technology development for beneficial re-use of the stored dewatered sediments.

## 6. BENEFICIAL RE-USE CONCEPTS AND APPLICATION TO GREAT LAKES SEDIMENTS

### 6.1. *Framework*

Methods for the handling and disposal of dredged material have been studied and developed for many years, and several literature reviews are available to help select the successful complete treatment trains (e.g. <http://www.bnl.gov/wrdadcon/publications/reports>; Kraus and McDonnell, 2000). The former reference pertains to the Water Resources Development Acts (WRDA), which set forth a program consisting of a series of progressive steps to lead to a full-scale demonstration of one or more decontamination technologies with a processing capacity of at least 500,000 cubic yards per year (WRDA 1990; 1992; 1996). The WRDA Decontamination Program draws on many disciplines, ranging from the basic science and engineering fields to support technology development, as well as marketing and commercialization related to beneficial use of the decontaminated materials. Even though the primary emphasis of the Acts was to address the dredging needs of the New York/New Jersey Harbor region, its results are applicable to other ports and harbors in the United States. Currently, applications of selected alternative technologies are under consideration or demonstrated at the pilot scale in the Great Lakes Region (EPA-GNPO Workshop, 2001).

The topics that enter into the overall WRDA Decontamination Program include: (i) Bench-, pilot, and full-scale technology testing and evaluation; (ii) Design of an integrated treatment train; (iii) Commercial-scale design engineering; (iv) Commercialization of decontamination technologies; and (v) Public outreach program/Citizen's advisory committee (CAC).

### 6.2. *WRDA Technology Integration*

Sediment decontamination ties together a series of operations starting with removing sediments, and finishing with production of a material that is suitable for beneficial use options. Hence, the reuse of dredged material requires a centralized containment facility to dewater sediment (e.g.

CDF), and a location for rehandling. The objective is then to provide and develop viable methods for incorporation into the decontamination and beneficial use portions of a conceptual treatment train as shown in Figure 10. An overview of the technologies considered in the WRDA Program is provided in Table 1. Complementary, in order to proceed with a concerted reuse/rehandling approach, a comprehensive management strategy needs to be developed that addresses the end-user community and public perceptions.

### 6.3. Technology Description

*Manufactured Soil (U.S. ACE-WES).* The manufactured soil is created by blending cellulose waste solids (yard waste compost, sawdust, woodchips) and biosolids (cow manure, sewage sludge) with the as-dredged sediment. Its inherent simplicity makes this an attractive option, requiring initial contaminant concentration reductions through dilution from the addition of materials needed for soil formation. The suitability of the soil for growth of different plant species was tested for tomato, marigold, ryegrass, and vinca, and indicated that a viable soil was formed. Alternative applications include recreational fields at Pearl Harbor (Hawaii), landscaping throughout the city of Toledo (Ohio), and as cap material at Brownfield and Superfund sites. The N-Viro Company produces soils using biosolids, kiln-dust, and fertilizer augmentation to produce a potting and topsoil product sold to the public. To date, there only have been small scale projects, and full scale production would require a large area.

*Solidification/Stabilization (WES, International Technology, Metcalf and Eddy, Inc.).* This is a treatment approach that creates solid aggregates from dredged material by addition of Portland cement, fly ash, lime, and cement kiln dust. After blending, the material is allowed to set into a hardened granular soil-like condition with a lower water content and improved structural/geotechnical properties. The contaminants become more tightly bound to the sediment matrix by chemical and mechanical means, thus preventing leaching and minimizing bioavailability. Beneficial uses for this product include structural and non-structural fill, grading material, daily/intermediate landfill cover, Brownfield redevelopment projects, and final landfill cover. No performance standards have been set at this time, but field-scale applications have been conducted in Japan to bottom sediments, and in the U.S. to industrial wastes and dredged sediments from New York/New Jersey and Boston Harbors.

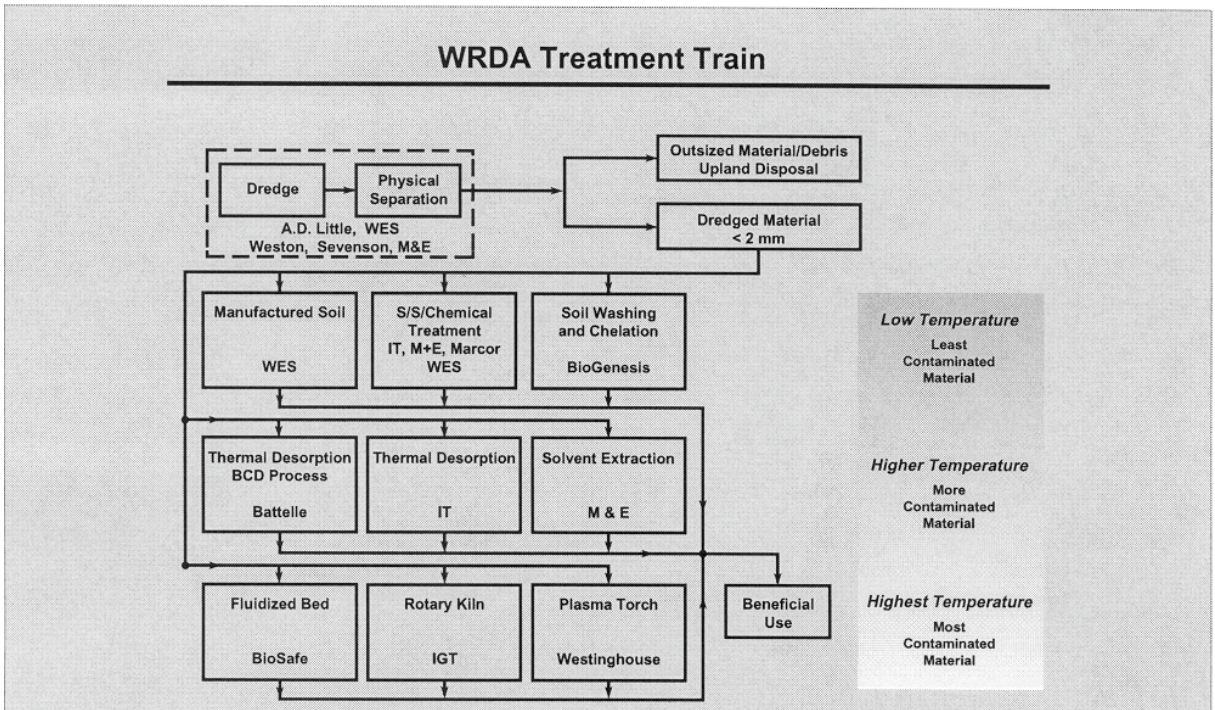
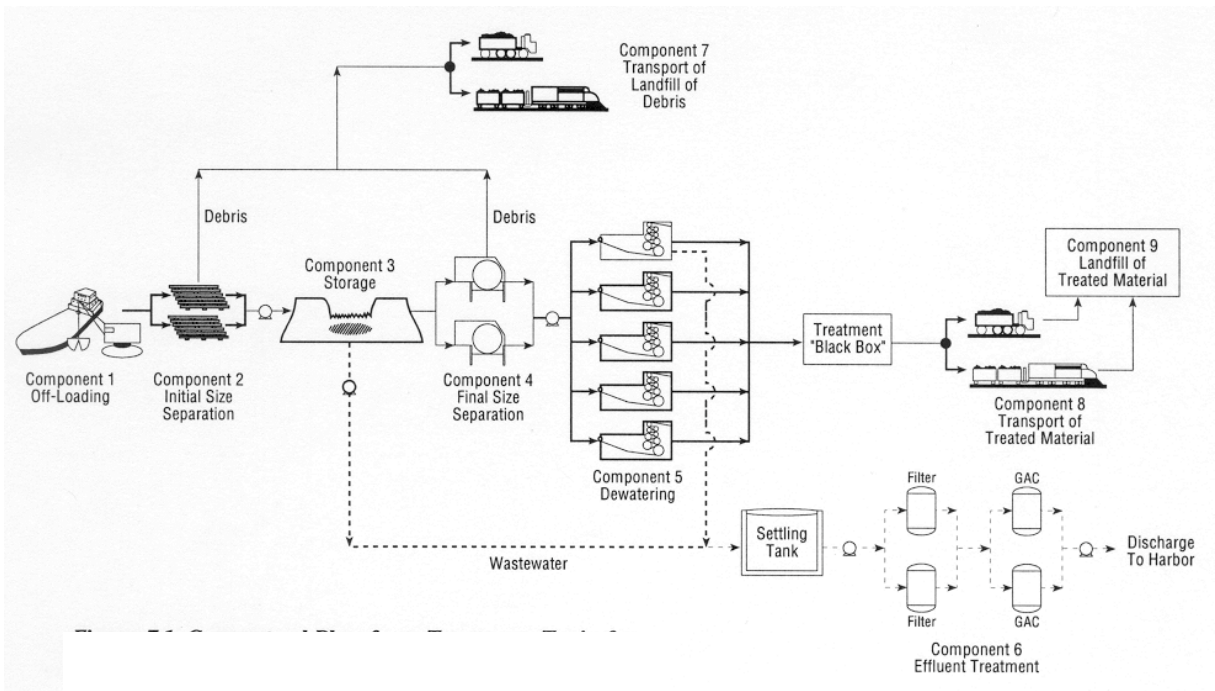


Figure 10. Conceptualization of a treatment train for dredged material (A), and incorporation of beneficial reuse technologies (B) (Adapted from EPA 000-0-99000)

*Sediment Washing (Biogenesis)*. This technology uses a proprietary blend of surfactants , chelating and oxidizing agents, and high pressure water jets to remove both organic and inorganic contaminants from the dredged material. The chelating agents serve to render the metals soluble, and the organic contaminants in the liquid phase are treated and destroyed by

cavitation-oxidation. Floatable organic material is separated by surface skimming in flotation tank, and metals are precipitated in the form of a sludge which is disposed of in a landfill. The technology approach is simple in concept, but relies on a solid understanding of sediment chemistry and particle-contaminant interactions in the liquid and solid phases. The end-material can be combined with humates, lime and other organics to form a manufactured soil. Large pilot-scale demonstrations have been conducted on NY/NJ materials.

*Base-catalyzed decomposition (BCD, Battelle).* This approach is an enhanced thermal desorption technology which removes (halogenated) organic contaminants from the dredged material and then passes them through a second stage where dehalogenation occurs in the presence of hydrogen-donating oil, sodium hydroxide and a catalyst, at elevated temperatures (340°C). PAH and metals were not substantially removed during technology application, and considering the elevated temperatures, complex material handling and pollution control systems will be required to treat the various side streams to minimize environmental emissions.

*Thermal destruction (BioSafe, Institute of Gas Technology, ENDESCO, Westinghouse).* As indicated in Table 1, these technologies aim at the destruction of organic materials in the dredged material, either using fluidized bed technology, rotary kiln, or plasma torch technology. All organic materials are converted to inorganic by products such as carbon monoxide, carbon dioxide, hydrogen, methane (a fuel gas which can be recycled), oxygen and dinitrogen gas. The metals are incorporated in the remaining product (e.g. glass) or removed in gaseous side streams. Beneficial end-products include construction-grade cement for cement blocks and paving material, glass aggregates and tile products.

#### 6.4. Application to Great Lakes Sediments

A number of technology demonstrations for treatment of Great Lakes contaminated sediments have indicated the application of WRDA technologies in freshwater environments. An early application was the Contaminated Sediment Treatment Technology Program (CoSTTeP), a sub-program of the Canadian Great Lakes Cleanup Fund (GLCF), which has evaluated six technology categories at 5 sites in Canada (Hamilton Harbor, Thunder Bay, St. Marys River, Welland River-Niagara, Toronto Harbor), including: (i) pre/post treatment; (ii) non-incineration

thermal treatment; (iii) chemical treatment; (iv) metal removal; (v) biological treatment; and (vi) solidification/stabilization (1990-97). All sites considered were non-PCB sites, and were contaminated with creosote, polycyclic aromatic hydrocarbons (PAH), and metals. The results, reported in <http://www.AboutREMEDATION.com>, have indicated that the technologies have difficulty competing with landfill options, on a cost basis. No assumptions were made with respect to cost recovery due to beneficial re-use, marketing and commercialization.

The State of Michigan-Department of Environmental Quality (M-DEQ) has been working with the GLNPO to investigate the application of beneficial treatment technologies for treatment of contaminated Black Lagoon sediments in the Trenton Channel of the Detroit River. Following favorable reviews of bench-scale studies (Cement-Lock, Biogenesis Soil Washing, and plasma vitrification), a pilot scale demonstration of the Cement Lock technology using 2,000-5,000 cubic yards of sediment will be conducted, with the objective of delivering a marketable final product for beneficial use. The sediments are contaminated with oils and grease (18,000 mg/kg), PAH (51 mg/kg), PCBs (11 mg/kg), and heavy metals (As: 7.8 mg/kg; Cd: 9.5 mg/kg; Cr: 138 mg/kg; Cu: 180 mg/kg; Pb: 218 mg/kg, and Hg: 0.55 mg/kg). Bench scale studies indicated that the cement product exhibited levels below detection limit for all chemicals.

Similarly, the State of Wisconsin is starting a pilot-scale demonstration of the Minergy plasma torch vitrification process on 70 tons of PCB-contaminated sediments, with the objective to destroy the contaminants and produce 30 tons of useable end-product (glass). In collaboration with the U.S. Army COE, the University of Wisconsin – Center for Byproducts Utilization is evaluating the applicability of commercial top soil products from dredged materials from Milwaukee and Green Bay, with the objective to grow corn, sunflower, sorghum, ryegrass, and clover. The sediments are contaminated with PCBs and PAH.

#### *6.5. End Users, Public Perception and Risk*

Industrial, municipal, and commercial users make up the majority of end-users of dredged material as a beneficial resource. Industrial users represent the largest sector in terms of numbers of potential users (POAK, 1999), while municipal users are the largest on a per cubic yard basis (USACE, 1999). Commercial users have embraced dredge material on a much smaller basis,



and have generally used pilot projects to determine marketability of products. A recent study of the end-user communities in the San Francisco Bay area, which included a detailed survey of the construction and redevelopment community, indicated very little enthusiasm to utilize dredged material as a resource. Perception of re-used dredged material as hazardous or a health hazard after contaminant destruction and stabilization is a major issue that needs to be addressed via a concerted outreach effort (EPA 000-0-9000).

One major effort to address these issues is a proposed framework for evaluating beneficial uses of dredged material in NY/NJ harbor (Bonnievie et al., 2001). This framework is consistent with the current US Army COE Dredged Material Management Plan (DMMP), to provide economically cost-effective, and environmentally sound management practice to satisfy the need for safe navigation. Since the DMMP presents a strong preference for management options that result in beneficial use of dredged material, the framework is intended to incorporate economic, environmental and policy-related information that would be supplemental to a standard cost-benefit analysis. To encompass the diverse array of potential benefit types, a wide range of assessment endpoints related to potential for environmental risk (economics, human health, ecological health, and resource management) are included. Considering that all stakeholders, including citizen groups, are involved, it is hoped that a normalized weight-of-evidence approach to beneficial reuse may address real and perceived impacts.

## 7. SUMMARY

Even though most sources of organic and inorganic contamination impacting the Great Lakes Basin have been identified and addressed, historic contamination and continuing contributions from tributaries and atmospheric composition indicate that sediment management strategies for navigational and remedial reasons are here to stay. On-going work to develop contaminant mass balances for the Great Lakes, and improved integrated risk assessment models will aid in the selection of appropriate sediment management strategies. The current emphasis on dredging and disposal in the Great Lakes AOCs is likely to continue, considering the uncertainties associated with mass balance estimations and incomplete risk characterization based on a myriad of endpoints. However, legal, economic and technical difficulties limit the potential to expand

these facilities, and ,hence, there is an increased need to focus on beneficial re-use strategies for industrial, commercial and public end-users. Many end-users and technologies have been identified for dewatered dredged sediments, but most have only been demonstrated at the bench or pilot scale. Implementation of marketing and commercialization strategies for the end-products will require further communication of associated risks and benefits to the end-user through interaction with NGOs and local, State, and Federal authorities.

## 8. RESEARCH NEEDS

The recommendations for future research needs to address the issues associated with sediment contamination will be largely drawn from three resources: the final summary report of the ARCS Program (USEPA, 1994), the NRC report on contaminated sediments in ports and waterways, and the EPA Workshop on Great Lakes contaminant sediment remediation held last year in Ann Arbor. Based on these reports, the research needs can be divided along sediment assessment, risk assessment, treatment technologies, and public involvement/education.

### *8.1. Sediment Assessment*

The ARCs program has advocated that an integrated assessment approach, including chemical analysis, toxicity testing, and benthic community surveys, be used to define the magnitude and extent of sediment contamination. Here, semi-quantitative screening level analysis to delineate the boundary conditions of the problem, and linkages/correlations between total sediment toxicity and contaminant-specific toxicity are needed, to quantify the importance of bioaccumulation under varying conditions and scenarios. Considering the high volume/low contamination scenario typically encountered in contaminated sediments, rapid throughput contaminant and microbial screening technologies are required to process large numbers of samples. In the area of sediment toxicity, suitable endpoints need to be refined, to enable proper comparisons between 'before' and 'after' remediation scenarios, as well as to aid in priority ranking. Part and partial to sediment toxicity assessment is the issue of beneficial re-use; the proper tests have to be developed or applicable to sediment material treated according to these innovative technologies, and the results properly and convincingly communicated to the various end-users. An important aid in both technical and non-technical communication is the need for

visual presentation of the data, and for quantifiable uncertainty associated with the measurements, and geographical distribution of chemicals and toxicity. The expertise in various aspects of this topical area is well represented at UM between the College of Engineering, the School for Public Health, SNRE, and Geology.

### *8.2. Risk Assessment*

Risk reduction strategies have meaning only if the baseline risk can be properly evaluated and quantified, and if predictive assessments can be incorporated. Since the approaches to quantify risk, and to help provide a scientific basis for making remedial response decisions, often rely on complex series of mass balance models, there is a need to quantify and propagate the associated uncertainty with contaminant mass and loss pathway estimates. Within this context, and for economic purposes, uncertainty analysis can also aid in defining the minimum amount of data that will be required to achieve acceptable levels of uncertainty, or aid in future sampling plans. Further, since risk is based on exposure and loss pathways, there is a great need to help establish scientific methods for measuring sediment bed stability (and thus contaminant transport), and contaminant bioavailability (i.e. organic and inorganic speciation). Finally, the use of mass balance models requires highly skilled personnel, and this need is likely to continue, unless more readily useable models can be developed. Some attempts have been made to develop Excel-based models which are more accessible to less experienced individuals. Again, the expertise in these various topical areas is well represented between the previously mentioned schools and departments.

### *8.3. Treatment Technologies*

The current evaluation of effectiveness, feasibility and cost of innovative treatment technologies indicated that most were feasible, all technologies exhibited some degree of contaminant-specific effectiveness, and most cost more than traditional confined disposal. Also, technologies for the treatment of contaminated sediment in situ are less developed than those applicable to dredged material. The following research needs were identified by the NRC for selected technologies:

Natural recovery: scientific underpinnings, protocols for in situ flux measurements and to quantify relative chemical release measurements

In place capping: data analysis of current efforts, controls for chemical release, simulation of temporal disturbances

Immobilization: scientific underpinnings, enhancement technologies, long-term effectiveness, simulation of temporal and spatial effects, lab/pilot/field demonstrations, applicability of material for beneficial re-use

In situ bioremediation: bioavailability and the effect of aging, enhancement approaches, laboratory, pilot and field demonstrations, impact of sediment composition and hydrodynamics, possible combination with capping.

Chemical separation, thermal desorption and destruction: systems integration for complete physical isolation and destruction, process control, effluent gas treatment for metals

CDF/CAD/landfills: design approaches (liners, covers), protocols for loss estimation, site restoration, and potential use of treated materials, subaqueous contaminant release pathways, methodology for dewatering (CDF, landfill), rehandling and transportation.

In all categories, reliable cost estimates, including cost recovery from potential beneficial re-use, have to be developed. The UM has well-represented expertise in various aspects of innovative technology development, but more needs to be done to include cost analysis and feasibility components during technology transfer.

#### *8.4. Public Involvement/Education*

Broad public involvement and education is critical in any sediment assessment and remedy selection study in order to develop a common understanding of the problem, and the environmental and economic impacts of alternative remedial actions. Given the potentially high cost of remediating all the contaminated sediment deposits in the Great Lakes, there is a need to better define the problem and determine its impacts, so that scarce resources can be strategically targeted at those sediments posing the greatest risk to the basin, through interaction with local communities. Whereas both SeaGrant and SNRE have established successful communication channels with various public entities, public outreach by the COE requires development.

Table 1. Innovative Technologies for Beneficial Re-Use of Contaminated Dredge Material (after Krause and McDonnell, 2000)

Technology	Process	Advantages	Disadvantages	Cost
<b>Contaminant Separation</b>				
Thermal Desorption/ Cement-Lock	Heat (up to 1400 °C) to remove contaminants from the sediment matrix, which is then added to cement mix. The process takes place in a rotary kiln.	Beneficial uses include construction fill and habitat restoration.  Existing cement plants may be able to handle large volumes of dredged material  Process can remove all organic and most metals	Site waste stream that requires disposal at a hazardous waste treatment facility	Processing cost of \$ 50 per cubic yard  Value of construction-grade product \$50-70 per cubic yard.  Disposal costs of waste stream depends on level of contamination
Fluidized Bed Treatment	High temperature heating unit (not oxidation or incineration) that converts all organic materials to carbon monoxide, hydrogen and methane	Material is 99.9% free of organic material and, depending on metal content, disposed of without restriction  Operation in continuous mode, and a priori dewatering not required	Extremely energy intensive and costly  Has only been demonstrated in a small pilot scale project	Pilot and full scale production costs are estimated between \$40 and \$120 per cubic yard.
Plasma Vitrification	Plasma torch (5000°C) melts sediment using fluxes to produce a glass product	Glass product can be sold to recover some of the costs associated with process	Production of small waste stream of oversized debris and CaSO <sub>4</sub> , which can be readily landfilled	Processing cost of \$90-120 per cubic yard (depends on cost of electricity)
<b>Contaminant Destruction</b>				
Base-Catalyzed Decomposition	Two-stage process which primarily removes (volatilizes) and destroys halogenated compounds	Product 99.8% free of chlorinated compounds.  Also removes volatile and semi-volatile compounds.  Remaining metals not leachable	Polycyclic aromatic hydrocarbons are not removed	Production costs are approximately \$108 per cubic yard.

Table 1. (Cont.) Innovative Technologies for Beneficial Re-Use of Contaminated Dredge Material

Technology	Process	Advantages	Disadvantages	Cost
<b>Contaminant Reduction</b>				
Soil Washing	Process blends detergents, chelating and oxidizing agents, and high pressure water jets	90% reduction in organic compounds, and 70% reduction in metals  Process (after blending) yields a product suitable for manufactured topsoils	Only useful for low to medium contamination levels	Processing costs estimated at \$30-50 per cubic yard
<b>Contaminant Stabilization</b>				
Solidification/Stabilization	Addition of cement, fly ash, lime and/or chemicals to soil aggregates  Can be used in conjunction with other processes	Bound aggregates can be used for some types construction processes as well as landfill closure and Brownfield remediation projects  Both freshwater and marine sediments used.	Highly contaminated sediments may need to be pre-cleaned with another process prior to S/S to produce a beneficial end-product  In some cases, changes in contaminant chemistry may render them more susceptible to leaching	Production costs vary by sediment/soil type, raw materials costs, and the level and type of contamination  Costs were not inclusive of contaminant removal prior to S/S (if required).  Production costs estimated at \$30-50 per cubic yard
Manufactured Soils	Dredge material mixed with other proprietary materials to produce a top soil	Provides beneficial use for other waste products  Topsoil product can be sold to municipalities and the public	Process requires relatively small batches (< 5000 cy)	Production costs estimated at approx. \$20 per cubic yard, but does not include contamination reduction costs
Construction Products	Dredged material used as raw material to manufacture construction products such as building blocks, tiles and bricks	Both freshwater and marine sediments can be used	Full-scale production is not yet available	Industry estimates at \$20-80 per cubic yard do not include contamination removal costs

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