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Review of Phytoreclamation and Management Approaches for Dredged Material Contaminated with Lead

PURPOSE: The purpose of this technical note is to provide information for managing dredged material contaminated with lead (Pb).

BACKGROUND: Dredged material can contain levels of contaminants that are of concern, especially to State regulatory agencies. Before the dredged material can be used for beneficial purposes, the level of contaminants or the risk of contaminants to human health or the environment may need to be reduced. Such situations have recently evolved in Milwaukee, WI, and in the Bronx, NY, at Van Cortlandt Lake Park. Results of recycled soil manufacturing technology (RSMT) screening tests indicated that a high-quality topsoil could be manufactured from Van Cortlandt Lake dredged material (Lee, Sturgis, et al. in preparation). The dredged material initially appeared to contain a moderate level of Pb, approximately 500 mg/kg. The final concentration of Pb in the dredged material blended topsoil was approximately 150 mg/kg. When the New York City Department of Parks and Recreation indicated a desire to manufacture topsoil using RSMT for athletic field reconstruction, the city and State regulatory agencies raised concern about the Pb in the dredged material. The regulators identified 33 mg/kg Pb as a safe soil concentration with regard to wildlife. However, the existing soil concentrations of Pb in the New York City area average approximately 55 mg/kg. Pb is a naturally occurring metal, and has always been present in soils, surface waters, and groundwaters. Concentrations in soils range from 2 to 200 mg/kg and overall average 16 mg/kg (Stubbs 1973). Lead content of agricultural soils ranges from >1 to 135 mg/kg with a median value of 11 mg/kg (Holmgren et al. 1993). In Milwaukee, WI, the desired concentration of Pb in dredged material for beneficial uses is 50 mg/kg according to State regulators.

INTRODUCTION: Lead contamination has been the focus of much research recently to determine how best to manage the Pb and render it harmless and reduce risks to human health and the environment. There is a vast amount of research studying Pb contamination in urban soils, agricultural soils, and Pb mine lands. Other recent research is evaluating Pb contamination in firing ranges on Department of Defense Military Training Installations (IT 1999, 2000a, 2000b; Woodward-Clyde 1987; Geofon, Inc., 2000; and Montgomery Watson 2000a, 2000b).

This technical note (TN) discusses available information and relates the more important research findings to Pb contamination in dredged material and potential management strategies that are feasible. This TN will refer to a comprehensive literature review of phytoremediation technology for the phytoextraction of Pb from contaminated soils as well as recent innovative research on the immobilization of soil Pb through the formation of pyrophosphate minerals (Lee, Price, et al. in preparation). This TN is a follow-up of TN-DOER-C9, which described the findings of a working group of experts on phytoreclamation of contaminants in dredged material (Price, Lee, and Simmers 1999).

Phytoreclamation/Phytoextraction of Lead. Soils contaminated with heavy metals are commonplace and may present a threat to the environment and human health. Lead contamination of soils has been associated with many ecological problems including toxicity to green plants, animals, and humans; contamination of groundwater; and loss of vegetative cover (Body, Dolan, and Mulcahy 1991). Lead contamination of soil and sediment, as well as surface and groundwaters, has occurred to significant degrees due to industrial processes, smelting and combustion of coal (Sawidis et al. 2001; Nriagu 1973), mining activities (Cotter-Howells and Thornton 1991; Blowes et al. 1991), use of Pb-based paints (Weitzman et al. 1993), automotive exhaust fumes, land applications of municipal wastewater and sludge (Logan and Chaney 1983; Nriagu 1973), agricultural uses of fertilizers and insecticides (Ma, Logan, and Traina 1995), as well as waste disposal in landfills (Gounaris, Anderson, and Holsen 1993; Nriagu 1973).

Natural mineral deposits containing particularly large quantities of heavy metals are present in many regions of the globe. These areas often support characteristic plant species that thrive in these metal-enriched environments. Some of these species can accumulate unusually high concentrations of toxic metals to levels that far exceed the soil levels. Accumulators of nickel (Ni), cobalt (Co), copper (Cu), manganese (Mn), Pb, zinc (Zn), and selenium (Se) have been reported (McGrath et al. 1993). As a result of their association with specific ore deposits, many metallophyte plants are used in prospecting for mineral deposits. Metal hyperaccumulating plants are plants that can accumulate unusual amounts of specific metals (Raskin et al. 1994). Baker and Brooks (1989), Baker et al. (2000), and Reeves et al. (2000) summarized research on hyperaccumulator plants. They defined a hyperaccumulator of Co, Cu, chromium (Cr), Pb, or Ni to be a plant that can have metal concentrations of 1,000 mg/kg or more in plant tissue. Hyperaccumulating plants are widespread throughout the plant kingdom. It is common for plants within a genus to widely differ in their ability to hyperaccumulate metals. Hyperaccumulators tend to grow in very specific soils (with high metal content). The *Thlaspi* genus (including species *caerulescens* and *rotundifolium*) was particularly effective at hyperaccumulating Pb (as well as cadmium (Cd) and Zn).

Although hyperaccumulators were promising, they have several disadvantages. First, they tend to be small, lacking the biomass to extract large amounts of metal. They also have shallow root systems, and many live in only narrow ecological ranges. Therefore, researchers developed more aggressive approaches. The use of chelating agents, such as EthylenediamineTetraacetate (EDTA), was developed. The approach also uses larger plants, such as Indian mustard (*Brassica juncea*). These plants were grown to near maturity. Then the EDTA solutions were applied to the soil to increase the solubility of the metal. The plants would then uptake and sequester the metal.

In an effort to improve the phytoextraction of Pb and other metals, researchers have worked extensively on the use of chelating agents. Chelating agents have been used to extract metals from soils (Norvell 1984) and have been widely studied and used in the field of agriculture and plant nutrition (Albasel and Cottenie 1985). The use of chelates to improve phytoextraction was introduced in 1997 by Huang et al. (1997) and Blaylock et al. (1997).

Huang et al. (1997) presented evidence that phytoextraction of metals requires high-biomass plants that can accumulate >1 percent Pb in shoots. It was determined that most hyperaccumulators, like *Thlaspi rotundifolium*, were too small and grow too slowly for practical phytoremediation. Most hyperaccumulator plants also had shallow root systems and many lived in only narrow ecological ranges. Therefore, Huang et al. (1997) studied the uptake of Pb using corn (*Zea mays*) and pea with the assistance of chelating agents. This study indicated the following effectiveness of various chelates: EDTA>HEDTA>DTPA>EGTA>EDDHA. The addition of EDTA increased Pb shoot concentrations in both species from <500 to >10,000 mg/kg. Furthermore, EDTA improved translocation of metals from roots to shoots.

Blaylock et al. (1997) studied the effects of EDTA and other chelates on improving uptake of Pb and other metals (Cd, Cu, Ni, and Zn) by Indian mustard. A combination of EDTA and acetic acid gave the best results. A pH range of 5 to 7.5 was tested. A pH of 5 performed the best, with or without EDTA. EDTA also improved translocation of the Pb to the shoot.

Blaylock (2000) presented results from two field studies: one at Bayonne, NJ, and the other at Dorchester, MA. Both sites were approximately 93 m^2 (1,000 ft^2) and were contaminated with Pb and other metals. Within a year, the sites were treated with three harvests using Indian mustard and EDTA amendments. In both cases, most treatment appeared to be in the upper 15 cm of the soil. Contour maps of metal concentrations in the upper 15 cm seem to indicate substantial removal. Treatment goals were not reached, however, the decrease in the upper 15 cm at the Bayonne site was, in fact, significantly larger than the authors had estimated based on bench top pilot testing.

A study was conducted at the Twin Cities Army Ammunition Plant in Minnesota that illustrated the difficulty with the use of chelates for phytoextraction of Pb (U.S. Army Environmental Center (USAEC) 2001). The first year of testing indicated that the water balance was too high due to too much rain and overwatering, and the growing season was shorter than expected. Consequently, plants were stressed by insufficient nutrients, and uptake of Pb was much less than anticipated (USAEC 2001). During the third year, the project was terminated when Pb was found contaminating the groundwater under the site and the U.S. Army was issued a notice of violation from Minnesota Pollution Control Agency (Twin Cities Army Ammunition Plant 2001).

Use of chelates to solubilize Pb in dredged material for phytoextraction requires consideration of appropriate controls to minimize potential leaching and migration offsite in surface runoff. Contaminant pathway testing as described in the Upland Testing Manual (UTM) (U.S. Army Corps of Engineers 2003) should be considered before the application of chelates to solubilize Pb in dredged material. Lining and collection of leachate may be required to eliminate potential groundwater contamination. Careful application of chelate will be required to maximize phytoextraction and minimize offsite migration of chelated Pb. Plants that take up large amounts of Pb will need to be harvested and disposed of in an appropriate manner. During phytoextraction of Pb, wildlife should be excluded from the area being treated. Because of these issues, phytoextraction of Pb from dredged material is not recommended at this time.

Phytostabilization/In Situ Immobilization of Soil Lead. The total soil content of Pb relates how much Pb is present in the soil, but does not indicate the impact of the Pb on the environment. Critical factors that influence the impact of Pb on the environment are the form and bioavailability of the Pb present in the soil. Extensive research has been and continues to be conducted on determining and predicting soil Pb bioavailability (Chaney, Mielke, and Sterrett 1989). Management of Pb in soil is currently a hot topic of research. Typical management scenarios have been to excavate Pb-contaminated soil, stabilize the Pb, and haul it to an appropriate landfill at an enormous cost. Under certain conditions, it is impossible to excavate and haul off large quantities of Pb-contaminated soil, such as in Pb mining impacted areas or areas of extensive training or firing ranges.

Young children can be exposed to Pb-contaminated soil and dust in their homes and during outdoor activities. There is heightened concern about the long-term effects of Pb exposure to children (Cotter-Howells and Thornton 1991; Weitzman et al. 1993). Reducing the bioavailability of Pb from contaminated soils has been the focus of a considerable amount of research (Logan and Chaney 1983; Cotter-Howells 1996; Cotter-Howells and Caporn 1996; Ma et al. 1993, 1994a, 1994b; Ma, Logan, and Traina 1995; Ma 1996; Ryan and Berti 2000; Ryan and Zhang 2000; Ruby, Davis, and Nicholson 1994; Ruby et al. 1996). A method that has proven to be very effective involves that of phosphate mineralization. Phosphates were found to rapidly immobilize and form stable insoluble compounds with Pb in soils, sediments, or water. Current research has focused on in situ stabilization and immobilization of Pb to render the Pb harmless and environmentally safe. The use of phosphate as an agent for immobilizing Pb by the formation of extremely insoluble Pb pyrophosphate minerals has received much attention (Cotter-Howells 1996; Cotter-Howells and Caporn 1996; Ma et al. 1993, 1994a, 1994b; Ma, Logan, and Traina 1995; Ryan and Berti 2000; Ryan and Zhang 2000; Ruby, Davis, and Nicholson 1994; Ruby et al. 1996). Phosphates have been used to treat and immobilize soluble Pb species in industrial effluents or solid waste by-products (Lewickw 1972; C.K.L. 1972), aqueous environments (Xu and Schwartz 1994; Zhang and Ryan 1998; Ma, Logan, and Traina 1995), and soils (Berti and Cunningham 1997; Zhang, Ryan, and Bryndzia 1997; Ma 1996).

Research to date indicates that immobilization of Pb with phosphate is a low-cost, long-term, environmentally safe method of remediating large areas of Pb contamination.

Ryan and Berti (2000) discussed the need for in-place inactivation and natural ecological restoration technologies due to the shortcomings of many current remediation methods. Many existing remediation methods are expensive. Alternatives are needed that are low cost, low input, and environmentally benign, but must provide protection to human health and the environment equivalent to or better than current soil remediation technologies.

In response to the need for in-place remediation, U.S. Environmental Protection Agency (USEPA) in collaboration with Ohio State University examined the feasibility of Pb immobilization by rock phosphate (Ryan and Zhang 2000). This approach was based on the hypothesis that Pb phosphates are the most insoluble Pb minerals, are resistant to acid weathering, and are less bioavailable than other Pb forms. This proposition is so potentially beneficial to society, both to health and economics, that a cooperative association of government, university, and industry researchers known as the In-Place Inactivation and Ecological Remediation Technology (IINERT) Soil-Metals Action Team has been formed under the Remediation Technology Development Forum (RTDF) to verify and improve on this “in-place inactivation” technology for soil Pb and eventually to educate regulators, problem owners, and technology applicators in its uses, benefits, and limitations.

Ryan and Berti (2000) explained that in-place inactivation is a site stabilization technique in which materials are applied and mixed into the soil to alter the soil contaminant chemistry, resulting in contaminant forms that are less soluble, less mobile, and less bioavailable. While in-place inactivation does not affect or reduce the total contaminant concentration, in-place inactivation reduces the risk of harm to an organism (e.g., humans) by making the contaminant unable to biologically interact with the organism. This technique is based on fundamental soil chemistry, plant biology, agricultural practices, and experience with the restoration of drastically disturbed mines, roadside lands, and construction sites. It appears to be a relatively simple, low-cost, low-input method, and should prove adaptable to a wide range of contaminated sites.

The three most basic and important hypotheses of the study by Ryan and Berti (2000) were that (1) surrogate relationships could be identified/confirmed among Pb availability to humans, pigs, rats, and glassware extractions (single and sequential); (2) good correlations existed between soil components (e.g., Pb species and non-Pb-containing components) and the soil Pb hazard; and (3) addition of materials to Pb-contaminated soils could induce the formation of less hazardous Pb forms, providing a practical approach to in-place inactivation.

The experimental approaches used to study Pb inactivation by phosphate have included laboratory scale solution studies, resin studies, dialysis studies, soil studies, and feeding studies. Chen et al. (1997) studied the effects of heavy metals sorption on mineral apatite. They found that Pb sorption and removal from solution was not affected by the presence of other metals or pH, with nearly 100 percent being removed.

Ryan and Zhang (2000) showed that hydroxyapatite removed Pb^{2+} from Pb-EDTA solution in the presence of excess EDTA. These results indicated that basic calcium-phosphates can sequester Pb even in the presence of strongly complexing organic ligands such as EDTA. In longer term experiments, Ryan and Zhang (2000) showed that hydroxyapatite was effective in immobilizing Pb^{2+} for up to 16 weeks, confirming the stability of the reaction product.

Ma, Logan, and Traina (1995) found that the effectiveness of phosphate rocks to immobilize Pb was increased significantly by increasing the amount of phosphate rocks added at the same initial Pb concentration. While increasing the reaction time from 2 to 6 hr showed only a small increase in Pb removal from aqueous solutions, increasing the reaction time to 2 days significantly increased the removal.

Significant theoretical and experimental evidence supports the hypothesis that Pb phosphates are the most insoluble and stable forms of Pb in soils and that they can form rapidly in the presence of adequate Pb and phosphate. Among all of the Pb phosphate minerals, chloropyromorphite has the lowest solubility, and thus it is most stable under favorable environmental conditions (Nriagu 1973, 1974, 1984). Nriagu (1973) found that Pb chloropyromorphite was the most stable Pb species formed in test systems and will be the Pb species formed in most natural aquatic systems since the concentration of chloride ions present in natural systems would allow for the formation of thermodynamically stable Pb chloropyromorphite.

Physiological-Biological Extraction Test (PBET). Considerable research has been conducted to develop a laboratory extraction test that simulates the human digestion process (Ruby et al. 1992; Davis, Ruby, and Bergstrom 1992; Ruby et al. 1993; Ruby, Davis, and Nicholson 1994; Medlin 1995; Ruby et al. 1996; Ruby, Schoof, and Brattin 1999). The tests developed essentially simulate a 1-hr stomach digestion (tests have varied from 30 min to 2 hr) at pH 1.3 to pH 2.5 using a stomach juice such as glycine buffered with hydrochloric acid followed by an intestine digestion

(1 to 4 hr) at a pH of ~7.0. Extraction test results have been correlated to animal bioavailability of the soil Pb (Ruby et al. 1993; Ruby et al. 1996) and to sequential soil extraction procedures (Basta and Gradwohl 2000). Ruby, Schoof, and Brattin (1999) have related the extraction test results to human risk assessment. Refinement of the PBET procedure is being conducted under the RTDF IINERT research program with direction from the Solubility/Bioavailability Research Consortium (SBRC). Currently, the best correlation of soil Pb bioavailability to animals and humans has been obtained with a 1-hr stomach digestion at pH 2.3 using 0.4M glycine in HCl. The recommended procedure does not call for the intestine digestion at pH 7.0 since this portion of the original procedure did not correlate as well with soil Pb bioavailability.[1]

Relation of Pb Pyrophosphate Treatment to Animal Feeding Tests. Animal models have been used as human surrogates to determine the contaminant bioavailability via the incidental ingestion pathway of contaminant migration from soil to humans. Immature swine, rat, and rabbit models have been evaluated for simulating ingested heavy-metal bioavailability to humans (Casteel et al. 1996; Dieter et al. 1993; Ruby et al. 1993).

In animal feeding experiments, Ryan and Zhang (2000) have illustrated that Pb bioavailability followed the order: Pb-acetate >> contaminated > soil pyromorphite = control and that the addition of apatite or rock phosphate to the contaminated soil reduced the bioavailability of the contaminated soil Pb. These results illustrated that the formation of pyromorphite in soils not only reduced the solubility of the soil Pb, but reduced its bioavailability. In fact, even without allowing time for reaction, the addition of the phosphate (apatite or rock) to the contaminated soil was effective at reducing soil Pb bioavailability as the Pb-contaminated soil and apatite or rock phosphate passed through the test animals. Zhang and Ryan (1998) indicated the primary concern of immobilization of soil Pb was the reduction in bioavailability of soil Pb that may be ingested by a small child. However, the results of the pH-dynamic study they performed illustrated that the transformation of biologically reactive Pb such as anglesite into chloropyromorphite can be completed within the time scale of the stomach emptying into the gastrointestinal (GI) tract. The significance of this is that even if the transformation of labile soil Pb forms such as anglesite to chloropyromorphite is not completed prior to ingestion of the soil, the transformation is potentially completed in the GI tract and the bioavailability of soil Pb will be controlled by chloropyromorphite if there are stoichiometrically adequate amounts of phosphate in the digestive system. Therefore, incorporating calcium phosphate materials into Pb-contaminated soil provides the stoichiometric amounts of phosphate to control soil Pb bioavailability if a child ingests the soil mixture.

The results of this research clearly indicate that application of phosphate amendments to dredged material should also serve to immobilize any Pb contained in the dredged material and substantially reduce the risk to human health and the environment. It would appear that a safe topsoil should be able to be blended with dredged material that is contaminated with Pb with stoichiometric amounts of phosphate. Consequently, soccer fields could be safely constructed with such a blended topsoil.

Lee, Sturgis, et al. (in preparation) attempted to test this hypothesis by blending two forms of phosphate materials with topsoil manufactured from Van Cortlandt Lake dredged material, city

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[1] R. L. Chaney, Personal Communication, 14 August 2001, U.S. Department of Agriculture Agricultural Research Service, Beltsville, MD.

wood chip yard waste, and Bronx zoo manure according to recycled soil manufacturing technology. Grass was grown in the blended topsoil and earthworms were exposed to the topsoil. Soil samples were tested according to the PBET procedures and earthworms were analyzed after 28 days exposure. Less than 1 percent of the 150 mg/kg Pb in the topsoil was bioavailable in the PBET test or in the earthworms.

CONCLUSIONS AND RECOMMENDATIONS: Phytoextraction of Pb from dredged material using chelates to solubilize Pb and the use of plants that can take up the chelated Pb appear to be potentially troublesome. Potential leaching of chelated Pb into groundwater has occurred in past attempts. Potential migration of chelated Pb offsite in rainfall runoff can occur if appropriate controls are not designed into the plan. Plants taking up Pb will require control and isolation from any wildlife near or on a CDF. All plant material will require harvesting and disposal in an appropriate place.

On the other hand, in situ immobilization of Pb in dredged material appears to be a most promising technology at this time. Comprehensive research has shown clearly that Pb can be transformed into insoluble minerals such as pyrophosphate and reduce the bioavailability to extremely low levels. Reduced risk to human health and the environment appears to be possible through the addition of appropriate amounts of phosphate materials.

This in situ immobilization and phytostabilization technology would be most appropriate for confined disposal facilities (CDFs) that have been filled to capacity and the process of removal of dredged material has been initiated. Once phosphate materials have been incorporated into the surface dredged material, the upper foot of dredged material should be removed for beneficial use. The same phosphate amendments could be applied to the next 1-ft layer of dredged material and the dredged material removed. The process can continue until a suitable quantity of reclaimed dredged material has been removed to restore storage capacity for future dredging projects.

This in situ immobilization and phytostabilization technology would also be an appropriate control measure for CDFs that have been filled to capacity and are scheduled to be used for wildlife habitat or a park or for some other beneficial site use.

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