

Mercury Contaminated Sites: A Review of Remedial Solutions

Jennifer Hinton, jhinton50@hotmail.com

Marcello Veiga, veiga@mining.ubc.ca

The University of British Columbia, Vancouver, BC Canada

The physio-chemical characteristics of mercury have made it a favourable component for many industrial and agricultural applications. Significant anthropogenic Hg sources include fossil fuel combustion, the chloralkali industry and pharmaceuticals. Modifications to industrial practices and stricter regulations have led to emission reductions in many countries; however, this has been countered by the increased use of Hg for gold amalgamation, particularly in tropical, developing countries. Under any of these circumstances, varying quantities and forms of Hg have been inevitably released to environments where it can pose a risk to human and ecological health.

Given the unique behaviour of Hg, remediation of Hg contaminated sites can be complicated and costly. This paper reviews commonly employed and emerging techniques to mitigate Hg pollution and describes key design considerations and concerns associated with each method. Well-established *ex-situ* (external) techniques, such as physical separation and thermal treatment of excavated materials, are discussed in theory and practice. Potential *in-situ* (*i.e.* in place) Hg recovery methods, such as soil vapour extraction combined with soil heating and the use of leaching agents, are also explored. Finally, containment strategies are described for sites where more conclusive measures cannot be employed.

Keywords: mercury sources, mercury pollution, remediation, mitigation.

CHARACTERIZATION OF CONTAMINATED SITES

Industrial activities use Hg for various purposes and can therefore introduce Hg into the environment in many different forms. As the toxicological impacts of Hg are largely dependent on speciation, understanding its transformations and the impacts of various chemical forms is vital to the prevention of harmful human and environmental health effects. Ultimately, the Hg species present in a given environment depends on the initial released form, the thermodynamic stability of this compound and the transformation rate of the released form to a more stable one (Baeyens *et al*, 1979). These issues must be well understood to effectively design and evaluate appropriate remedial solutions in Hg impacted areas.

Depending on the mercury source and how it was released into the environment, mercury may be present in concentrated "hot spots" or dispersed over extensive areas, as is often the case in agricultural applications or atmospheric deposition adjacent to power generating facilities. Often, contamination is initially suspected on the basis of historical land use (*e.g.* in the production of primary batteries). Once an initial investigation of land use has been conducted, soil and

groundwater should be sampled to determine the presence of contaminants. A detailed site will determine the extent and distribution of contamination and characterize geologic units significant to contaminant mobility. This typically involves subsurface drilling and/or shallow trench sampling, generally using a backhoe, and may be accompanied by the installation of piezometers to sample and monitor groundwater and collection of surface water samples. Primarily due to the volatile nature of many forms of Hg, the health of those carrying out work at the site must be protected throughout the investigation.

The recent metallic Hg spill in Choropampa, Peru, brought to light the importance to consider the form of Hg present when conducting sampling programs. During the clean-up that followed this accident, an analytical procedure was employed to detect metallic Hg “hot spots” along the roadsides. Although the procedure was accurately conducted, the size of the sample analyzed (1 gram) was too small to sufficiently represent the content of Hg in soils from a given location.

When released in the environment, metallic Hg often produces a “nugget effect”, i.e. individual droplets increase the analyzed concentration in discrete locations. Consequently, larger samples are always needed to avoid sampling errors. It has been established that the most effective procedure for detection of metallic Hg “hot spots” is a semi-quantitative method involving pyrolysis of 30g soil samples (CETEM, 1989). In the screening stage, analytical precision of small, discrete samples is essentially irrelevant to the practical identification of hot spots. For comparison, the appropriateness of sample sizes for gold sampling is shown in Table 1. It has been established that visible identification by panning is a reasonable and inexpensive method to identify metallic Hg hotspots until levels of 3 ppm (CETEM, 1989).

Size of Gold Particle (mm)	kg of sample required	
	4 ppm Au	1 ppm Au
2.0	400	1000
1.0	50	200
0.5	8	30
0.25	1	4
0.125	0.1	0.5
0.062	0.02	0.05
0.031	0.002	0.006
0.015	0.0002	0.002
0.008	0.00002	0.0001

Table 1. - Required Sample Sizes for Characterization (adapted from Clifton et al., 1969)

Sampling of biota present at the site and/or the implementation of bioavailability studies should be performed at this point in the site assessment. Bioaccumulation evidence must be obtained or predicted in order to evaluate appropriate courses of action. If impacts to biota are not proven or probable, containment and long-term management is more appropriate than other aggressive measures. This, of course, is based on the acceptability of this to regulators. If bioaccumulation is occurring or likely to occur, then remediation must be implemented. The decision now is to excavate and treat or isolate impacted materials *or* address the contamination using *in-situ* methods. Currently, excavation is widely practiced as it is frequently the most cost-effective option for Hg impacted sites. However, alternative *in-situ* technologies will likely become more prevalent as they become more established and less costly.

In some cases, the decision to remove contaminated soils or sediments is based exclusively on Hg concentrations in excess of numerical criteria. In Japan, for example, the decision to dredge sediments from Minamata Bay with Hg concentration above 25 ppm was based on many site-specific factors such as tidal range, sediment-to-water transfer rate and a safety factor of 100 in fishing zones (Kudo and Turner, 1999). In British Columbia, Canada, after the construction of a Convention Center on a old contaminated site, the Government established guidelines for Hg concentration in soils. The BC Ministry of Environment (1989) determines that soils or sediments with Hg concentration between 2 and 10 ppm require remediation to levels below 2 ppm if the land is to be used for residential and recreation purposes. For sites with concentrations above 10 ppm Hg, all uses of land are restricted pending the application of appropriate remedial measures that reduce contaminant concentrations to less than 10 ppm.

It is clear that the simple soil or sediment analysis does not provide enough evidence to support remediation actions. Sites with 25,000 ppm of Hg in British Columbia have been analyzed. This tailing from a chlor-alkali operation must definitely be properly contained. It is evident that a risk based approach as warranted in many situations.

REMEDICATION TECHNOLOGIES

Many factors are used to assess the suitability of specific remedial measures for a given site. The distribution and properties of stratigraphic units (individual soil and rock types) and a comprehensive understanding of the site hydrogeology and hydrology, are used in conjunction with physio-chemical properties of the contaminant to predict contaminant mobility and distribution and subsequently develop means for mitigation. Once determined, these elements can contribute to a preliminary assessment of risk to ecological and human health in the impacted area and provide a basis for evaluation and subsequent design of measures for mitigation.

The determination of clean-up goals and prescribed responses are becoming increasingly dependent on identified risks to human and ecological health. Although numerical criteria are valuable indicators of the occurrence and extent of contamination, these levels are not economically or technically attainable in some situations. As shown in Figures 1 and 2, if Hg is detected in biota (*i.e.* it is bioavailable) or the risk of its subsequent incorporation into organisms is appreciable, then more aggressive and often more costly measures are warranted. In the case of dispersed mercury contamination, remedial measures are typically not feasible (Figure 1). If bioaccumulation is identified, then exposure pathways must be addressed. Possible responses

include the implementation of consumption advisories to reduce intake of contaminated food, or educational programs and technology modifications to lessen exposure to Hg vapour. If Hg is distributed in a localized area (Figure 2) and bioaccumulation is evident, then remedial actions are warranted. If the potential for assimilation into the food chain is low, then containment methods are sufficient. In either situation, long term management is required until adequate protection of ecological and human health is ensured. Of course, any measure employed must be acceptable to regulators.

Excavation and Treatment

Excavation and *ex-situ* (*i.e.* off-site or aboveground) treatment of Hg-contaminated soils is the most frequently employed practice for Hg recovery. Although excavation can be complicated if it extends below the water table or costly if the contamination is distributed over a large area, it is essentially a well-understood practice. Mercury is subsequently liberated from excavated soil by one of the three processes described below.

Figure 1: Appropriate Response to Dispersed Mercury

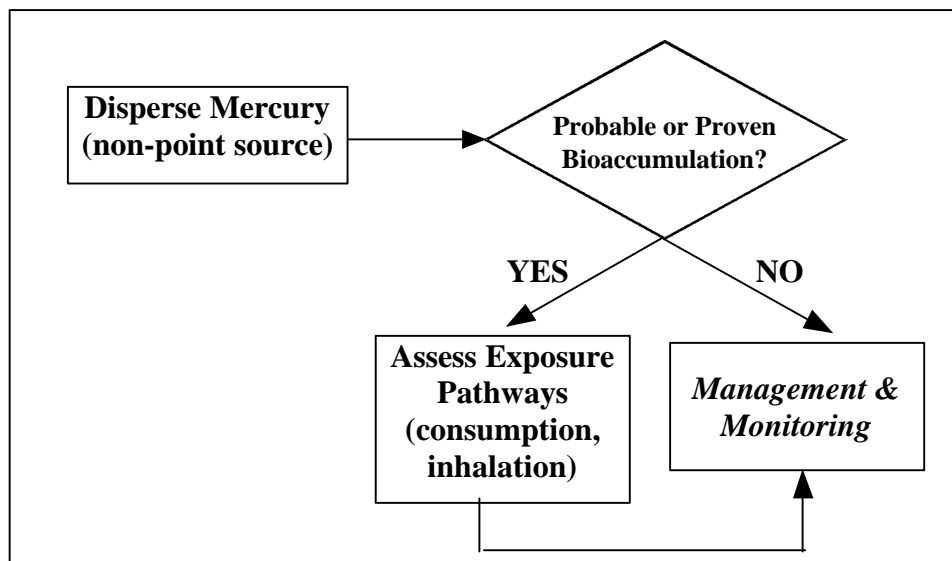
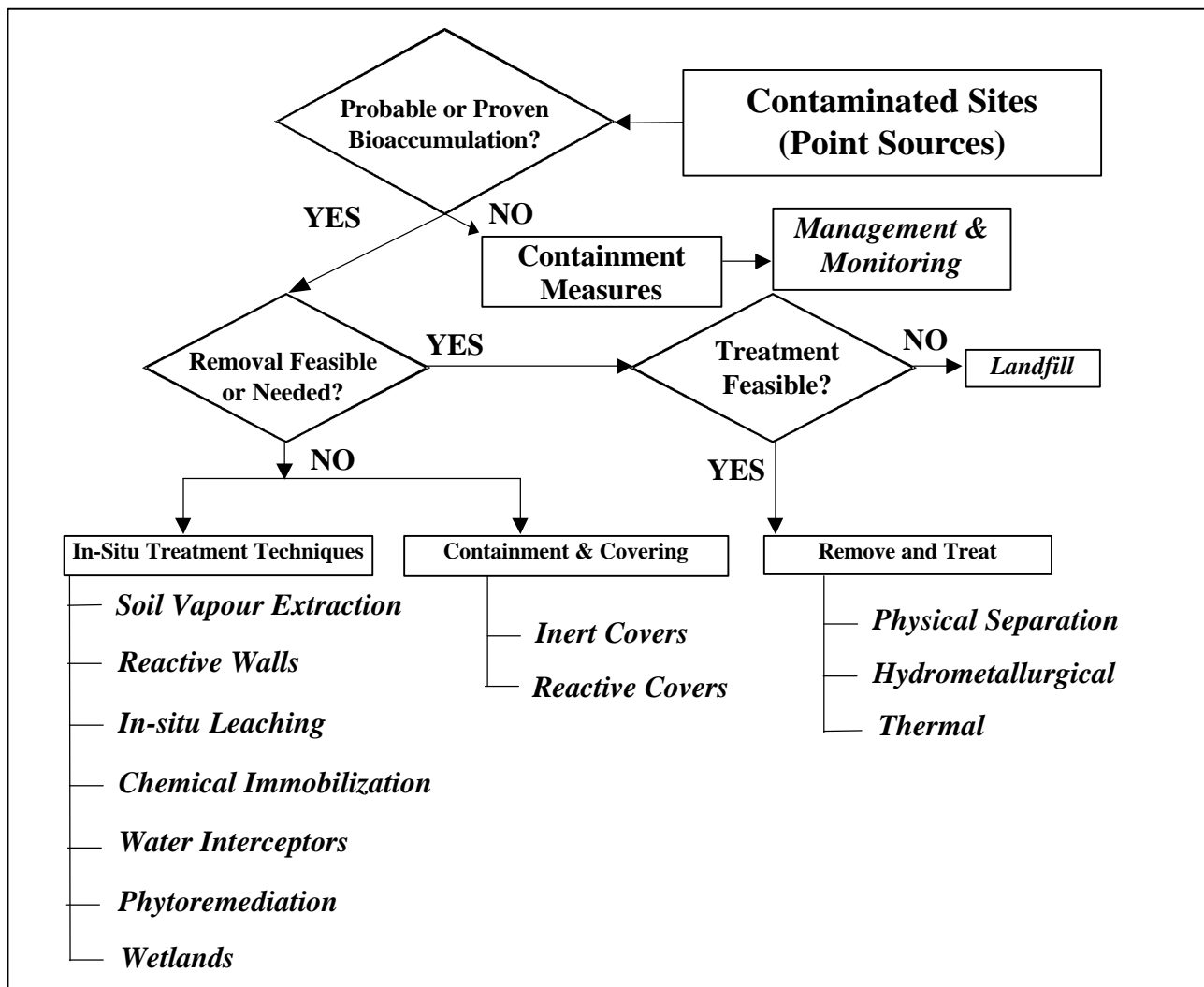


Figure 2 - Appropriate Responses to Point Source Mercury Contamination



Physical Separation

Classification on the basis of size fraction simple process is based on the premise that most Hg compounds have a strong affinity for the fine fraction of soils. In wet screening, metallic Hg droplets behave as a separate liquid phase being concentrated into the fines. If there is an intention to analyze Hg grain size distribution, dry sieving must be adopted. Rubble and coarse constituents of soils, such as gravel, are initially removed via sieving. Water is used to wash small to medium fractions (~50mm to >0.1mm diameter) and fines (*i.e.* silts, clays or organics) are separated from these fractions, generally using hydrocyclones, spiral classifiers, and fluidized beds (Hempel and Thoeming, 1999). The remaining Hg-enriched sludge or fine fraction is dewatered and subsequently isolated or treated further (for example, via thermal methods). This physical separation procedure well established, effective at reducing the volume of contaminated soils and generally does not require the use of other chemicals. This technique is most effective for soils dominated by coarse materials (*i.e.* sand and gravel) with some (< 20%) fines. Due to the toxicity of Hg, the occupational health of adjacent workers naturally must be considered during processing.

Thermal Treatment

As the volatility of Hg and its compounds increase with temperature, thermal heating of excavated soil is a potentially effective means for Hg recovery from contaminated soils. Hempel and Thoeming (1999) determined that all Hg compounds should volatilize at temperatures below 600 °C and, in the absence of HgO, 250 °C should be sufficient. Larry and Jose (1990) recommended heating temperatures between 600-900 °C. Matsuyama *et al* (1999) determined that low-temperature thermal treatments could adequately recover Hg from soils, were less costly and less likely to negatively alter soil properties. Matsuyama *et al* found that at temperatures of 200-210°C, even stable compounds like HgS begin to volatilize. In the presence of a catalyst, specifically iron chloride, Hg (as HgS) removal increased from 50% to 99.5% in a one-hour period at 300 °C.

Organic contaminated soils are commonly treated using thermal processes. Hempel and Thoeming (1999) described the method for organic pollutants, as it is principally the same for Hg recovery from soils. Soil is processed in batches as follows: first it is dried at a temperature of 100 °C., then transferred to a heating drum where temperatures of 600°C are maintained. Gas from these stages passes through an afterburner where, at temperatures near 800-900°C, conversion of organic contaminants to CO₂ is anticipated. This is followed by gas cooling (150°C), flow through a dust filter and a spray tower to remove dust and SO₂. If this typical configuration were applied to Hg, it would be recovered from the gas phase using a gas washing system (Hempel and Thoeming, 1999), charcoal filter (Renner, 1995), iodine impregnated scrubber or through condensation.

Hydrometallurgical Treatments

Chemical extraction of Hg from excavated soils can be induced through four primary mechanisms: desorption of adsorbed species; oxidation of metallic Hg; use of strong complexing agents; and through dissolution of precipitated Hg (*after* Hempel and Thoeming, 1999). Efficiency of any mechanism employed may decrease over time due to recomplexation and readsorption and removal of the most soluble compounds at early time. The two most promising hydrometallurgical techniques employed are electrokinetic or electroleaching and leaching methods.

Leaching agents commonly applied to excavated materials include halide compounds, such as hypochlorite or hydrobromic acid, iodine in the form of potassium iodine, and a mixture of nitric acid and NaCl (Hempel and Thoeming, 1999). Sodium hypochlorite and sodium pyrophosphate are believed to be particularly effective at liberating metals from organics, but hydrogen peroxide releases more metals from oxides and sulfides (Papp *et al.*, 1991).

In-Situ Recovery

Methods for *in-situ* recovery of Hg are far less established than *ex-situ* techniques. As well, due to subsurface heterogeneity, more uncertainty generally exists concerning the effectiveness of *in-situ* processes, and clean-up times tend to be longer than *ex-situ* treatments. Despite these factors, many *in-situ* technologies are very promising and – mainly due to the fact that contaminated soil and groundwater remain in the subsurface – may become more cost-effective and practical than excavation and treatment methods for many Hg-contaminated sites.

Soil Vapour Extraction coupled with Soil Heating

Soil Vapour Extraction (SVE) uses a vacuum to force air through the unsaturated zone. Volatile and semi-volatile compounds easily partition into the vapour phase and are subsequently removed by the vacuum. The ground surface is covered with a tarp or other cover system to minimize vacuum extraction of “clean” air from the surface and ensure lateral airflow through the impacted area. The effectiveness of these systems is primarily dictated by contaminant volatility and availability to air channels, which is mainly governed by contaminant solubility and tendency to sorb to solid surfaces and soil properties (*e.g.* grain size).

It is well demonstrated that Hg fluxes into the vapour phase increase significantly with airflow and temperature (Ebinghaus *et al.*, 1999, Sexauer Gustin *et al.*, 1997). Thus, soil vapour extraction may be an effective technology for *in-situ* remediation of Hg if performed in conjunction with soil heating. Currently, soil heating can be costly over expansive areas and difficult to homogeneously heat a soil volume. As well, the effects of soil heating on physical, chemical and biological properties of soil are not well known. Despite this, soil heating combined with soil vapour extraction may in the future become an effective means of Hg removal in the vadose zone.

Permeable Reactive Walls

Permeable Reactive Walls, wherein dissolved compounds react with wall constituents to precipitate contaminants, have successfully been employed at many organic and metal impacted sites. Permeable reactive walls are engineered structures installed below the ground surface perpendicular to the flow of contaminant-laden groundwater. Walls are geochemically engineered to transform contaminants to relatively benign and/or immobile forms and ideally can operate passively for extended periods with little or no maintenance (Domenico and Schwartz, 1998). Two main criteria that must be addressed in effective design include wall permeability (ideally slightly greater than the parent material) and the level of reactivity with wall constituents (Waybrandt *et al.*, 1998). Depending upon the contaminant of concern, wall constituents have ranged from organic-rich composted sewage sludge or wood chips combined with neutralizing materials (*e.g.* limestone) for removal of metals from acidic mine drainage to zero-valent iron (ZVI) for various organic and inorganic contaminants. Other materials proposed for metals studied include hydroxyapatite (Xu and Schwartz, 1994), zeolites (Li, in Press), hydrous ferric oxides and bone char phosphate (USEPA, 1997).

In-situ Leaching and Extraction

Used in conjunction with Pump-and-Treat systems, *In-situ* Leaching and Extraction involves the injection of chemicals to enhance Hg solubility in groundwater, thereby reducing clean-up time and improving recovery rates from groundwater. Pump-and-treat is a frequently practiced, cost-effective remedial alternative employed either for removal of contaminants from the subsurface and/or hydraulic containment of a contaminant plume (Domenico and Schwartz, 1998). This technology is generally limited to treatment of contaminants impacting groundwater in a dissolved form (*e.g.* HgCl⁻, HgS⁰) or as a non-aqueous phase liquid (NAPLs, *e.g.* metallic Hg). In many natural systems, Hg is strongly adsorbed to soils and/or is present as a precipitate – in either situation, Hg may dissolve or desorb over time and thereby provide a long-term source to groundwater. To reduce clean-up time and increase the efficacy of the clean-up system, solubility-enhancing chemicals may be injected up-gradient from the zone of contamination. Two issues limit the applicability of this method of *in-situ* Hg extraction. First, it is not well demonstrated and second, the injection of leaching agents into the subsurface for the purpose of enhancing contaminant mobility is often unacceptable to regulators.

Electro-Kinetic Separation

This process involves the generation of an electric field through application of a low-voltage direct current (DC) in a soil matrix (USAEC, 2000). Heavy metals, such as Hg, migrate towards electrodes placed in the soil where they accumulate and can subsequently be removed at a lower cost than excavating the entire impacted area. In fine-grained soils (*e.g.* clays), migration of charged and uncharged species may occur by *electroosmosis* (Hempel and Thoeming, 1999). The electrokinetic separation process occurs in three stages (Sobolev *et al*, 1996). First, the metal must be transformed into a soluble form, with or without the injection of solutions. Second, the electric current mobilizes the solubilized metal towards an electrode. Finally, accumulated metals are collected at the electrode, typically through excavation.

Interceptor Systems

Interceptor Systems, such as trenches and drains, are extremely simple and effective at recovering Hg as “free product” (essentially as metallic Hg), however this treatment is limited by topography and stratigraphy and does not address Hg held in residual saturation.

Phytoremediation

Phytoremediation is a promising albeit unproven technology, wherein plants assimilate and concentrate metals from soils. Metal recovery typically occurs through subsequent harvesting and combustion of plants (Anderson *et al*, 1999). Plants that accumulate substantial quantities of metals from soils, known as *hyperaccumulators*, may be species specially developed for extraction of certain metals, or native species possessing a resistance to toxic effects of specific metals. This technique holds much promise for the cost effective remediation of shallow soils over a fairly widespread area, but issues such as limited access to vegetation by wildlife and time required for clean-up must be addressed.

Passive Remediation - Wetlands

The use of wetlands for Hg immobilization is a somewhat controversial issue as wetland-type environments are intrinsically amenable to the conversion of Hg to its highly toxic form, methylmercury (MeHg). Despite this potential, the US Department of Energy (Anon, 2000) has had notable success in significantly limiting the amount of MeHg generated (<1 ng/L) in an artificial wetland, mainly by controlling the amount and form of sulfur present. Mercury bound to decomposed organic matter descends to the bottom of anaerobic cells where it reacts with gypsum to form relatively insoluble sulfide. Ultimately, the wetland will treat up to 1 million gallons of water daily.

Containment

Clean-up of many contaminated sites is often not feasible, generally due to financial or technical reasons. Several methods of containment are commonly employed to inhibit mobilization of contaminants and minimize ecological and human health risks of exposure.

Pump-and-Treat

As discussed in the Section on *In-Situ* Leaching and Extraction, pump-and-treat is a frequently employed, cost-effective alternative. Due to the characteristics of the contaminant or system, contaminant removal may not be possible and hydraulic containment may be necessary to protect the surrounding environment. As the mass of contamination remains in the subsurface, pump-and-treat systems must operate in perpetuity to prevent off-site migration. Basically, pump-and-treat systems involve the installation of extraction wells below the water table within or slightly down-gradient from the zone of contamination. As extracted water must be treated at the surface, well placement and pumping rate should be chosen to ensure capture of contaminated groundwater and limit recovery of clean water. Monitoring wells are installed around the contaminant plume to assess containment and evaluate hydrogeochemical conditions.

Impermeable Barriers, Surface Seals and Drains

There are several geotechnically-engineered approaches for prevention of the off-site migration of contaminants. Each system has limitations with respect to emplacement depth and uncertainty concerning permeability and barriers may intend to surround the contaminated zone entirely or remove the potential for groundwater flow through the source. *Slurry walls* are low permeability barriers generally composed of excavated material mixed with bentonite or a mixture of cement and bentonite. Walls can be constructed as a trench, which are typically 0.5 to 2m wide and installed to a maximum depth of 50m, or using specialized equipment (Domenico and Schwartz, 1998). Notable installation systems include augers that mix bentonite with native soil (limited to 60m depths) and a procedure wherein parallel steel plates are forced into the ground using vibration and filled with bentonite slurry as the steel is retracted. The latter method is more susceptible to leakage, as it must be constructed in sections and is considerably thinner

than the other methods. *Grout walls* or *grout curtains* are similar to slurry walls constructed through the pressurized injection of gelling or solidifying liquids into the subsurface (Domenico and Schwartz, 1998). *Sheet pile walls* are installed using driving or vibratory forces. In the past, joints between adjacent piles have tended to leak until voids are filled with fines. Recently, however, a technique proposed by the University of Waterloo in Canada in which joints are sealed during construction has been demonstrated in field tests to be extremely effective (Domenico and Schwartz, 1998).

Surface seals and drains are used to control infiltration and limit mobilization of contaminants into groundwater (Domenico and Schwartz, 1999). Low permeability materials, such as compacted clays, natural soils mixed with stabilizers or bentonite, or geosynthetic membranes, are frequently used to inhibit infiltration. Design considerations include grading of surfaces to direct drainage and the long-term integrity of the seals. Elements such as weather effects (frost, desiccation of clays), damage from burrowing animals or vegetation or degradation of synthetics by sunlight can all be significant.

Stabilization and Solidification

Solidification and stabilization techniques involve the *ex-situ* or *in-situ* mixing of impacted soils with additives that intend to reduce the mobility or leachability of contaminants (Domenico and Schwartz, 1998). Stabilization specifically attempts to bind contaminants to the solid and is often accompanied by reductions in soil permeabilities. Solidification techniques will improve the physical characteristics of materials, such as sediments or sludges, so they can be excavated and transported more easily.

Many suppliers propose various concoctions of stabilizing agents (*e.g.* organic polymers), although cement, calcium carbonate spiked with binders (furnace slag silicates or fly ash), asphalt or bitumen are most commonly used. Augers or adapted digging tools can mix additives with soil that has been excavated or directly in the subsurface. Subsurface mixing is less established than aboveground techniques, primarily because specialized injection and mixing equipment must be used, homogeneous treatment is difficult to ensure, and it is still a fairly costly procedure. Despite these issues, *in-situ* stabilization may in the future become an effective solution for deep and difficult to access zones of contamination.

Sediment Capping

In-situ capping involves the placement of a subaqueous cap of clean and ideally isolating material over contaminated sediments (Palermo, 1998). As dredging is often difficult to achieve without re-suspension of sediments and thus contaminants, removal of contaminated material is not always practical. The U.S. Army Corp of Engineers has developed detailed specifications for design, construction and monitoring of these systems (Palermo, 1998).

Containment via sediment capping should satisfy three basic purposes: physical isolation of the contaminated sediments from benthic organisms, prevention of sediment re-suspension and transport, and reduction of the contaminant migration into the water column (Palermo, 1998). In the case of mercury in sediments, the increased solubility and diffusability of methylmercury must be considered during evaluation of this alternative.

Site specific issues that must be assessed prior to cap design include the following: qualities of the watercourse (bathymetry, currents, wave energies and seasonal variability, etc.); functions of the waterway (water supply, wastewater discharge, recreational use, etc.); and geoenvironmental properties (sediment, soil and rock stratigraphy and individual attributes, hydrogeologic conditions, etc) (*after* Palermo, 1998). Sufficient understanding of groundwater flow is particularly important as it will influence the flux of contaminants into the water body and may modify or be modified by the capping system.

CONCLUSIONS

Due to historic and recent industrial practices, varying quantities and forms of Hg have been released to environments where it can pose a risk to human and ecological health. In response to this, several techniques exist or are currently being developed for the remediation of mercury and other contaminants. Techniques such as excavation and *ex-situ* treatment of impacted soils and containment measures are frequently practiced and well understood. Despite this, these methods are often not cost effective or technically feasible. Emerging *in-situ* procedures, such as permeable reactive walls and leaching, may ultimately become more appropriate for many Hg-impacted sites. In some situations, acceptable clean-up goals are simply not economically or technically possible, thus long term management through containment and monitoring must be employed. Ultimately, any measure employed must consider the risk to ecological or human health and have the acceptance of regulators.

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