

ERDC TR-05-9

Engineer Research and
Development Center



US Army Corps
of Engineers®

Environmental Quality Technology Program

Rainfall Lysimeter Evaluation of Leachability and Surface Transport of Heavy Metals from Six Soils With and Without Phosphate Amendment

Steven Larson, Barbara Tardy, Ken Rainwater, and
Jeb S. Tingle

September 2005

Rainfall Lysimeter Evaluation of Leachability and Surface Transport of Heavy Metals from Six Soils With and Without Phosphate Amendment

Steven Larson and Barbara Tardy

*Environmental Laboratory
U.S. Army Engineer Research and Development Center
3909 Halls Ferry Road
Vicksburg, MS 39180-6199*

Ken Rainwater

*Water Resources Center
Texas Tech University
Lubbock, TX 79409*

Jeb S. Tingle

*Geotechnical and Structures Laboratory
U.S. Army Engineer Research and Development Center
3909 Halls Ferry Road
Vicksburg, MS 39180-6199*

Final report

Approved for public release; distribution is unlimited

ABSTRACT: The addition of phosphate materials such as hydroxyapatite (HAP) has been suggested and used for treatment and stabilization of lead-contaminated soils. HAP has been used at some small arms firing ranges (SAFRs) to contain the lead in berm soils onsite. The two primary pathways for lead migration from the sites are via leachate production and surface runoff water from rainfall. The focus of this study is the in situ treatment of various representative berm soils with HAP using large berm lysimeters under simulated field conditions.

Six soils were enriched with lead by firing military rounds of M855, 5.56-mm, lead/copper/antimony bullets into the soils. The six lead-enriched soils were treated with HAP amendment and six of the same enriched soils were tested without HAP treatment. The soils were placed in the lysimeters and subjected to rainfall over a period of time. Leachate and runoff samples were collected. The effectiveness of the HAP treatment was determined by the amount of lead and other metals lost from the treated soils as compared to the soils without any treatment.

DISCLAIMER: The contents of this report are not to be used for advertising, publication, or promotional purposes. Citation of trade names does not constitute an official endorsement or approval of the use of such commercial products. All product names and trademarks cited are the property of their respective owners. The findings of this report are not to be construed as an official Department of the Army position unless so designated by other authorized documents.

Contents

Conversion Factors, Non-SI to SI Units of Measurement.....	ix
Preface.....	x
Executive Summary.....	xi
1—Introduction.....	1
Background.....	1
Objectives.....	3
2—Methods and Materials.....	4
Method Summary.....	4
Soil Collection, Preparation, and Characterization.....	4
Lead Partitioning.....	5
Physical and Chemical Analysis Procedures.....	6
Metal Concentration and Particle Size Analysis.....	7
Metal Speciation and Availability, Sequential Extraction Procedure.....	8
Laboratory Lysimeter Procedure.....	9
Lysimeter design.....	9
Experimental design.....	11
Lysimeter loading.....	11
Toxicity Characteristic Leaching Procedure.....	12
3—Results and Discussion.....	13
Soil Properties.....	13
Soil permeability.....	14
Clay analysis.....	14
Lead partition coefficient.....	15
Metal Content of Clean Soils.....	15
Particle Size Distribution in Lead-Enriched Soils.....	16
Metal Content of Lead-Enriched Soils.....	16
Metal Distribution in Lead-Enriched Soils by Particle Size.....	18
Sequential Extraction Results.....	19
Laboratory Lysimeter Procedure Results.....	25
Effluent production.....	25
Leachate and runoff pH values.....	26
TSS in leachates and runoff.....	27
Lead Loss in Leachate and Runoff.....	30
Dissolved lead.....	31
Particulate lead.....	32

Percent lead reduction	32
Accumulated Lead Mass Lost.....	33
Digested leachate lead.....	33
Digested lead in runoff.....	34
Correlation of total lead lost with sequential extraction results	38
Lead partitioning	38
Comparison of Other Metals Lost with Sequential Extraction Results	39
Copper	39
Antimony.....	40
Chromium.....	42
Nickel	43
Zinc.....	45
Iron	46
Manganese.....	48
Vanadium	48
TCLP.....	50
Clean soils	50
Untreated, lead-enriched control and lead-enriched, HAP-treated soils before LLP testing	50
Untreated, lead-enriched control and lead-enriched, HAP-treated soils after LLP testing.....	52
Phosphate Loss in Lysimeters with HAP Amendments	55
4—Conclusions and Recommendations.....	56
References	60
Appendix A: Analytical Data	A1
SF 298	

List of Figures

Figure 1.	Cross-sectional diagram of lysimeter cell	10
Figure 2.	Lysimeter cell showing plumbing fixtures	10
Figure 3.	Experimental design showing lysimeter array and content.....	12
Figure 4.	Particle size distribution of the six lead-enriched soils	17
Figure 5.	Percent of total of selected metals by particle size	20
Figure 6.	Percent of total of selected metals by particle size.....	21
Figure 7.	Comparison of collected volumes of leachate and runoff	26
Figure 8.	Average pH values of leachates and runoff from lysimeters	27

Figure 9.	Accumulated TSS in leachates from lysimeters containing: A, Clay, Silty Sand A, and Glacial Till; and B, Silty Sand B, Peat, and Loess Silt lead-enriched soils.....	29
Figure 10.	Accumulated TSS in runoff from lysimeters containing: A, Clay, Silty Sand A, and Glacial Till; and B, Silty Sand B, Peat, and Loess Silt lead-enriched soils.....	30
Figure 11.	Accumulated total lead mass in leachates from treated and untreated lead-enriched Peat and Loess Silt lysimeters	34
Figure 12.	Accumulated total lead mass in leachates from treated and untreated lead-enriched Clay and Silty Sand A lysimeters	35
Figure 13.	Accumulated total lead mass in leachates from treated and untreated lead-enriched Glacial Till and Silty Sand B lysimeters.....	35
Figure 14.	Accumulated total lead mass in runoff for treated and untreated lead-enriched Peat and Loess Silt lysimeters	36
Figure 15.	Accumulated total lead mass in runoff from treated and untreated lead-enriched Clay and Silty Sand A lysimeters	37
Figure 16.	Accumulated total lead mass in runoff from treated and untreated lead-enriched Glacial Till and Silty Sand B lysimeters	37
Figure 17.	Correlation of average digested runoff lead concentration with percent of lead in residual fraction of the sequential extraction test	38
Figure 18.	Plot comparing percent of antimony in sequential extraction residual fraction with average antimony concentration in digested runoff.....	42
Figure 19.	Percent reduction in TCLP lead by soil type before and after LLP	53
Figure A1.	Grain-size distribution for Loess Silt soil.....	A2
Figure A2.	Grain-size distribution for Clay soil.....	A3
Figure A3.	Grain-size distribution for Glacial Till soil	A4
Figure A4.	Grain-size distribution for Peat soil.....	A5
Figure A5.	Grain-size distribution for Silty Sand A soil.....	A6
Figure A6.	Grain-size distribution for Silty Sand B soil	A7

List of Tables

Table 1.	Preparation of Lead-Enriched Simulated SAFR Soils	5
Table 2.	Chemical Analytical Methods and Procedures.....	8
Table 3.	Properties of Clean Soils	13
Table 4.	Mineralogical Summary of the Six Clean Soils	14
Table 5.	Partition Coefficients (K_d) Between Lead and the Soils.....	15
Table 6.	Metal Concentrations of the Six Clean Soils Before Lead Enrichment	16
Table 7.	Average Metal Concentration of Lead-Enriched Soils (average of nine replicates, 1.7-mm fraction)	18
Table 8.	Percent of Lead and Copper in Sequential Extraction Fractions.....	22
Table 9.	Percent of Antimony in Sequential Extraction Fractions.....	22
Table 10.	Percent of Chromium in Sequential Extraction Fractions.....	22
Table 11.	Percent of Nickel in Sequential Extraction Fraction	23
Table 12.	Percent of Zinc in Sequential Extraction Fractions.....	23
Table 13.	Percent of Iron in Sequential Extraction Fractions	23
Table 14.	Percent of Manganese in Sequential Extraction Fractions.....	24
Table 15.	Percent of Vanadium in Sequential Extraction Fractions.....	24
Table 16.	Summary of Percent of Lead, Copper, and Antimony in the Least and Most Tightly Bound Fractions of the Sequential Extraction	25
Table 17.	Total Volumes of Effluent Collected Over the 16-Week Test Period.....	25
Table 18.	Total Mass of TSS Lost and Average TSS Concentration in Effluents	28
Table 19.	TSS Leachate Concentrations as Percent of Runoff Concentrations, and Percent Reduction in TSS Concentrations in Effluents After HAP Treatment	28
Table 20.	Average Effluent Lead Concentrations Over 16-Week Test Period.....	31
Table 21.	Percent Reduction in Average Lead Concentrations in Effluents	33

Table 22.	Comparison of Average Digested Effluent Lead Concentrations with the Partition Coefficient of the Soils.....	39
Table 23.	Average Effluent Copper Concentrations Over 16-Week Test Period.....	40
Table 24.	Percent Reduction in Average Copper Concentrations in Effluents	40
Table 25.	Average Effluent Antimony Concentrations Over 16-Week Test Period	41
Table 26.	Comparison of Percent of Antimony in Effluents and Percent Reduction in Average Antimony Concentrations.....	42
Table 27.	Average Effluent Chromium Concentrations Over 16-Week Test Period	43
Table 28.	Percent Reduction in Average Chromium Concentrations in Effluents	44
Table 29.	Average Effluent Nickel Concentrations Over 16-Week Test Period.....	44
Table 30.	Comparison of Percent Nickel in Effluents and Percent Reduction in Average Nickel Concentrations	45
Table 31.	Average Effluent Zinc Concentrations Over 16-Week Test Period.....	46
Table 32.	Percent Reduction in Average Zinc Concentrations in Effluents	46
Table 33.	Average Effluent Iron Concentrations Over 16-Week Test Period.....	47
Table 34.	Percent Reduction in Average Iron Concentrations in Effluents	47
Table 35.	Average Effluent Manganese Concentrations Over 16-Week Test Period	48
Table 36.	Percent Reduction in Average Manganese Concentrations in Effluents	49
Table 37.	Average Effluent Vanadium Concentrations Over 16-Week Test Period	49
Table 38.	Percent Reduction in Average Vanadium Concentrations in Effluents	50
Table 39.	TCLP Results for the Six Clean Soils	51
Table 40.	Average TCLP Metal Concentrations of Untreated, Lead-Enriched Control Soils and Lead-Enriched, HAP-Treated Soils Before LLP Testing.....	51

Table 41.	Average TCLP Metal Concentrations: Lead-Enriched Control Soils and Lead-Enriched, HAP-Treated Soils After LLP Testing	52
Table 42.	TCLP Lead Concentration Pass or Fail Results for HAP-Treated Soils Before and After LLP.....	53
Table 43.	Average Phosphate Concentrations in Leachates and Runoffs from Lysimeters.....	55
Table 44.	Phosphate Loss in Lysimeters with HAP Amendment	55
Table 45.	Comparison of Percent Lead and Antimony in the <1.0-mm Soil Fractions with the Total Amount of Lead and Antimony Lost in Effluents from Lead-Enriched Control Soils.....	58
Table 46.	Comparison of Partition Coefficient with the Total Amount of Lead Lost in Filtered Effluents from Lead-Enriched Control Soils	59
Table A1.	Sequential Extraction Procedure	A8
Table A2.	Metal Concentration Results for Clay, Silty Sand A and Glacial Till Lead-Enriched Soils.....	A9
Table A3.	Metal Concentration Results for Silty Sand B, Peat and Loess Silt Lead-Enriched Soils	A10
Table A4.	Total Mass of Metals in Lysimeters	A11
Table A5.	Leachate pH Results.....	A11
Table A6.	Runoff pH Results.....	A12
Table A7.	Leachate TSS.....	A12
Table A8.	Runoff TSS.....	A13
Table A9.	TCLP Lead Concentration Results.....	A14

Conversion Factors, Non-SI to SI Units of Measurement

Non-SI units of measurement used in this report can be converted to SI units as follows:

Multiply	By	To Obtain
cubic feet	0.02831685	cubic meters
feet	0.3048	meters
gallons (U.S. liquid)	0.003785412	cubic meters
pounds (mass)	0.4535924	kilograms

Preface

This report describes the evaluation of in situ stabilization of lead in representative small arms firing range (SAFR) soils using hydroxyapatite (HAP). Large-scale environmental systems simulating field conditions at SAFRs were used to assess the effectiveness of the HAP in stabilizing and reducing lead migration offsite. The study was conducted by the Environmental Laboratory (EL), U.S. Army Engineer Research and Development Center (ERDC), Vicksburg, MS, under the Environmental Quality Technology Program, Inorganics in Soils focus area. Dr. M. John Cullinane, Jr., EL, was Program Manager, and Dr. Steve Larson was Project Manager.

This report was prepared by Dr. Steve Larson, and Ms. Barbara Tardy, Environmental Processes and Engineering Division (EPED), Mr. Jeb S. Tingle, Engineering Systems and Materials Division, Geotechnical and Structures Laboratory, and Dr. Ken Rainwater, Director, Texas Tech University Water Resources Center, and Professor, Department of Civil Engineering, Texas Tech University. Mr. Milton Beverly and Ms. Michelle Thompson, EPED student contractors contributed to the report. This report was reviewed by Dr. Larry Jones, OA Systems Corporation, Tucson, AZ. Dr. Robert M. Engler, Senior Scientist, Mr. Roy Wade, and Mr. Michael Channel (EPED) provided in-house technical review of this report.

This study was conducted under the direct supervision of Dr. Patrick Deliman, Chief, Environmental Restoration Branch, and under the general supervision of Dr. Richard E. Price, Chief, EPED, and Dr. Elizabeth C. Fleming, Acting Director, EL.

COL James R. Rowan, EN, was Commander and Executive Director of ERDC. Dr. James R. Houston was Director.

Executive Summary

The Department of Defense operates a number of small arms training sites for military weapons proficiency. Over the years the soil at these small arms firing ranges (SAFRs) has become contaminated with various types of small caliber, metal-based ammunition. This study describes the use of a laboratory lysimeter procedure (LLP) to evaluate the potential migration of heavy metals from berm soils associated with small arms training. The study also evaluates the effectiveness of hydroxapatite (HAP) in treating, stabilizing, and reducing metals migration. Six different “clean” soil types were collected from various sites. Six lead-enriched, simulated berm soils prepared by firing lead/antimony/copper bullets into the clean soils were treated with HAP and six untreated, lead-enriched soils were loaded into twelve lysimeters. The lysimeters were exposed to sixteen weeks of simulated precipitation. Leachate and runoff samples were collected and analyzed for ten metals. The fates of the three major metal components in the simulated SAFR soils – lead, copper, and antimony – were emphasized.

The soils were thoroughly characterized to assess the effects of the soils’ physical and chemical properties on metals transport at SAFRs. A sequential extraction test was used to separate and quantify the distribution of the metals in each soil matrix. To evaluate the permanency of the HAP treatment process, the LLP leachate and runoff results were compared to the results obtained from the Toxicity Characteristic Leaching Procedure (TCLP). Unlike laboratory batch tests, the LLP more closely simulates the physical transport processes that occur on SAFRs. Due to the design of the LLP, evaluation of transport as leachate and surface runoff, as well as measuring both particulate and dissolved transport by these modes, was possible.

The laboratory lysimeter study described in this report supports the idea that HAP treatment of certain types of berm soils can cause reductions in metal losses relative to the same soils without HAP treatment. The following conclusions can be drawn from these experiments:

- Most metals were transported predominantly as particulates and exited the lysimeters in both leachate and surface runoff.
- Metal losses were dependent on soil type and initial metal concentration.

- HAP treatment reduced the lead concentrations in the dissolved and particulate leachates and runoffs by 6 to over 90 percent across the soil types, but appeared to increase the concentrations of antimony.
- HAP treatment lowered the total suspended solids (TSS) in the leachates and runoffs, which could provide the greatest reduction in lead mobility at SAFR sites.
- The permanency of the HAP treatment process as evaluated by the TCLP appears to be dependent on soil characteristics.

1 Introduction

Background

Lead contamination from spent ammunition at small arms firing ranges (SAFRs) is a widespread problem for military and civilian agencies. The Department of Defense oversees more than 3,000 active SAFRs and is currently in the process of closing more than 200 of them (Interstate Technology and Regulatory Council (ITRC) 2003). More than 160 million pounds¹ of lead are added to SAFR berms in the form of spent bullets each year. In addition, it is estimated that 9,000 nonmilitary, outdoor firing ranges are now active in the United States (U.S. Environmental Protection Agency (USEPA) 2001).

Several methods have been suggested for the in situ remediation of lead-contaminated soils. Martin and Ruby (2004), in their recent review of the subject, identified seven potential technologies: solidification/stabilization (S/S), vitrification, electrokinetic remediation, soil flushing, phytoextraction, phytostabilization, and chemical stabilization. Chemical stabilization differs from S/S in that it involves the use of chemical amendments to induce specific chemical reactions within the soil matrix that render the metal contaminants nontoxic or non-bioavailable. Unlike many S/S techniques, chemical stabilization techniques do not attempt to encapsulate the metal contaminant or reduce the permeability of the soil. Chemical stabilization usually can be accomplished in situ as it does not substantially alter the structure or potential uses of the treated soil.

Several studies have shown the potential of phosphate minerals for the in situ chemical stabilization of lead-contaminated soils (Martin and Ruby 2004, Tardy et al. 2003). The majority of these studies have been bench-scale laboratory investigations focusing on the reduction of lead solubility in the treated soils. The extent of reduction in lead solubility was usually assessed by the reduction of lead concentrations in leaching tests such as the Toxicity Characteristic Leaching Procedure (TCLP) or the Synthetic Precipitation Leaching Procedure (SPLP; USEPA 1998). The TCLP was designed to mimic the action of landfill leachate on contaminated substrates and uses mildly acidic leachants that are filtered before analysis. It is the regulatory tool used to determine if a particular material requires disposal in a Resource Conservation and Recovery Act (RCRA) certified landfill. Regulated compounds from the material that exceed a concentration value of 5 mg/L in the leachate indicate that the material will require disposal in a

¹ A table of factors for converting non-SI units of measurement to SI units is presented on page ix.

secure landfill without further treatment. The SPLP was designed to evaluate contaminate release in response to the infiltration of meteoric water. The maximum contaminate concentration level as established by the USEPA for lead in drinking water is 0 mg/L and the actionable level as required by the Safe Drinking Water Act (SDWA) is 0.015 mg/L (15 µg/L); dissolved lead concentration above this level is considered a potential health concern (USEPA 2002).

Application of soluble or solid phase phosphates (such as hydroxyapatite, HAP) to lead-contaminated soil induces the dissolution of the native lead minerals, the desorption of lead adsorbed by hydrous metal oxides, and the subsequent formation of a very stable pyromorphite [e.g., $Pb_{10}(PO_4)_6X_2$, where X may be OH, Cl, or F]. The overall result is a decrease in the bioavailability of the lead without its encapsulation or removal from the contaminated soil.

The current study (reported herein) extends work at the U. S. Army Engineer Research and Development Center (ERDC) utilizing specially designed, large-scale lysimeters for evaluating the total amount of lead leaving firing range berms at SAFRs from all pathways. In the earlier study (Tardy et al. 2003) using the Laboratory Lysimeter Procedure (LLP), application of simulated rainfall at either a neutral pH or pH 3 allowed for the collection of both neutral and slightly acidic leachate through the soil mass and runoff from the surface. One SAFR soil and two phosphate amendments were used in that study. The SAFR soil used (8,000 mg/kg lead) was amended with 1 or 5 percent of either HAP or potassium dihydrogen phosphate (KHP). Simulated rain events were conducted weekly over a 16-week test period. Leachate and runoff samples were analyzed after the samples were filtered and also after a total acid digestion without filtration. Results of the study showed that the addition of phosphate reagents did lower lead solubility so that lead levels were considerably reduced in filtered leachates (and in TCLP tests) in agreement with earlier laboratory studies (Sims 1990; Ma et al. 1993, 1995; Laperche et al. 1997; Ma and Gade 1997; Berti and Cunningham 1997; Zhang and Ryan 2000).

However, further analysis of the unfiltered leachate and runoff samples showed that substantially larger amounts of lead were lost in the particulate fraction than in the filtered (0.45-µm) effluents. Of the total mass of lead lost from all lysimeters over the 16-week test period, only 2 percent remained in the effluent fractions after they had been filtered. Ninety-eight percent of the lead was found in suspension in the filterable particulates. In addition, the vast majority of the total mass of lead that was lost (>99 percent) was found in the runoff fractions. The total amount of lead lost in many runoff samples correlated closely with the levels of total suspended solids (TSS) in the effluents.

To effectively manage lead on SAFRs, physical processes such as soil erosion and the mechanisms of lead transport must be understood. In addition, the role that soil type plays in lead transport needs to be investigated.

In comparison to the previous study, this study uses one phosphate amendment at a single concentration level to treat six different soils representative of the types of soils found at SAFRs. This study also included a thorough evaluation of the characteristics of the soils used in order to assess the effects of soil proper-

ties on lead transport at SAFRs. Unlike the previous study, simulation of “acid rain” was not included in this study.

Objectives

The objectives of this study were to:

- Compare lead losses from untreated and phosphate-treated simulated SAFR soils.
- Assess lead losses through the pathways of leaching through the soil mass and in runoff from the soil surface.
- Compare lead losses in filtered effluent samples with the total lead losses in unfiltered effluent samples.
- Evaluate permanency of phosphate treatment.
- Quantify phosphate loss.
- Identify soil characteristics that impact lead transport at SAFRs.

2 Methods and Materials

Method Summary

Based on the results from previous studies in this laboratory (Tardy et al. 2003; Larson et al. 2004), HAP powder was selected as the soil amendment for this study. Six soils with diverse properties were collected from various sites. The soils were sieved over a 26.5 mm sieve to remove rocks, debris, and particles larger than 26.5 mm. Three variations of each of the six soils were prepared and used in the study. One third of each soil was designated as the “clean” soil and was not fired on and was not enriched with lead. Two-thirds of each of the sieved soils was then enriched with lead by firing large numbers of M855 rounds into them simulating actual conditions at SAFRs. The metallic components found in the M855 round include the metallic components of brass (copper and zinc) used for the jacketing material; the possible metallic components of the carbon steel penetrator (iron, manganese, chromium, nickel, tungsten, molybdenum, vanadium); and the metallic components of the bullet core (alloyed lead/antimony). The total mass of an M855 round is approximately 4 g. The bullet core portion (lead/antimony) has a mass of 2 g of which approximately 1.9 g is lead, 0.1 g is antimony. The rest of the bullet mass is 1.23 g copper/zinc alloy for jacketing and 0.66 g steel. One-half of each of the lead-enriched soils was treated with HAP at a 5-percent weight/weight (w/w) level based upon the air dried weight of the soil. The remaining lead-enriched soil was not treated with HAP and was designated “lead-enriched” control samples. The lead-enriched control soils and the lead-enriched HAP-amended soils were loaded into separate rainfall lysimeters that were developed previously at ERDC and described in detail in Larson et al. (2004). The LLP was used to simulate rain events that occur at SAFRs and provided a pilot-scale validation of the effectiveness of the amendment to stabilize lead and other metals in the soils. Leachate and runoff effluents were collected and analyzed after each simulated rainfall event.

Soil Collection, Preparation, and Characterization

During the soil selection process, consideration was given to select soils that were representative of actual SAFR sites with active ranges and to analyze a wide spectrum of soil types, i.e., grain size, plasticity, and alkalinity. The six soils selected were:

- Clay – a loamy soil, CL

- Silty Sand A – a lateritic soil, an iron rich quartzose soil, SM
- Glacial Till – a rocky, saline soil, SM
- Silty Sand B – a quartzose soil, SM
- Peat – a Peaty soil, MH or OH
- Loess Silt – a wind-deposited silty soil, ML-CL

The clean soils were collected and transported to the ERDC in 208-L, polyethylene-lined drums or in polyethylene-lined, 1.13-cu-m super-sacks. A composite sample from all drums or super-sacks of each soil from a given location was made by placing the contents of the containers on a polyethylene lining in a large concreted area at a depth of about 30 to 60 cm. Each soil was air dried to below its natural field moisture content to the point of workability (if needed), mixed with rakes, and tilled with a rotary tiller. The soils were sieved to remove particles greater than 26.5 mm (large rocks, debris, etc.) and labeled as clean soils, and were then placed into tared, lined 208-L drums.

The lead-enriched soils were produced by loading large masses of the six clean soils into six separate catch boxes and subjecting the soils to live fire bullet impacts from M855, 5.56-mm bullets at a distance of 98.5 m at the ERDC’s Big Black River Test Site. These representative SAFR soils were labeled and are identified as “lead-enriched” soils. Table 1 presents the soil type, soil mass, and number of rounds fired per soil for the six catch box firings.

Table 1 Preparation of Lead-Enriched Simulated SAFR Soils		
Soil Type	Soil Mass, kg	Number of Rounds Fired
Clay (CL)	576	2,874
Silty Sand A (SM)	660	3,293
Glacial Till (SM)	660	3,293
Silty Sand B (SM)	660	3,293
Peat (MH)	243	1,211
Loess Silt (ML-CL)	660	3,293

Lead Partitioning

The batch technique (USEPA 1999) for determining the partition coefficient (K_d) values, the most common laboratory method for understanding the partitioning behavior of a compound with soil, was used. The six clean soils were used along with soluble lead. The soluble lead was prepared by allowing lead slugs to dissolve in water at pH 7. This solution was used instead of spiking water with soluble metal salts in order to more accurately simulate the soluble lead expected to be present on SAFRs. The concentration range for the metal in solution was between 0 and 200 ppm. A known mass of each soil was added to a beaker and a known volume and concentration of the aqueous solution of lead was added to the soil. The beaker was sealed and mixed for 72 hr. A scoping

study found no significant change in concentration (± 5 percent) occurring between 72 and 96 hr. The aqueous metal/soil solution was then centrifuged and filtered and the remaining metal concentration in the supernatant was measured. Analysis of the water samples was performed using Inductively Coupled Plasma (ICP) spectroscopy. Control experiments were performed to eliminate artificial adsorption phenomena such as adsorption to container surfaces and precipitation. The aqueous lead concentration value (mg/L) and the soil concentration value (mg/kg) were plotted. The slope of the resulting line represented the K_d . USEPA (1999), "Understanding Variations in Partition Coefficient, K_d Values," contains a detailed description of the process along with the advantages and disadvantages of techniques for measuring K_d values.

Physical and Chemical Analysis Procedures

The chemical and physical properties of the soils before and after lead enrichment were determined by standard test methods. Specific gravity, grain size analysis, and Atterberg limits were determined according to the American Society for Testing and Materials (ASTM) (2001) procedures D854, C136, and D2487, respectively.

The amount of leachate and surface runoff is related to the engineering parameters of infiltration and surface drainage, respectively. These two parameters should be inversely proportional. In other words, the higher the infiltration rate, the less surface drainage or runoff. The amount of infiltration and surface drainage is related to the permeability or hydraulic conductivity of the soil, the vegetation, and the slope of the terrain. The coefficient of permeability of soils is dependent upon several factors including fluid viscosity, grain-size distribution, pore-size distribution, void ratio, soil grain mineralogy, and degree of saturation. The coefficient of permeability varies widely with soil type, and thus the infiltration characteristic of soils is a function of soil type. The Clay (CL) used in this experiment would have a typical coefficient of permeability of less than 0.000001 cm/sec. Silty Sand A and B (SMs) would have typical coefficients of permeability ranging from 0.01 to 0.001 cm/sec. The Glacial Till (SM) would have a slightly larger permeability ranging from 1.0 to 0.01 cm/sec due to the larger aggregate particles. The coefficient of permeability of the Loess Silt (ML-CL) would range between 0.001 and 0.00001 depending upon the Clay content, and the Peat (MH/OH) material would have a similar permeability, perhaps slightly higher depending upon the organic content. Consideration of the relative permeability of the various soils should help explain the infiltration or leachate data, although other factors influence the process.

The clay content (mineralogy) of the soils was determined to provide information on the soil structure and insight into the porosity of the soil. These characteristics ultimately affect leachate quality. To determine the type of phyllosilicates present, oriented samples of the $<0.45\text{-}\mu\text{m}$ size fraction of each sample were prepared and x-ray diffraction (XRD) patterns obtained. These samples were placed in an ethylene glycol atmosphere overnight at room temperature, and XRD patterns were collected for each sample. Samples that showed expansion of the crystal structure after exposure to an ethylene glycol atmosphere compared to

air-dried pattern indicated expandable smectitic clays. Comparisons of patterns obtained before and after exposure to ethylene glycol were used to determine the amount of expandable clay present.

The initial lead content of the representative SAFR soils was determined after the samples were digested following the procedures in SW-846 Method 3051 (USEPA 1999). Chemical analyses of the soils were conducted according to the standard analytical methods listed in Table 2. The detection limit for each analyte is also listed in Table 2. Chemical characterization included metals concentrations, designated TCLP metals, pH, total organic carbon (TOC), cation exchange capacity (CEC), phosphate, sulfate (SO₄), chloride (Cl), total Kjeldahl nitrogen (TKN), total phosphorus (TP), ortho phosphate (OPO₄), ammonia nitrogen (NH₃-N) and nitrate/nitrite (NO₃/NO₂). Quantification for metals was determined on a Perkins Elmer Optima 4300DV. TOC, CEC, and phosphate concentrations were measured on a Zellweger Astro Lab TOC Analyzer Model 2100, a Thermal Jarrell Ash Model 61E, and a Dionex Ion Chromatograph Model CD 20, respectively. Sulfate, Cl, NO₃/NO₂, and OPO₄ were determined on a Dionex Ion Chromatograph Model DX-120. Following a distillation procedure on a Labconco Rapid Still II, TKN and NH₃-N were measured by manual titration. TP was analyzed on a Lachat QuickChem 8000 FIA. Each analytical run included triplicate distilled-deionized water (DDI) blanks and triplicate quality control (QC) reagent standards. The percent standard deviation of the reagent QC standards was typically less than 5 percent. If blank values exceeded the method detection limit (MDL), these values were subtracted from the measured values.

Metal Concentration and Particle Size Analysis

The distribution of metals by particle size of the soil was determined for the six lead-enriched soils. A series of eight sieve sizes (6.7, 3.35, 1.00, 0.850, 0.500, 0.250, 0.125, and 0.063 mm) were used on each of the soils. Following separation with the sieves, each particle size fraction was dried and weighed in order to determine the mass present in each fraction. The concentrations of heavy metals in each of the size fractions were determined after grinding, digesting, and analyzing the resultant soil digest for the metals of interest.

The fine material that passed through the smallest of the sieve sizes was passed through a filter press. The volume of water used for the wet sieving procedure was recorded and analyzed for soluble heavy metals. The material that remained on the filter press was dried and also analyzed for heavy metals.

Table 2 Chemical Analytical Methods and Procedures			
Parameter/Procedures	Method	Detection Limit	
		Water, mg/L	Soil, mg/kg
Digestion Procedure TCLP Extraction	SW-846-3051 ¹ SW-846-1311 ¹	NA ³ NA	NA NA
Lead	SW-846-6010B ¹	0.003	0.40
Chromium	SW-846-6010B	0.0006	0.20
Copper	SW-846-6010B	0.006	0.60
Nickel	SW-846-6010B	0.0016	0.30
Zinc	SW-846-6010B	0.003	1.2
Iron	SW-846-6010B	0.006	0.09
Antimony	SW-846-6010B	0.020	1.6
Antimony	SW-846-7000 ¹	0.003	0.30
Manganese	SW-846-6010B	0.0003	0.25
Vanadium	SW-846-6010B	0.0003	0.33
Molybdenum	SW-846-6010B	0.003	0.33
Phosphate	SW-846-9056 ¹	0.1	0.1
TOC	SW-846-9060 ¹	0.05	0.5
CEC	SW-846-9081 ¹	ND ⁴	ND
pH	SW-846-9045C ¹	NA	NA
TSS	Standard Method 2540D ²	NA	NA
SO ₄	EPA Method 300.1 SW-846-9056	0.206	0.206
Cl	SW-846-9056	0.015	0.015
TKN	Standard Method 4500-N Standard Method 4500-NH ₃ (E)	>5.0	>5.0
NH ₃ -N	Standard Method 4500-NH ₃ (B & C)	0.02	0.02
OPO ₄	EPA Method 300.1 SW-846-9056	0.061	0.061
TP	Standard Method 4500-P (B&F)	0.01 to 6.0	0.01 to 6.0
NO ₃ /NO ₂	EPA Method 300.1 SW-846-9056	0.013/0.004	0.013/0.004

TCLP = Toxicity Characteristic Leaching Procedure.
¹ USEPA (1999).
² American Public Health Association (1998).
³ NA = Not applicable.
⁴ ND = No data provided.

Metal Speciation and Availability, Sequential Extraction Procedure

The clean soils were subjected to a modified sequential extraction procedure (SEP) described by Tessier et al. (1979). This procedure (Appendix A Table A1) was used to separate the metals into five operationally defined fractions described below. The names of the fractions are attributed to the predominant phases found in the environment. The definitions are operational, e.g., any metal that dissolves in 1 M NaOAc under these conditions is referred to as a carbonate. This does not mean that the metal was necessarily in the carbonate form, only

that it dissolved in this fraction. In each case, the suspension was centrifuged at 2000 rpm for 30 min. The supernatant was decanted, filtered through a 0.45- μ m syringe filter, and analyzed for Pb following the procedures in SW846 Method 6010.

Exchangeable Pb fraction: Eighty grams dry weight of soil was mixed with 1,000 mL of 1 M MgCl_2 at pH 7 and shaken for 1 hr.

Pb carbonate fraction: The soil residue was then mixed with 800 mL of 1 M sodium acetate at pH 5 (adjusted with acetic acid) and shaken for 3 hr.

Fe-Mn oxides associated fraction: The soil residue was then extracted for 3 hr in a 95 °C water bath with 800 mL of a reducing agent, 0.04 M hydroxylamine hydrochloride ($\text{NH}_2\text{OH}\cdot\text{HCL}$) in 25 percent (v/v) acetic acid with occasional agitation. The reducing agent was used to prevent the subsequent reprecipitation of the Fe and Mn.

Organic and sulfide associated fraction: The soil residue was extracted with 240 mL each of 0.02 M HNO_3 and 30 percent H_2O_2 and adjusted to pH 2 with HNO_3 . After 2 hr in an 85 °C water bath, a second 240-mL aliquot of H_2O_2 was added and the extraction continued for another 3 hr with intermittent agitation. After cooling, 400 mL of 3.2 M NH_4OAc in 20 percent (v/v) HNO_3 was added and the solution was brought to 1,600 mL with DDI water.

Residual Pb: The remaining soil residue was removed from the centrifuge bottle, dried, ground, and digested in concentrated HNO_3 in a microwave oven according to the procedures in SW846 Method 3051.

Laboratory Lysimeter Procedure

The LLP is designed to mimic field conditions resulting from the direct action of rainwater on target berms at SAFRs. In comparison to the TCLP that only tests filtered leachates, use of the LLP allows for collection and analysis of both leachate and runoff water and additional testing of the berm soil, thus furthering the goal of understanding metal transport pathways at SAFRs. The large-scale lysimeters used in this study are described in an earlier study (Larson et al. 2004) and are briefly summarized in the following sections.

Lysimeter design

A cross-section of the lysimeters designed for this study is diagramed in Figure 1. Constructed from black, 1.9-cm thick, high-density polyethylene 78.7 cm x 78.7 cm x 61 cm (inside length times width times height) in dimension, the lysimeters were placed on stands (fabricated from angle iron) that were designed with a 0.0625 slope for collecting runoff water from the surface of the soil (Figure 2). Depending on their construction, the slopes at different SAFR sites may vary; however, this slope was selected after pre-testing indicated that a steady runoff water stream could be collected during the test without creating gullies

and excessive washout effects in the soil. The system was also designed with the added flexibility of increasing the slope, if necessary.

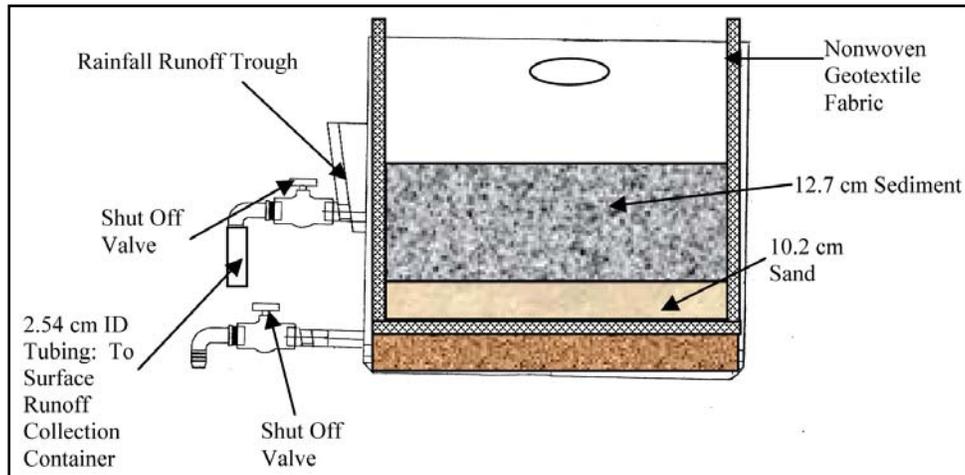


Figure 1. Cross-sectional diagram of lysimeter cell

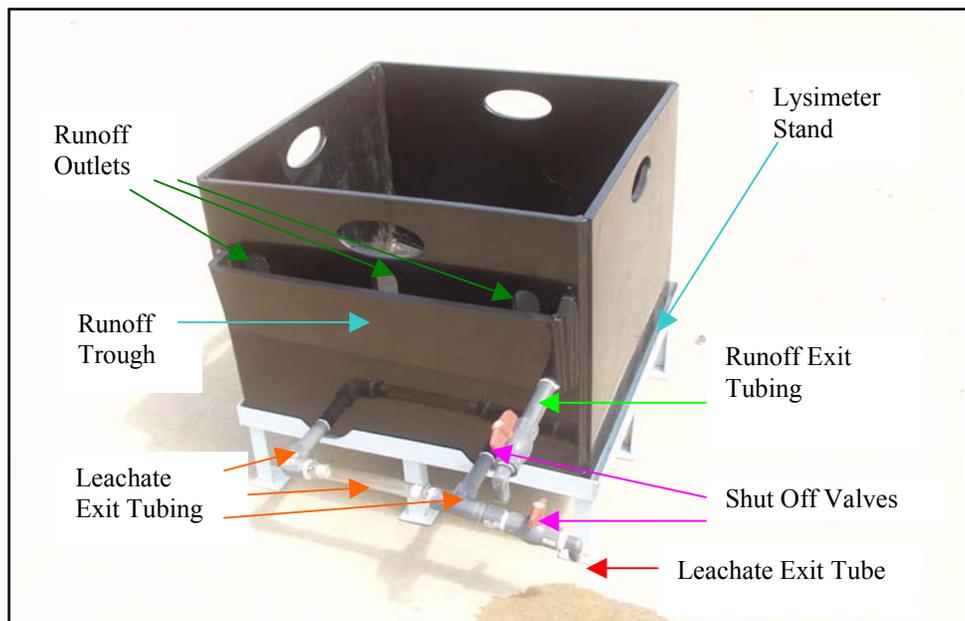


Figure 2. Lysimeter cell showing plumbing fixtures

The lysimeters were designed to allow for the collection of leachate flowing through the soil as well as runoff from the soil surface. Sufficient space remained above the soil mixture for a portion of the simulated rain to puddle and flow through the runoff trough into the runoff collection system. Leachate produced by rainwater flowing through the soil passed through the pea gravel and was collected for analysis. All tubing in the collection system was made from non-reactive silicon or polyethylene. Leachate and runoff samples were collected in polyethylene pans. Subsamples of both effluents were tested for the parameters listed in Table 2.

The rainfall was introduced through a rainfall simulator made from a clear Plexiglas box that was fitted with a piece of porous polyethylene on its bottom side. The water reservoir containing reverse osmosis (RO) water rested on mobile carts directly above the top of the lysimeter and supplied artificial rain to each lysimeter. Air pressure regulators were fitted into the top of the simulators to control and vary airflow, thus controlling rainfall rates. In this study the pressure regulators were not needed since a moderately steady rainfall rate was achieved at atmospheric pressure.

Each week eighteen liters of RO water were applied to the lysimeters in about 26 min. The weekly application of 18 L or 2.94 cm/week of simulated rainfall over the 16-week period replicated an annual rainfall of about 46.7 cm/year.

Experimental design

Twelve lead-enriched soil subsamples weighing an average of 110 kg were prepared for the lysimeters (Figure 3). Six of the lead-enriched soils were control soils without addition of HAP amendment. HAP amendment was added to the corresponding six lead-enriched soils. The HAP-amended soils were prepared by dividing each soil type into five subsamples of approximately 22 kg and mixing each soil subsample with 1.1 kg (5 percent w/w, chemical weight /soil weight on a dry weight basis) of HAP in a rotary cement mixer for 15 min. Leachate and runoff samples were collected and analyzed for specific metals after filtration through a 0.45- μ m filter and on the corresponding unfiltered samples after microwave digestion in nitric acid. The effectiveness of the HAP treatment in stabilizing metals in the soils was determined by comparing metal concentrations in effluents from the chemically treated, lead-enriched soils to their corresponding untreated, lead-enriched control soils. Comparative analyses of the clean soils provided data that quantified the level of lead enrichment attained after lead ammunition had been fired into the soils.

Lysimeter loading

A 7.6-cm layer of pea gravel was placed on the bottom of the lysimeter to prevent sediment clogging the drainage holes throughout leachate collection. A layer of non-woven geotextile was placed on the pea gravel and draped around the inside of the vessels. A 10.2-cm layer of coarse sand was placed on the geotextile and compacted to about 7.6 cm before the test soils were added. The 12 soil samples were then placed in three 7.6-cm increments over the sand layer and compacted to form a sediment layer approximately 32 cm deep.

To minimize piping and excessive void areas in the soil, prior to introducing artificial rain, the soils were saturated with RO water by allowing approximately 45 L of pressurized water to drain into each lysimeter in an upward mode through all layers. After complete saturation of the soils occurred as evidenced by a standing layer of water on the surface of the soils, the excess water was drained from each lysimeter. The lysimeters were left open at all times during the LLP.

Clay Lead-enriched Control Soil 0% HAP CELL #1	Clay Lead-enriched Soil 5% HAP CELL #2	Silty Sand A Lead-enriched Control Soil 0% HAP CELL #3	Silty Sand A Lead-enriched Soil 5% HAP CELL #4	Glacial Till Lead-enriched Control Soil 0% HAP CELL #5	Glacial Till Lead-enriched Soil 5% HAP CELL #6
Silty Sand B Lead-enriched Control Soil 0% HAP CELL #7	Silty Sand B Lead-enriched Soil 5% HAP CELL #8	Peat Lead-enriched Control Soil 0% HAP CELL #9	Peat Lead-enriched Soil 5% HAP CELL #10	Loess Silt Lead-enriched Control Soil 0% H CELL #11	Loess Silt Lead-enriched Soil 5% HAP CELL #12

Figure 3. Experimental design showing lysimeter array and content

Toxicity Characteristic Leaching Procedure

The TCLP was developed as a tool for determining if a particular material required disposal in a Resource, Conservation and Recovery Act (RCRA) certified landfill. Although the use of this technique in this manner is not consistent with the purposes for which the TCLP was developed, the procedure was performed on both the clean and the lead-enriched (HAP-treated and untreated) soils for comparison with results from other tests. The soils were subjected to the TCLP according to the procedure listed in Table 2 both before and after lead enrichment, after treatment with HAP, and after completion of the LLP.

3 Results and Discussion

Soil Properties

The initial properties of the six clean soils sieved to remove particles larger than 26.5 mm are summarized in Table 3. Graphs of the grain-size distribution for each of the same soils are presented in Appendix A, Figures A1–A6. The six soils were classified according to the Unified Soil Classification procedure (ASTM D 2487). The soils represented a wide array of soils typically found in SAFR berms. They varied from Silt and Sands with very low fines and acidic pHs between 5 and 5.5 (SM soils), to a Loess Silt soil with 99 percent fines and a pH of 8.7 (ML), to an acidic Clay soil with 89 percent fines (CL), and to a highly organic soil (Peat) with 92 percent fines and over 5 percent organic carbon (MH).

Property	Clay	Silty Sand A	Glacial Till	Silty Sand B	Peat	Loess Silt
Specific gravity	2.72	2.65	2.52	2.62	2.58	2.73
Percent fines	88.9	24.2	47.6	22.3	91.5	98.9
Percent sand	8.3	75.8	48.8	77.2	8.5	1.1
Unified soil classification	CL	SM	SM	SM	OH	ML
Percent total organic carbon (TOC)	0.427	0.134	1.8	1.24	5.16	0.87
Cation exchange capacity (CEC) meq/100 g	11	9	20	8	27	5
pH	5.12	5.51	4.96	5.48	6.75	8.67
Ca	227	412	730	152	3200	2780
Fe	3.17	1.76	8.68	1.88	5.87	2.36
Mg	113	61	58.1	26.4	602	466
Mn	1.58	0.26	2.67	0.4	0.37	1.97
K	27.7	61.4	61.1	12.7	210	17
Na	10.4	<4.00	32.9	<4.00	224	59.3
SO ₄	31	20	66	19	168	21
Cl	46	42	60	36	227	60
TKN	6.4	18	53	1.6	110	1
TP	4.7	8	15	1.5	43	13
OPO ₄	<0.3	<0.3	<0.3	<0.3	1.5	1.2
NH ₃ -N	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0
NO ₃ /NO ₂	0.93	32	49	2.2	28	0.93

Note: The unit of measurement for all metals and TKN, TP, OPO₄, and NH₃-N is milligrams per kilogram.

Soil permeability

As previously noted, the permeabilities of the soils directly affect the infiltration rates of rainfall and thus also affect the quantities of leachate and runoff generated. Granular materials are typically characterized as having relatively large coefficients of permeability, while clay soils typically have very small permeability coefficients. The Clay (CL) used in this experiment would have a typical coefficient of permeability of less than 0.000001 cm/sec. Silty Sands A and B (SMs) would have typical coefficients of permeability ranging from 0.01 to 0.001 cm/sec. The Glacial Till (SM) would have a slightly larger permeability ranging from 1.0 to 0.01 cm/sec due to the larger aggregate particles. The coefficient of permeability of the Loess Silt (ML-CL) would range between 0.001 and 0.00001 depending upon the clay content, and the Peat (MH/OH) material would have a similar permeability, perhaps slightly higher depending upon the organic content. Consideration of the relative permeability of the various soils should help explain the infiltration or leachate data, although other factors influence the process. Thus, the leachate quantity should be proportional to the permeability, while the surface runoff should be inversely proportional to both the soil permeability and measured leachate quantity.

Clay analysis

Changes in soil chemistry can degrade some clay minerals and reduce the ability of the soil to retain adsorbed metal ions. The results of both bulk and clay X-ray crystallography were used to generate the mineralogical summary presented in Table 4. Determining the clay content (mineralogy) of soils is critical because this characteristic provides important data that help determine the ability of soils to remove soluble metals from ground water and ultimately trap metals, hence, mitigating potential metal migration from berms and off ranges. The clay mineralogy also provided information concerning the soil structure that can provide insight into the porosity of the soil, ultimately affecting the leachate potential.

Table 4 Mineralogical Summary of the Six Clean Soils					
Soil	Detected Clay Components (x = mineral present, xx = predominant component)				
	Quartz	Expandable Clay	Non-expandable Clay	Feldspar	Carbonate
Peat	x	x	x	x	
Loess Silt	x	xx	x	x	x
Clay	x	x	x	xx	
Silty Sand A	x	x	x		
Silty Sand B	x		x		x
Glacial Till	x		x	xx	

Lead partition coefficient

The partition (or distribution) coefficient or constants (K_d) between lead and the clean soils were determined and are listed in Table 5. The partitioning constant describes the relative affinity for lead between the soil particles and the surrounding aqueous environment. Soils with high K_d values strongly adsorb the lead onto the soil particles and slow the rate of migration of the lead in the soil solution. A small K_d suggests faster migration rates and more rapid migration with the soil solution.

Comparison of the K_d values obtained shows a large effect due to soil type on the partitioning between sorbed and desorbed lead levels. The values varied from 157 L/kg for the sand to 36,050 L/kg for the Loess. Lead would be expected to migrate more rapidly in Silty Sand A and Silty Sand B than in the Peat Loess Silt, or Clay. However, the prediction of migration rates based upon K_d values alone does not take into account the migration of metals as colloids or metals partitioned onto suspended sediments.

Soil	Partition Coefficient for Lead, L/kg
Clay	320
Silty Sandy A	254
Glacial Till	697
Silty Sand B	157
Peat	2,670
Loess Silt	36,000

Metal Content of Clean Soils

The metal content of the six clean soils before bullet firings is presented in Table 6. The variations in results for the soils can be attributed to the differences in the soil chemistry and characteristics. The two metals Fe and Mn that are often found at high levels in soils are present in a varied range of concentrations from 1,200 to 38,000 mg/kg and from 9 to 900 mg/kg, respectively. Lead concentrations in the soils also varied with a minimum of 3.74 and a maximum of 42.8 mg/kg. These lead concentrations are consistent with lead levels found in many native soils. Antimony was below detection limits in all of the soils. The clean Loess Silt soil had low concentrations of all of the remaining metals.

**Table 6
Metal Concentrations of the Six Clean Soils Before Lead Enrichment**

Metal	Metal Content of Clean Bulk Soils, mg/kg					
	Clay	Silty Sand A	Glacial Till	Silty Sand B	Peat	Loess Silt
Lead (Pb)	12	16.3	7.41	42.8	19.1	3.74
Chromium (Cr)	20.7	18.6	10.4	6.8	28.4	2.18
Copper (Cu)	10.5	20	9.87	8.15	12.4	0.44
Nickel (Ni)	25.9	18.5	13.6	1.86	18.4	1.27
Zinc (Zn)	53.3	34.5	33	7.09	101	<2.00
Iron (Fe)	37,800	15,600	13,600	5,190	33,200	1,200
Manganese (Mn)	146	284	562	26.6	913	8.8
Molybdenum (Mo)	0.38	0.23	0.47	<0.33	0.39	<0.33
Vanadium (V)	26.1	30.3	15.7	11.6	36.2	3.27
Antimony (Sb)	<0.600	<0.600	<0.600	<0.600	<0.600	<0.600

Particle Size Distribution in Lead-Enriched Soils

Samples of the < 26.5-mm fraction of the six lead-enriched soils were wet-sieved to determine their relative particle-size distribution as shown in Figure 4. The soils can be grouped into fine-grained and granular materials. The Clay, Loess Silt, and Peat are fine-grained soils with a majority of particles < 0.076 mm. The two Silty Sands and the Glacial Till are granular materials with the majority of particles > 0.076 mm. The granular materials should generate higher quantities of leachate due to their higher permeability, but the Glacial Till's high quantity of fines would inhibit flow making it slightly less permeable than the Silty Sands. Of the fine-grained soils, the Clay and the Peat should have the lowest permeability, and thus the least leachate based upon their grain-size distribution. The Loess Silt would be expected to produce a slightly higher quantity of leachate than either the Clay or Peat, but less than the granular materials.

Metal Content of Lead-Enriched Soils

Based upon the weights of the lead and copper in the M855 rounds, the number of rounds fired, and the mass of soil, the theoretical Cu and Pb concentrations in each of the soils were 6,600 and 10,000 mg/kg, respectively. Table 7 shows the actual average metal concentrations for nine replicate samples and Tables A2 and A3 (Appendix A) show the individual, standard deviation, and percent standard deviation results obtained in the 1.7-mm fraction after analysis of each soil. Enrichment in lead and copper ranged from a 4.8-fold increase in copper for the Peat soil to over a 3,000-fold increase in lead for the Glacial Till soil. Also, considering that antimony was below the detection limit in the clean soils, enrichment of this metal in the bullet-fired soil also occurred.

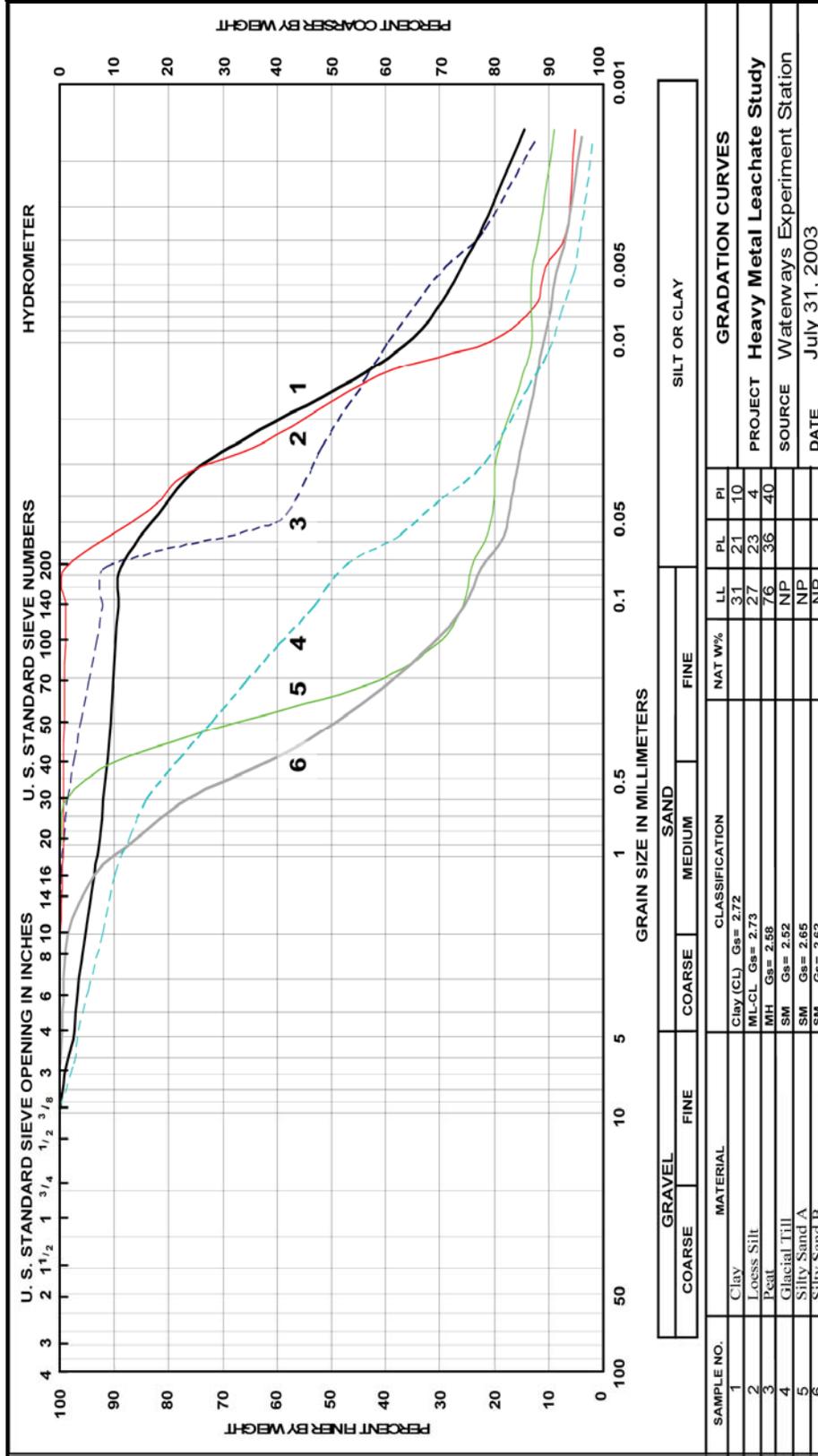


Figure 4. Particle size distribution of the six lead-enriched soils

Table 7
Average Metal Concentration of Lead-Enriched Soils (average of nine replicates, 1.7-mm fraction)

Metal	Metal Concentration of Soils After Being Fired Upon, mg/kg					
	Clay	Sandy Clay	Glacial Till	Sand	Peat	Loess
Lead (Pb)	11,300	5,900	26,000	5,930	3,857	1,918
Chromium (Cr)	44.5	12.5	35.7	19.7	39.1	27.7
Copper (Cu)	74.6	310	1883	260	58.9	88.8
Nickel (Ni)	37.3	6.7	28.7	9.7	32.3	18.7
Zinc (Zn)	97.4	42.4	225.0	42.0	150	53.7
Iron (Fe)	31,100	5,500	21,000	6,828	37,700	16,400
Manganese (Mn)	735	85.9	358	100	1137	429
Molybdenum (Mo)	1.86	1.90	1.53	1.35	0.53	1.53
Vanadium (V)	75.0	17.9	57.7	25.7	47.2	43.2
Antimony (Sb)	198	139	463	144	1.7	46.3

The total mass of each metal (the product of the metal's concentration and the mass of soil placed in the lysimeters) in the soils is shown in Table A4 in Appendix A.

Metal Distribution in Lead-Enriched Soils by Particle Size

Samples of the six lead-enriched soils were wet-sieved to determine their relative particle-size distribution. The relative amount of the five metals of interest in each particle-size fraction was also determined. The results for the soils are shown in Figures 5 and 6. In the Clay soil, about 50 percent of the metals (except copper) was found in particles greater than about 850 μm and the bulk of the remaining in the $>63\text{-}\mu\text{m}$ fraction. More than 95 percent of the copper was found in the 1,000- to 3,350- μm fraction. For the Silty Sand A soil, over 90 percent of the metals (except copper) were found in fractions between 250 and 850 μm . Copper was again found generally in the larger particle fractions.

The lower concentration of fines in the Glacial Till, Silty Sand A, and Silty Sand B soils was evident in that most of the metals in these soils was found in the larger particle-size fractions, especially in the Glacial Till where over 85 percent of the metals were found in the $>850\text{-}\mu\text{m}$ fractions. An exception was the lead content of the Silty Sand B soil; lead fairly evenly distributed throughout the particle-size fractions.

The Peat and Loess Silt soils are similar in that 90 percent of both chromium and iron was found in the $<63\text{-}\mu\text{m}$ fraction, lead was more evenly distributed, and copper was found in the larger soil fractions. The majority of the lead, copper, and antimony in these soils were found in larger particles, which comprised less than 5 percent of the total.

In general, the bulk of the copper mass was found to be in particle sizes larger than 1.0 mm in all the soils; over 90 percent of the copper was in particle sizes greater than 0.25 mm. Chromium and iron concentrations tracked closely in all of the soils.

Sequential Extraction Results

The sequential extraction technique (Appendix A Table A1) separates the metals in a soil sample into five operationally defined categories: exchangeable, carbonates, Fe-Mn oxides, organic matter and sulfides, and residual. The names of the categories are attributed to typical metal phases found in the environment. The extractions become more aggressive through the series. The latter fractions are generally considered to be tightly bound and have a lower environmental impact.

The highest percentage of lead from the lead-enriched soil fractions (Table 8) was found in the carbonate fraction for the Silty Sand A, Silty Sand B, and Glacial Till soils. These soils also had the lowest percentage of lead in the residual fraction. In contrast, the Clay, Peat, and Loess Silt all had over 50 percent of the lead found in the organic/sulfide and residual fractions with very little in the exchangeable or carbonate fractions. This condition was especially true for the Peat and Loess Silt soils, which had less than 10 percent of the lead in the exchangeable or carbonate fractions. There were considerable differences between the two soil groups, granular and fine-grained, and these differences play an important role in the amount of metal lost in the effluents. Thus, the granular materials displayed weaker bonds on the lead than the fine-grained soils indicating that granular materials are more susceptible to producing leachate contaminated with heavy metals than fine-grained soils.

Over 97 percent of the copper (Table 8) was found in the residual fractions of the sequential extraction tests in three of the soils: Clay, Glacial Till, and Loess Silt. The Peat soil had 95 percent of the copper in the residual and organic/sulfide fractions. The Silty Sand A and B soils had appreciable percentages of copper in all fractions except the exchangeable fraction. It is hypothesized that the high fines content of the Glacial Till increased the amount of copper in the residual fractions compared to the Silty Sand materials.

Antimony percentages varied widely between the different soils (Table 9) in a manner similar to that previously discussed for lead. Over 90 percent of the metal was found in the organic/sulfide and residual fractions in Clay, Peat, and Loess Silt soils. For the Silty Sands A and B and Glacial Till soils, antimony was distributed throughout the fractions.

Chromium was found in the soils at levels varying between 12.5 mg/kg in Silty Sand A to 44 mg/kg in the Clay soil. Over 98 percent of the chromium was found in the residual fraction after sequential extraction for all soils except Silty Sand A soil where 75 percent was in the residual and 25 percent in the Fe-Mn oxides fraction (Table 10). Overall chromium was tightly bound in all soils.

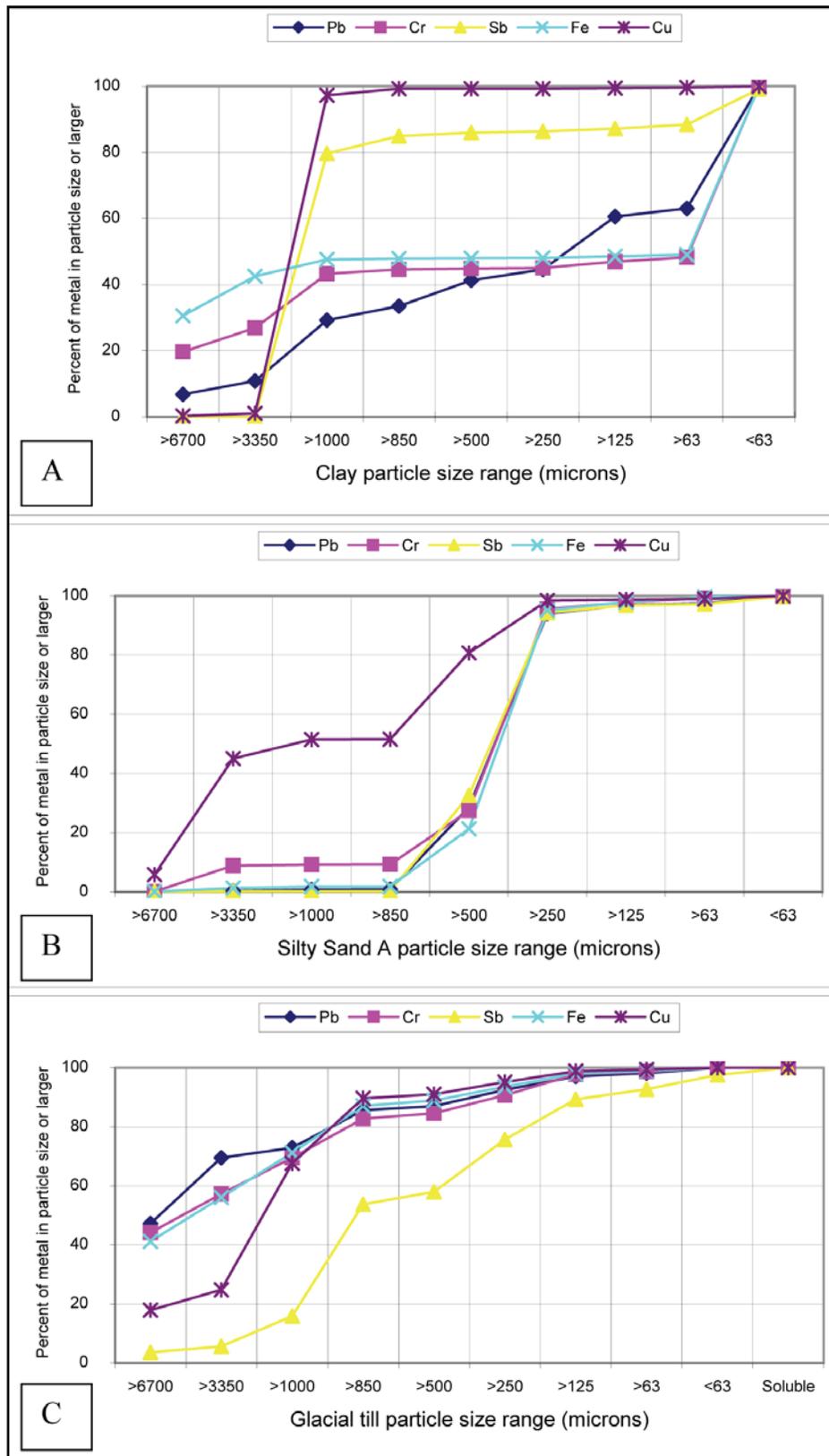


Figure 5. Percent of total of selected metals by particle size: A. Clay, B. Silty Sand A, and C. Glacial Till soils

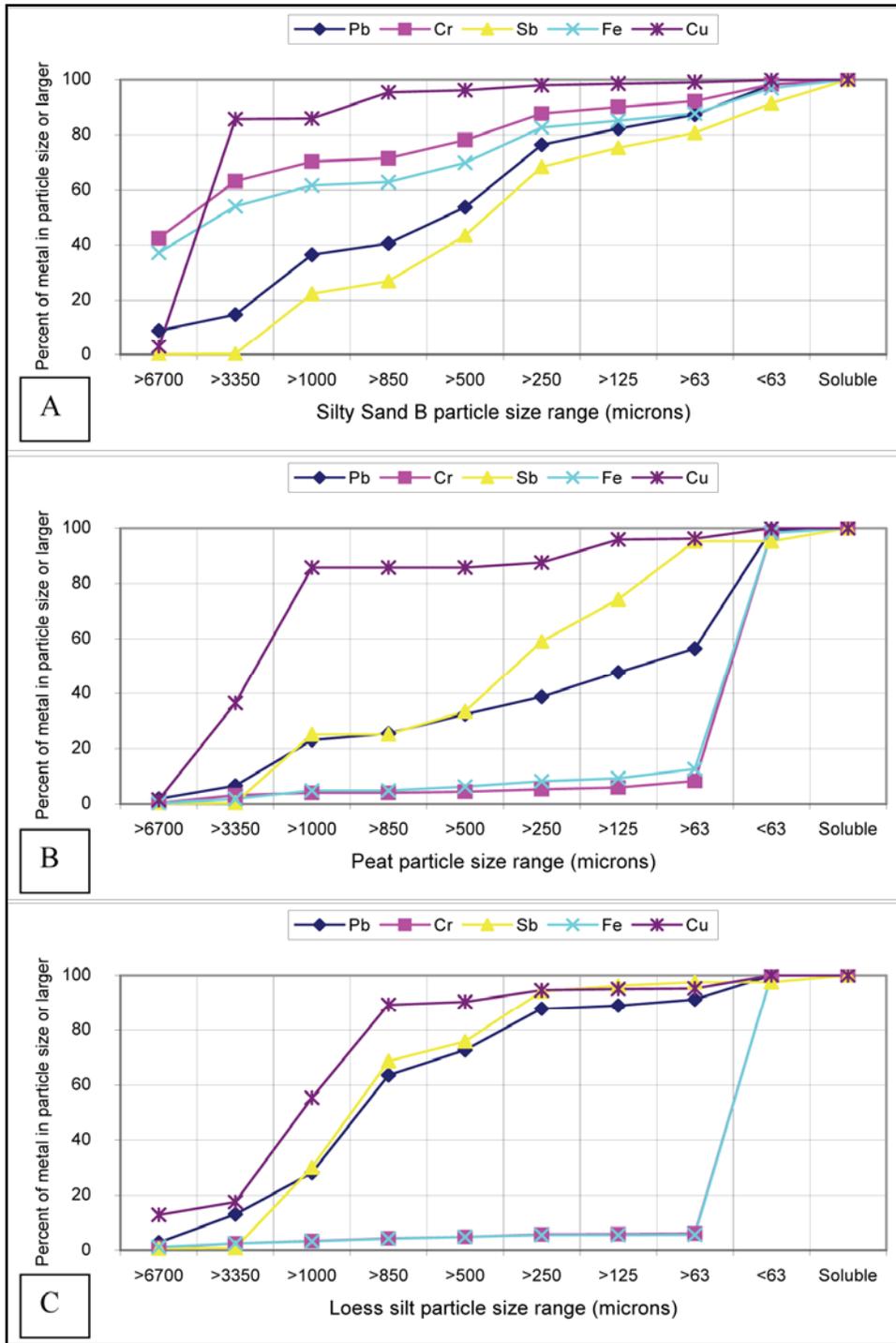


Figure 6. Percent of total of selected metals by particle size: A. Silty Sand B, B. Peat, and C. Loess Silt soils

Table 8					
Percent of Lead and Copper in Sequential Extraction Fractions					
Soil	Percent Lead in Sequential Extraction Fraction				
	Exchangeable	Carbonates	Fe-Mn Oxides	Organic Matter and Sulfides	Residual
Clay	2.2	15.9	20.3	28.3	33.4
Silty Sand A	10.1	33.8	15.7	27.9	12.5
Glacial Till	13.5	60.7	11.7	10.5	3.5
Silty Sand B	16.9	33.5	12.9	30.9	5.7
Peat	1.5	4.1	4.2	23.1	67.0
Loess Silt	0.1	7.1	12.0	26.8	53.9
Soil	Percent Copper in Sequential Extraction Fraction				
	Exchangeable	Carbonates	Fe-Mn Oxides	Organic Matter and Sulfides	Residual
Clay	<0.1	0.1	0.13	0.19	99.5
Silty Sand A	0.5	20.5	11.8	20.7	46.5
Glacial Till	<0.1	0.7	0.3	1.0	97.9
Silty Sand B	1.9	23.6	12.9	31.9	29.7
Peat	1.6	2.0	1.8	27.1	67.5
Loess Silt	<0.1	1.1	1.0	0.5	97.4

Table 9					
Percent of Antimony in Sequential Extraction Fractions					
Soil	Exchangeable	Carbonates	Fe-Mn Oxides	Organic Matter and Sulfides	Residual
Clay	<0.1	6.4	2.4	13.7	77.4
Silty Sand A	1.7	27.0	8.0	5.4	58.0
Glacial Till	8.2	54.7	4.5	5.3	27.2
Silty Sand B	6.2	24.7	6.6	23.5	39.0
Peat	<0.1	<0.1	<0.1	8.7	91.3
Loess Silt	<0.1	<0.1	<0.1	12.4	87.6

Table 10					
Percent of Chromium in Sequential Extraction Fractions					
Soil	Exchangeable	Carbonates	Fe-Mn Oxides	Organic Matter and Sulfides	Residual
Clay	<0.1	<0.1	<0.1	1.4	98.6
Silty Sand A	<0.1	<0.1	24.6	<0.1	75.4
Glacial Till	<0.1	<0.1	<0.1	<0.1	100.0
Silty Sand B	<0.1	<0.1	<0.1	1.7	98.3
Peat	<0.1	<0.1	<0.1	0.5	99.5
Loess Silt	<0.1	<0.1	2.2	<0.1	97.8

Nickel concentrations in the soils varied from 6.7 mg/kg in Silty Sand A to 37 mg/kg in the Clay soil. Between 65 and 75 percent of the nickel was found in the residual fraction of the sequential extraction test for all of the soils except Silty Sand A in which nearly 60 percent was found in the exchangeable fraction (Table 11). A significant percentage (about 9 to 12 percent) of the nickel was in the exchangeable fraction in all of the soils.

Table 11					
Percent of Nickel in Sequential Extraction Fraction					
Soil	Exchangeable	Carbonates	Fe-Mn Oxides	Organic Matter and Sulfides	Residual
Clay	10.1	5.3	4.4	4.1	76.0
Silty Sand A	58.1	20.2	<0.1	<0.1	21.8
Glacial Till	10.3	7.0	3.8	3.1	75.7
Silty Sand B	18.1	8.7	4.6	3.7	65.0
Peat	8.6	5.6	3.4	8.5	73.9
Loess Silt	11.9	5.5	11.9	bdl	70.8

Zinc concentrations in the soils varied from 42 mg/kg in the Silty Sand A and B soils to 225 mg/kg in the Glacial Till soil. In three of the soils, Clay, Glacial Till, and Loess Silt (Table 12), over 90 percent of the zinc was in one component, the residual fraction of the sequential extraction test, while in the Peat soil the organic matter/sulfide and residual fractions together made up over 90 percent of the zinc. Only in the Silty Sand A and B soils, which had the lowest zinc concentrations, was the zinc distributed more uniformly throughout the fractions. The fine-grained soils and the Glacial Till with its significant fines content appear to tightly bind the heavy metals compared to the cleaner granular soils.

Table 12					
Percent of Zinc in Sequential Extraction Fractions					
Soil	Exchangeable	Carbonates	Fe-Mn Oxides	Organic Matter and Sulfides	Residual
Clay	1.0	0.3	0.7	1.4	96.6
Silty Sand A	3.9	14.5	22.4	15.3	43.9
Glacial Till	0.2	0.7	0.5	1.0	97.5
Silty Sand B	8.4	18.5	16.9	21.4	34.8
Peat	1.4	2.0	4.1	19.1	73.4
Loess Silt	<0.1	1.4	6.4	1.6	90.6

Large amounts of iron in the soils were found in the residual fraction of the sequential extraction tests. The only exception was the Silty Sand A in which 30 percent of the iron was found in the Fe-Mn oxide fraction (Table 13).

Table 13					
Percent of Iron in Sequential Extraction Fractions					
Soil	Exchangeable	Carbonates	Fe-Mn Oxides	Organic Matter and Sulfides	Residual
Clay	<0.1	<0.1	3.1	0.2	96.8
Silty Sand A	<0.1	0.2	30.2	1.3	68.3
Glacial Till	<0.1	0.0	1.7	0.3	98.0
Silty Sand B	<0.1	0.1	6.2	0.2	93.4
Peat	<0.1	<0.1	1.6	0.2	98.2
Loess Silt	<0.1	<0.1	4.9	0.1	95.1

The original manganese concentrations in the soils varied from 96 mg/kg in the Silty Sand A to 1,137 mg/kg in the Peat soil. The distribution of the metal in the sequential extraction fractions was quite variable (Table 14). In general, very little manganese was in the exchangeable fraction. Large percentages were found in the Fe-Mn oxides and the residual fractions for all soils except the Loess Silt in which 88 percent was found in the organic matter/sulfide fraction.

Soil	Exchangeable	Carbonates	Fe-Mn Oxides	Organic Matter and Sulfides	Residual
Clay	0.4	3.1	50.7	11.0	34.8
Silty Sand A	1.9	43.4	41.6	2.6	10.5
Glacial Till	0.5	5.6	9.9	2.6	81.4
Silty Sand B	1.4	6.6	80.1	3.8	8.1
Peat	0.2	0.8	19.3	14.6	65.1
Loess Silt	<0.1	0.6	6.4	88.0	4.9

As seen in Table 15, over 95 percent of the vanadium was found in the residual sequential extraction fraction of all soils except for the Silty Sand A and B soils in which higher percentages were in the Fe-Mn oxides and organic matter/sulfide fractions.

Soil	Exchangeable	Carbonates	Fe-Mn Oxides	Organic Matter and Sulfides	Residual
Clay	<0.1	0.2	1.8	0.6	97.4
Silty Sand A	<0.1	<0.1	34.7	8.5	56.9
Glacial Till	<0.1	0.1	1.7	1.2	97.0
Silty Sand B	<0.1	1.5	7.0	7.0	84.6
Peat	<0.1	0.1	1.8	0.3	97.8
Loess Silt	<0.1	<0.1	4.4	0.6	95.0

Table 16 summarizes the percentages of lead, copper, and antimony in the least (exchangeable and carbonate) and most tightly bound (organic matter and sulfide, and residual) fractions. The soils fell into two major groups: Silty Sand A and B and Glacial Till soils all had large amounts of the metals in the most available fractions, while the Clay, Peat, and Loess Silt soils had the majority of the metals in the least available fractions. Lead and antimony were particularly high in the easily available fractions especially in the Glacial Till. Copper was found largely in the least available fractions in all soils.

Fractions	Metal	Clay	Silty Sand A	Glacial Till	Silty Sand B	Peat	Loess Silt
Exchangeable + carbonate	Pb	18	44	74	50	6	7
	Cu	0.1	21	1	25	4	0
	Sb	<0.1	29	63	31	<0.1	<0.1
organic matter and sulfide + residual	Pb	62	40	14	37	90	81
	Cu	100	67	99	62	95	98
	Sb	91	63	33	62	100	100

Laboratory Lysimeter Procedure Results

Leachate and runoff samples were collected each week and analyzed for pH, metal content, and phosphate concentration. The results are discussed in the following sections.

Effluent production

In general, from 61 to 81 percent of the effluent was collected as leachate passing through the soil mass and from 19 to 39 percent as runoff. The total effluent volumes are given in Table 17 and are illustrated in Figure 7.

Soil and Treatment	Total Leachate, L	Total Runoff, L	Total Effluent, L	Leachate as % of Total	Runoff as % of Total	Leachate to Runoff Ratio
Clay	193.0	57.1	250	77	23	3.38
Clay + 5% HAP	167.7	83.0	251	67	33	2.02
Silty Sand A	183.0	78.6	262	70	30	2.33
Silty Sand A + 5% HAP	185.8	78.8	265	70	30	2.36
Glacial Till	208.8	54.6	263	79	21	3.82
Glacial Till + 5% HAP	166.1	86.8	253	66	34	1.91
Silty Sand B	162.7	82.7	245	66	34	1.97
Silty Sand B + 5% HAP	175.1	67.8	243	72	28	2.58
Peat	203.3	47.8	251	81	19	4.25
Peat + 5% HAP	184.9	70.2	255	72	28	2.63
Loess Silt	153.3	65.0	218	70	30	2.36
Loess Silt + 5% HAP	144.5	90.8	235	61	39	1.59
Average	177.3	72.0	249	71	29	2.46

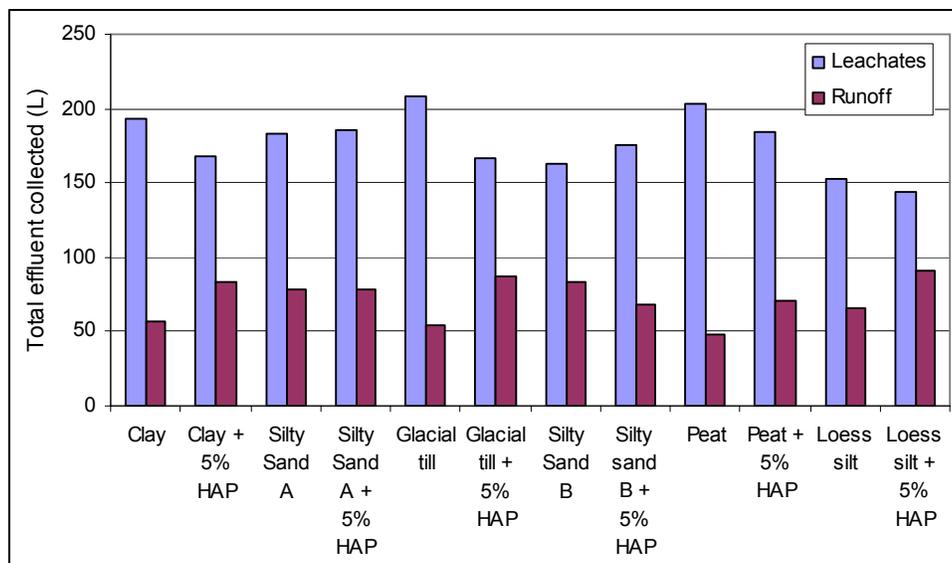


Figure 7. Comparison of collected volumes of leachate and runoff

In four of the six lysimeters, HAP addition decreased the total volumes of leachate by as much as 6 to 20 percent and increased the amount of runoff by 40 to 60 percent. The addition of HAP may have lowered the permeability of the soils slightly. Silty Sand A treated and control effluent volumes were nearly identical. Although Silty Sand B had slightly larger amounts of leachate and smaller volumes of runoff after HAP treatment, this result may also be attributable to other factors such as absorptive capacity, evaporation losses from the open lysimeters between weekly tests, and/or more rapid surface drying and rewetting of the sand soils.

The ratio of leachate to runoff collected during the LLP is also shown in the last column in Table 17. A comparison of the ratios of leachates and runoffs from the lead-enriched control soils without HAP treatment shows broad variations for different soil types. The ratios for the untreated soils from highest to lowest were in the order of Peat, Glacial Till, Clay, Loess Silt, Silty Sandy A, and Silty Sand B. SAFRs have a variety of different soil types, rainfall amounts, rainfall frequencies, rainfall intensities, and rainfall durations. Since artificial rain conditions were about the same for all 12 lysimeters and soil type varied, these results indicate that soil type impacts leachate and runoff production and is a major factor in addressing reductions of metal losses from contaminated sites.

With the exception of Silty Sand A and Silty Sand B, a decrease in the ratios of leachates to runoffs was observed for the same lead-enriched soils after HAP treatment. A slight increase in the ratio was noted for Silty Sand B after HAP treatment while the Silty Sand A remained about the same.

Leachate and runoff pH values

The average pH values of the leachate and runoff samples are shown in Figure 8. The individual results are shown in Tables A4 and A5 in Appendix A.

The pH values varied closely around pH 7 throughout the test period. In general, the Peat effluents were slightly more acidic and the Loess Silt effluents more basic than effluents from the other soils. HAP addition had only a minimal effect on effluent pH values except in the Loess Silt soils in which increased leachate and runoff average leachate were observed.

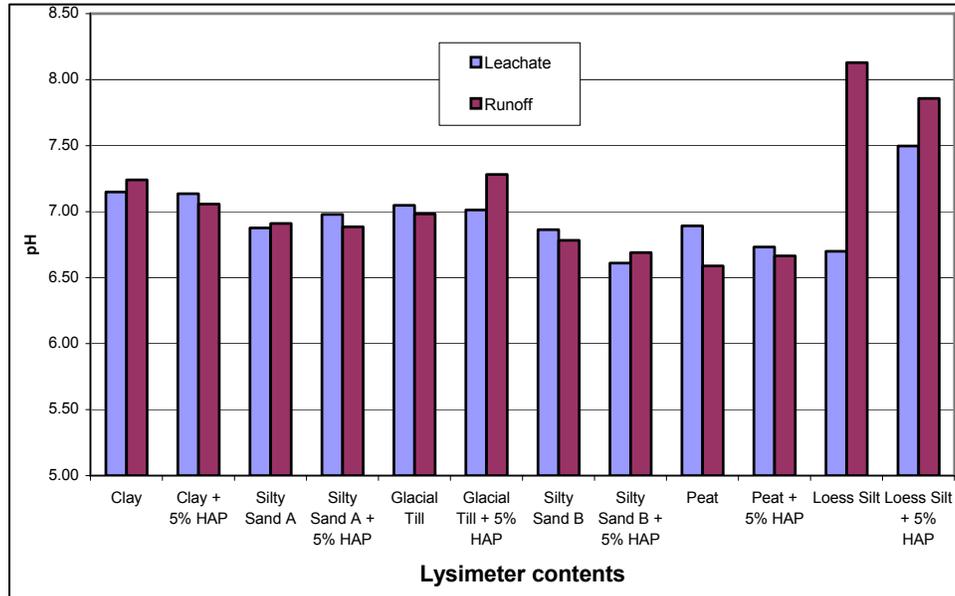


Figure 8. Average pH values of leachates and runoff from lysimeters

TSS in leachates and runoff

The TSS concentrations in the effluents varied widely depending upon the soil composition and structure. The individual weekly TSS results are shown in Appendix A, Tables A6 and A7, and the average TSS concentrations are shown in Table 18. TSS concentrations varied from 21 mg/L for the HAP-treated Silty Sand B leachate to over 4,140 mg/L for the HAP-treated Loess Silt runoff. The soils with the highest to lowest TSS concentrations in leachates were Clay, Glacial Till, Loess Silt, Silty Sand A, Peat, and Silty Sand B, respectively. The soils with the highest to lowest TSS concentrations in runoff were Loess Silt, Clay, Silty Sand B, Silty Sand A, Glacial Till, and Peat, respectively. TSS leachate concentrations as a percentage of runoff TSS concentrations ranged from about 4 to 60 percent (Table 19). Reductions in TSS concentrations in the treated effluents were above 40 percent for 9 of the 12 soils, while the remaining three effluents were below 40 percent.

Table 18
Total Mass of TSS Lost and Average TSS Concentration in Effluents

Soil and Treatment (% fines)	TSS in leachate		TSS in runoff	
	Total Mass Lost, mg	Avg. Conc., mg/L	Total Mass Lost, mg	Avg. Conc., mg/L
Clay (89)	45,500	243	125,500	2,300
Clay + 5 HAP	7,500	43	42,900	473
Silty Sand A (24)	18,500	106	111,800	1,300
Silty Sand A + 5 HAP	12,500	69	50,800	584
Glacial Till (48)	36,700	180	68,500	1,300
Glacial Till + 5 HAP	13,200	76	77,000	859
Silty Sand B (22)	6,200	41	125,900	1,400
Silty Sand B + 5 HAP	3,380	21	34,100	488
Peat (91.5)	12,200	58	9,060	158
Peat + 5 HAP	5,300	31	4,630	53
Loess Silt (99)	21,500	152	366,400	4,100
Loess Silt + 5 HAP	18,500	126	161,000	2,200

Table 19
TSS Leachate Concentrations as Percent of Runoff Concentrations, and Percent Reduction in TSS Concentrations in Effluents After HAP Treatment

Soil and Treatment	Leachate TSS Conc. as % of Runoff TSS Conc.	Percent Reduction in TSS Conc. in Leachates by HAP Treatment	Percent Reduction in TSS Conc. in Runoff by HAP Treatment
Clay	10.8	82.3	79.0
Clay + 5% HAP	9.1		
Silty Sand A	8.0	34.8	55.7
Silty Sand A + 5% HAP	11.8		
Glacial Till	14.3	57.9	31.7
Glacial Till + 5% HAP	8.8		
Silty Sand B	2.9	48.8	65.6
Silty Sand B + 5% HAP	4.3		
Peat	36.5	47.1	66.7
Peat + 5% HAP	58.0		
Loess Silt	3.7	17.1	46.9
Loess Silt + 5% HAP	5.8		

Total accumulated TSS in leachates for the six soils is shown in Figure 9 and in runoff in Figure 10. Total accumulated TSS was calculated by multiplying individual weekly TSS concentrations by the volume of leachate or runoff collected then summing the totals over the 16-week test period. The x-axis shows the individual weekly leachate and runoff sample number. The y-axis shows the accumulated TSS in milligrams. As illustrated for all lysimeters, HAP treatment lowered the total accumulated TSS in all the leachates (Figure 9) and also lowered the total accumulated TSS in the runoffs. The implication of these findings is that HAP treatment may produce a reduction of lead mobility at SAFR sites.

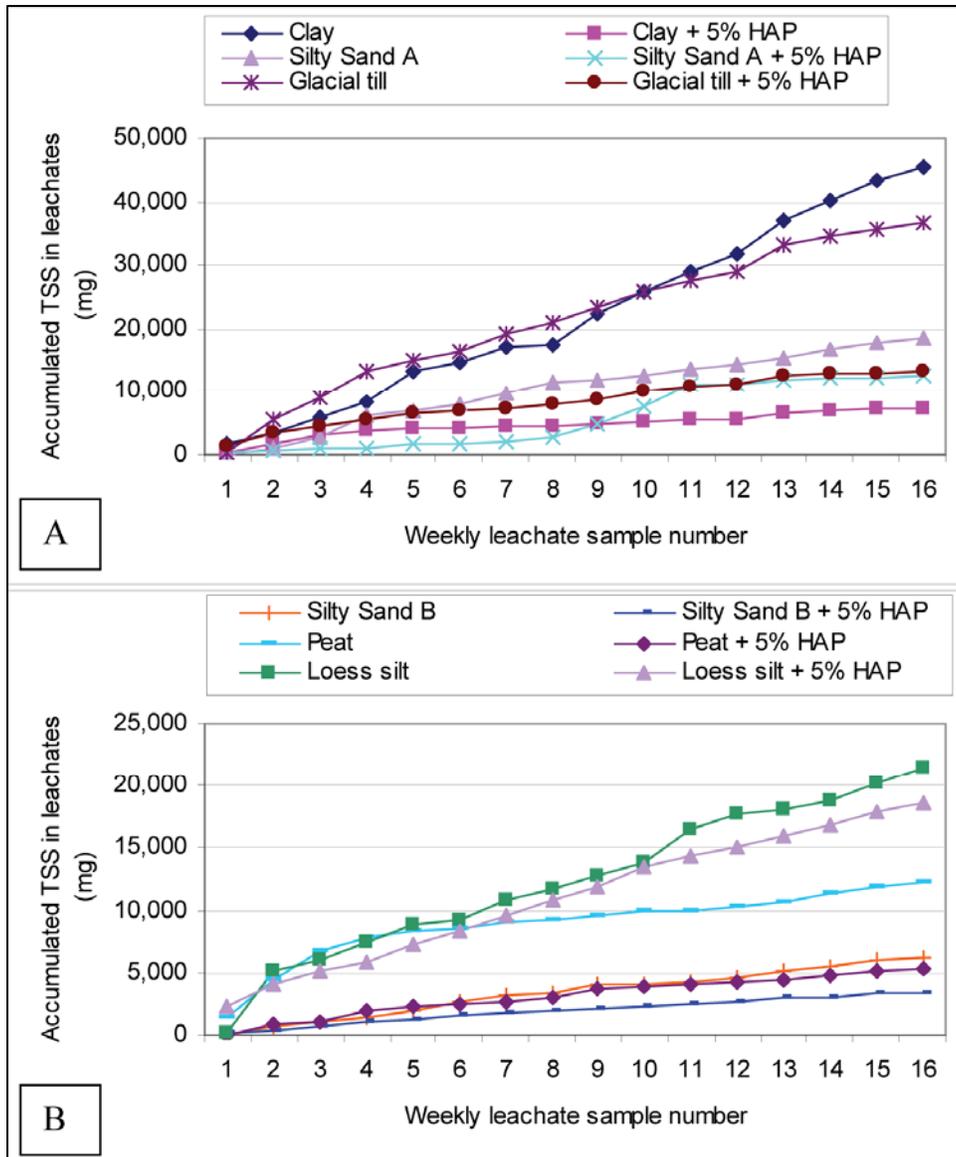


Figure 9. Accumulated TSS in leachates from lysimeters containing: A, Clay, Silty Sand A, and Glacial Till; and B, Silty Sand B, Peat, and Loess Silt lead-enriched soils

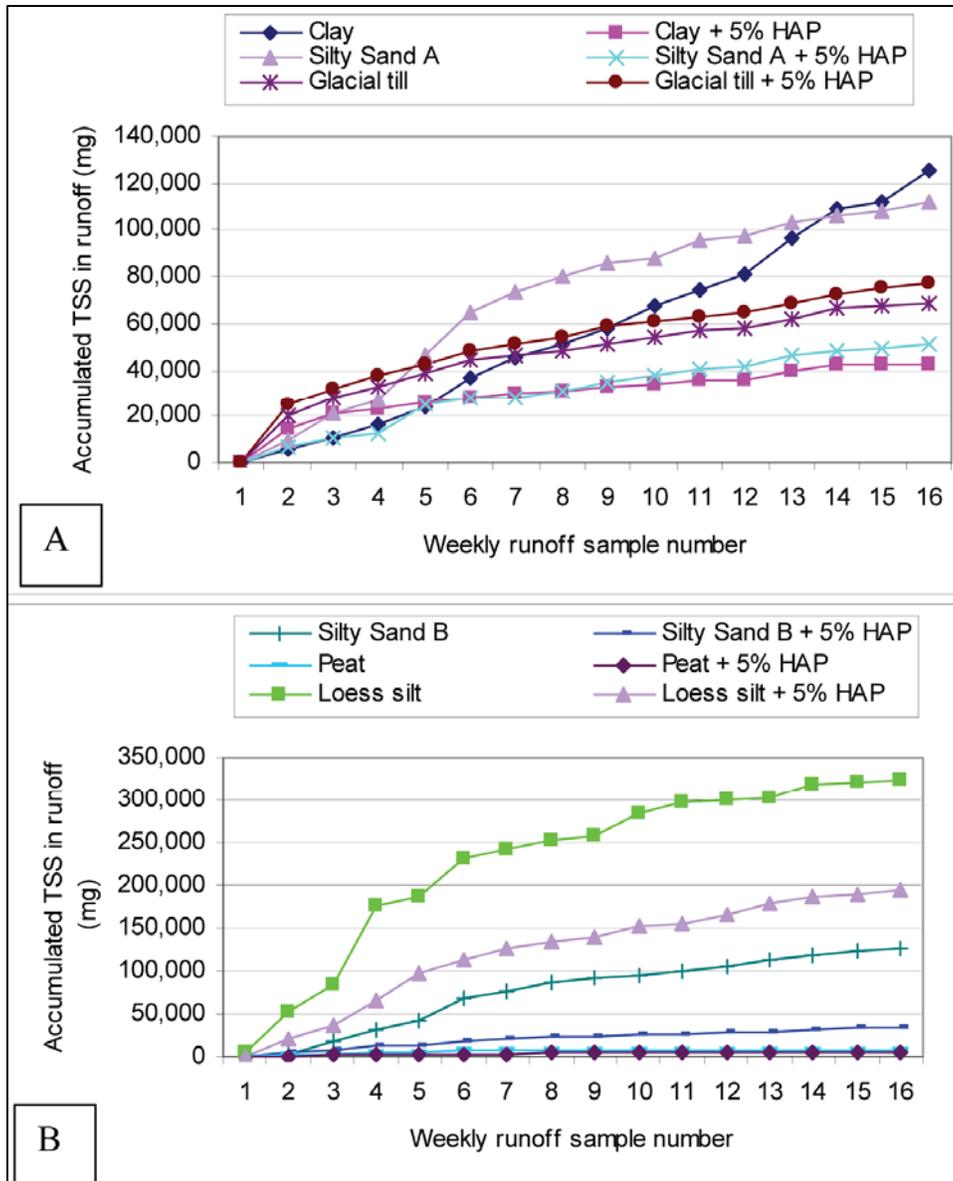


Figure 10. Accumulated TSS in runoff from lysimeters containing: A, Clay, Silty Sand A, and Glacial Till; and B, Silty Sand B, Peat, and Loess Silt lead-enriched soils

Lead Loss in Leachate and Runoff

After each weekly, simulated rain event, samples of leachate and runoff from each lysimeter were analyzed for nine metals including lead for both dissolved and total metal concentration. Dissolved metals are defined in the Standard Methods for the Examination of Water and Waste Water (Method 3010) as “those constituents (metals) of an unacidified sample that pass through a 0.45 μm membrane filter” and total metals are defined as “the concentration of metals determined on an unfiltered sample after vigorous digestion, or the sum of the concentrations of metals in both dissolved and suspended fractions” (American

Public Health Association 1998). The metal concentrations in the filtered (dissolved metals) samples were an estimation of the metals lost in solution or on very small (<0.45 µm) microparticulates while metal concentrations in the digested samples included metals in, or adsorbed onto, larger particulates. The discussion in the following sections includes all of the metals analyzed with an emphasis placed on lead, copper, and antimony which are the three major metals components in the M855 military bullets used to enrich the simulated SAFR soils.

Dissolved lead

The lead concentration in the soils loaded into the lysimeters was expected to be approximately 10,000 mg/kg based on the soil mass and number of bullets fired into each soil (Table 1). As shown previously in Table 7, the lead concentrations in the < 1.7-mm size fraction of the soils loaded in the lysimeters were typical of contaminated SAFR soils ranging from about 1,920 mg/kg in the Loess Silt to 26,000 mg/kg in the Glacial Till. The average concentrations of dissolved (filtered) lead and particulate (digested) lead found in the effluents from the lysimeters over the 16-week test period are shown in Table 20.

Table 20								
Average Effluent Lead Concentrations Over 16-Week Test Period								
Soil and Treatment	Average Lead Concentration, µg/L							
	Leachate				Runoff			
	Filtered		Digested		Filtered		Digested	
	Avg	Std	Avg	Std	Avg	Std	Avg	Std
Clay	<3	5.2	1,150	722	100	117	9,450	6,150
Clay + 5% HAP	<3	0.0	114	85	<3	0.0	1,470	742
Silty Sand A	36.9	35.0	980	433	347	315	17,600	8,940
Silty Sand A + 5% HAP	<3	0.0	90	56	109	123	6,610	3,440
Glacial Till	30.3	47.5	8,680	5,130	439	347	48,780	9,520
Glacial Till + 5% HAP	<3	1.6	1,770	1,130	127	185	30,900	21,560
Silty Sand B	96.0	78.7	423	176	4,570	4090	26,600	17,330
Silty Sand B + 5% HAP	4.28	4.6	130	66	275	303	6,240	4,050
Peat	<3	2.7	102	75	105	100	267	129
Peat + 5% HAP	<3	7.4	40	37	19	23	120	102
Loess Silt	<3	0.5	261	147	23	26	2,250	861
Loess Silt + 5% HAP	<3	0.0	233	37	8	15	2,110	1,650

Avg = Average value.
Std = Standard deviation.

For the six untreated, control lysimeters, dissolved lead average concentrations in the filtered leachate samples ranged from less than the method detection limit to 96 µg/L in the Silty Sand B soil; the untreated filtered runoff samples ranged from 23 to 4,570 µg/L. Lead concentrations in leachates from the HAP-treated cells were lower relative to the untreated control cells in all of the filtered samples. In comparing the filtered leachates to the SDWA standard of 15 µg/L,

prior to HAP addition, one-half of the soils (Silty Sand A and B and Glacial Till) exceeded the standard and one-half was below the standard (Clay, Peat, and Loess Silt). After HAP addition, average lead concentrations in the filtered leachates from the three soils that were higher than the standard were reduced to below the 15- $\mu\text{g/L}$ action level. In comparing the filtered runoffs to the SDWA standard of 15 $\mu\text{g/L}$, all six of the untreated control soils exceeded the standard. After HAP addition, average lead concentrations in the filtered leachates from two of the treated soils (Clay and Loess Silt) were reduced to below the 15- $\mu\text{g/L}$ action level.

Particulate lead

The majority of the lead was found in the particulate fractions in the digested effluents (Table 20). The digested particulate samples contained lead levels higher than their corresponding filtered (dissolved lead) samples. Before HAP treatment, average lead concentrations in the leachate samples ranged from 102 $\mu\text{g/L}$ in the Peat soil to about 8,700 $\mu\text{g/L}$ in the Glacial Till simulated soils. The range was 267 $\mu\text{g/L}$ in the Peat soil to about 48,000 $\mu\text{g/L}$ in the Glacial Till for the untreated digested runoff samples. Lead concentrations in the digested runoff samples ranged from 3 to 73 times greater than the lead concentrations in the corresponding digested leachate samples, an indication that particulate lead from runoff is a significant route of lead migration on SAFRs.

Percent lead reduction

The percent reduction in average lead concentrations in effluents from HAP treatment is shown in Table 21. If the metal concentrations were below the method detection limits or the standard deviation exceeded the average metal concentration, then the percent reduction after HAP treatment was listed as a negligible change.

HAP treatment reduced the lead concentrations in the digested leachate and runoff samples. HAP lowered the lead concentration in the digested leachate samples from the Glacial Till soil by the largest amount, 6,910 $\mu\text{g/L}$, representing an 80-percent reduction. However, the highest percentage reduction in the digested leachate samples (91 percent) occurred after HAP treatment of the Silty Sand A soil. The lowest reduction of 11 percent from 261 to 233 $\mu\text{g/L}$ or 28 $\mu\text{g/L}$ occurred in the Loess Silt soil.

The same trend was noted for the digested runoff samples. The largest actual reduction in concentration from 26,600 $\mu\text{g/L}$ to 6,240 $\mu\text{g/L}$ (Table 20) occurred in the Silty Sand B soil, representing a decrease of 20,360 $\mu\text{g/L}$ and a corresponding reduction of 77 percent (Table 21), however, the highest percentage reduction (84 percent) was noted for the Clay soil (Table 21) in which the lead concentration was reduced from 9,450 to 1,470 $\mu\text{g/L}$. The results in Tables 20 and 21 may indicate that soils with higher initial lead concentrations might need HAP treatment at levels above the 5 percent amount used in this study. On the other hand, soils with relatively low initial lead concentrations might not benefit

from HAP treatment. The issue of optimal HAP dosage is left for further research. These data indicate that the HAP treatment was more effective in the more loosely bound heavy metals of the granular soil.

Soil and Treatment	Percent Reduction in Lead Conc. After HAP Treatment			
	Leachate		Runoff	
	Filtered	Digested	Filtered	Digested
Clay	Negligible change	90	Negligible change	84
Clay + 5% HAP				
Silty Sand A	Negligible change	91	Negligible change	63
Silty Sand A + 5% HAP				
Glacial Till	Negligible change	80	Negligible change	37
Glacial Till + 5% HAP				
Silty Sand B	Negligible change	69	Negligible change	77
Silty Sand B + 5% HAP				
Peat	Negligible change	61	Negligible change	55
Peat + 5% HAP				
Loess Silt	Negligible change	11	Negligible change	6
Loess silt + 5% HAP				

Accumulated Lead Mass Lost

Lead was lost from all lysimeters over the 16 simulated rain events. The results expressed as accumulated lead mass lost and reported in micrograms are the product of multiplying the volume of each effluent by its corresponding concentration. The cumulative results are a summation of the data beginning with the first week of testing through the sixteenth week. The accumulated lead mass loss in digested leachates for the six soils is illustrated in Figures 11, 12, and 13, and for digested runoff samples in Figures 14, 15, and 16.

Digested leachate lead

Figure 11 illustrates the total (digested) accumulated leachate lead mass losses for the Peat and Loess Silt soils. There is a steady increase in lead mass lost from the untreated and HAP-amended Loess soils over the 16-week period. Although HAP treatment lowered the amount of lead leaving the system, the two almost similar trend lines indicate some uniformity in lead mass losses. The results show that HAP treatment had a minimal effect in reducing lead mass loss from the Loess Silt soil. HAP addition moderately lowered the lead mass loss from the Peat soil in comparison to the control, untreated soil. The two trend lines for the Peat soils are somewhat dissimilar. The HAP-treated Peat soil showed a lower, steady, and consistent lead mass loss, with a greater than 50-percent reduction in the total mass of lead leaving the lysimeter via leaching when compared to the untreated control.

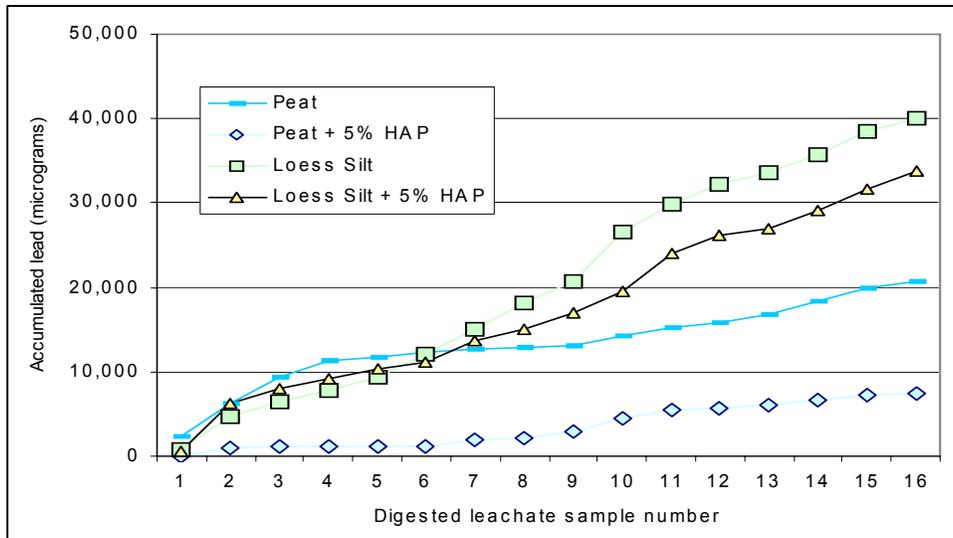


Figure 11. Accumulated total lead mass in leachates from treated and untreated lead-enriched Peat and Loess Silt lysimeters

Figure 12 shows total accumulated (digested) leachate lead mass losses for the Clay and Silty Sand A soils. The trend lines show a pattern of continual, steady leaching of lead from the Clay and Silty Sand A untreated soils. In addition, lead mass loss from the untreated Clay soil was virtually linear. In contrast, lead mass losses from the HAP-treated Clay and Silty Sand A soils were much lower than the losses from the untreated control soils indicating that the HAP treatment was effective in reducing lead mass losses from the two soils. The almost flat trend lines for the Clay and Silty Sand A soils that appear to plateau near the end of the 16-week period may also indicate that HAP amendment may have a stabilizing effect on lead mass losses for the two soils.

Figure 13 shows the accumulated leachate lead mass loss for the Glacial Till and Silty Sand B soils. The trend line for the untreated Glacial Till soil shows a steadily increasing lead mass loss and is almost linear while the HAP-treated soil showed only a slight increase in lead mass loss over the 16-week LLP period. The results for the treated and untreated Silty Sand B soils were nearly identical.

Digested lead in runoff

The runoff lead mass loss for the Peat and Loess Silt soils is depicted in Figure 14. The lead mass losses from the treated and untreated Peat soils were comparable while there was a decrease in lead mass loss after HAP treatment of the Loess Silt soil.

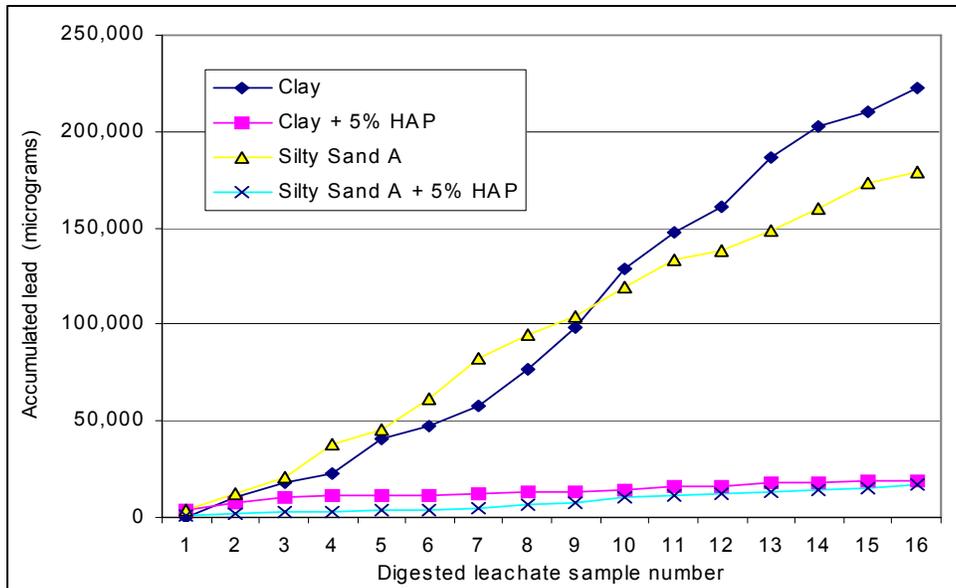


Figure 12. Accumulated total lead mass in leachates from treated and untreated lead-enriched Clay and Silty Sand A lysimeters

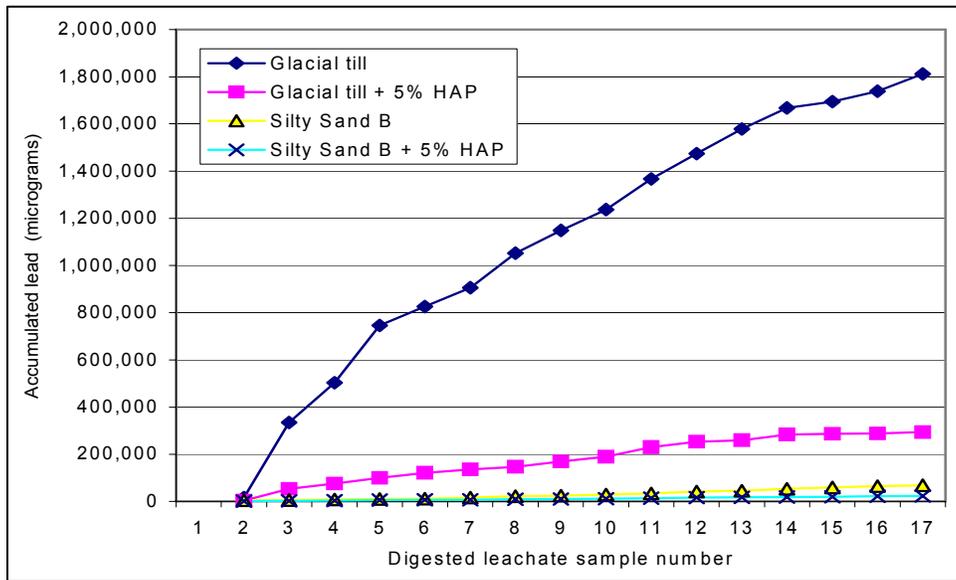


Figure 13. Accumulated total lead mass in leachates from treated and untreated lead-enriched Glacial Till and Silty Sand B lysimeters

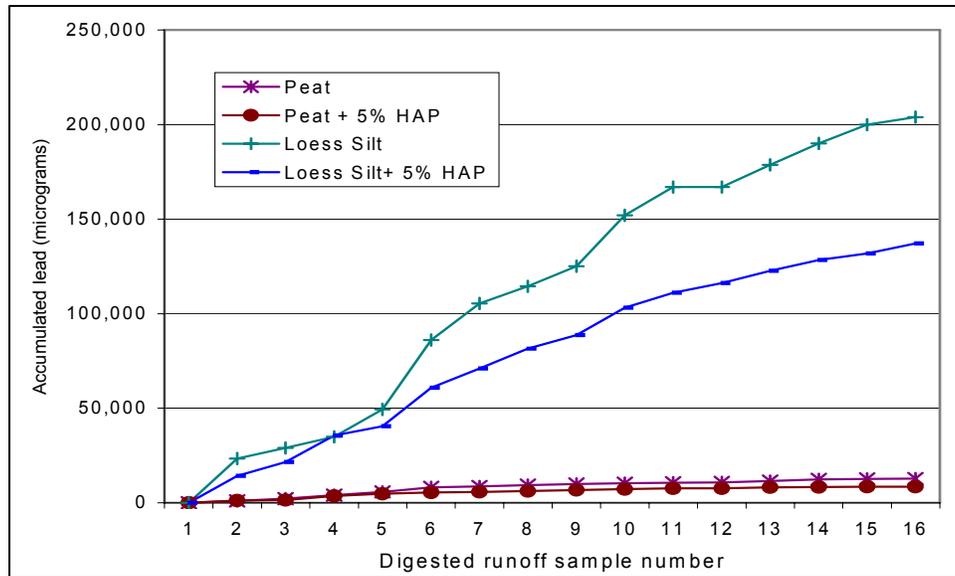


Figure 14. Accumulated total lead mass in runoff for treated and untreated lead-enriched Peat and Loess Silt lysimeters

Figure 15 illustrates the total accumulated (digested) lead mass losses in runoff from the treated and untreated Clay and Silty Sand A soils. The untreated Silty Sand A soil exhibited a pattern of steadily increasing lead mass losses in runoff and, with a few exceptions, again the trend line was almost linear. A similar pattern was observed for the untreated Clay soil although lead mass losses were much lower. These results may have serious implications for long-term lead mass losses at SAFRs resulting from runoff during rain events.

Lead mass losses in runoff were reduced in both soils after treatment with HAP, demonstrating that HAP treatment was effective in reducing lead losses in the two soils. Nonetheless, the treated Silty Sand A soil still showed a steadily increasing trend of lead loss, while lead losses from the treated Clay soil appeared to plateau near the end of the 16-week test period, indicating some lead stabilization after HAP treatment.

As shown in Figure 16, the untreated and HAP-treated Glacial Till soils tracked closely and exhibited similar, but steadily increasing, lead mass losses in runoff. Near the end of the 16-week period lead mass losses were practically the same for both the treated and untreated soils. These results show that HAP treatment was ineffective in reducing lead mass losses from the Glacial Till soil.

More lead was lost from the untreated Silty Sand B soil than the HAP-treated soil. The HAP-treated Silty Sand B soil showed a much lower but gradual increase in lead mass loss during the same period. With the exception of week 5, the lead mass loss trend line for the untreated Silty Sand B soil was almost linear while the trend line for the treated soil was nonlinear. Over the test period, HAP was effective in reducing the lead mass loss in the Silty Sand B soil.

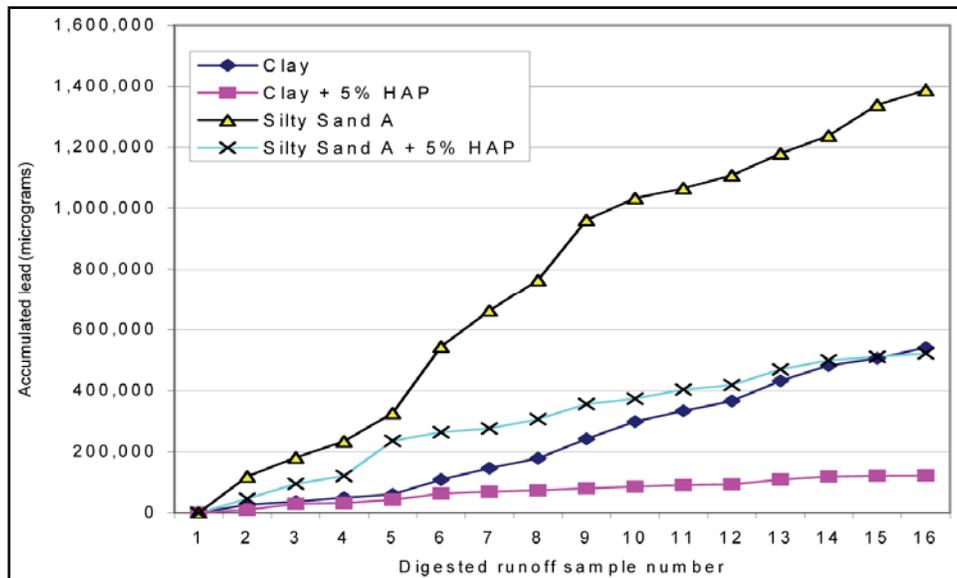


Figure 15. Accumulated total lead mass in runoff from treated and untreated lead-enriched Clay and Silty Sand A lysimeters

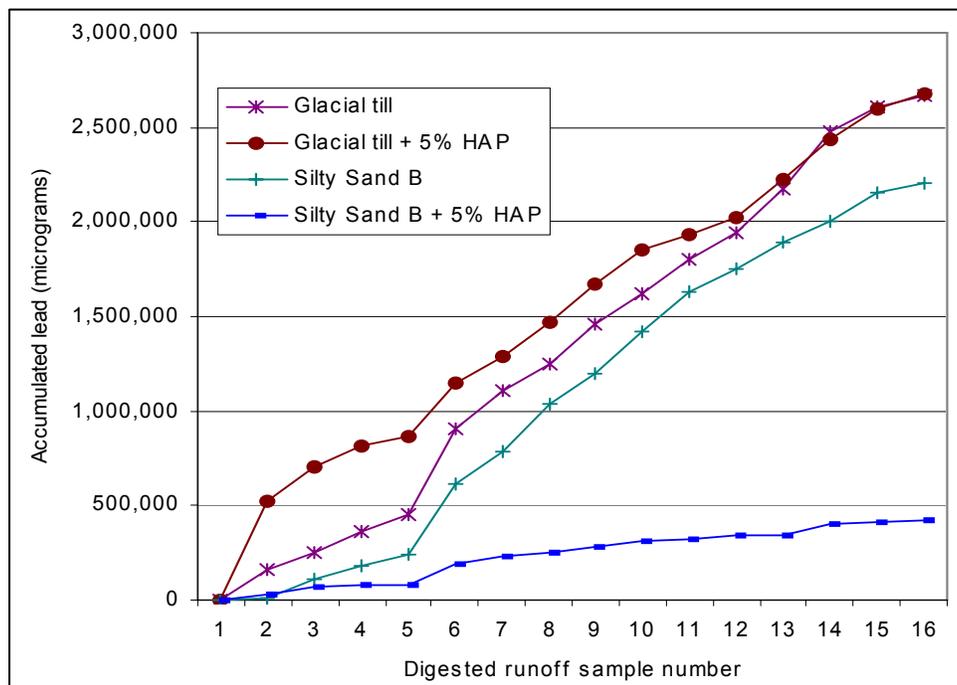


Figure 16. Accumulated total lead mass in runoff from treated and untreated lead-enriched Glacial Till and Silty Sand B lysimeters

In comparing runoff lead mass losses from the lead-enriched Clay and Silty Sand A soils without HAP amendment (Figure 15), to the corresponding leachate lead mass losses (Figure 12), the total lead mass losses from runoff was nearly 2½ and 7½ times greater, respectively, than the lead losses from leaching of the soils. In general, less than 1 percent of the lead was removed from all of the soils through leaching or runoff during the 16-week test period.

Correlation of total lead lost with sequential extraction results

The average effluent lead concentrations followed the same general pattern as was noted for the sequential extraction results. The three soils with the majority of the lead in the more available fractions also had much higher effluent concentrations. Conversely, the soils with the majority of the lead in the residual fraction had very low average lead concentrations. The logarithmic correlation between the percent of lead in the residual fraction and the average lead concentration in the digested runoff is shown in Figure 17. The coefficient of determination (r^2) was 0.94, indicating a strong correlation between the two factors. The logarithmic coefficient of determination calculated for the digested leachate was 0.45, indicating a poor correlation. The same relationship between the percent residual lead and the filtered leachate was 0.81 and the filtered runoff was 0.59, indicating moderate correlation.

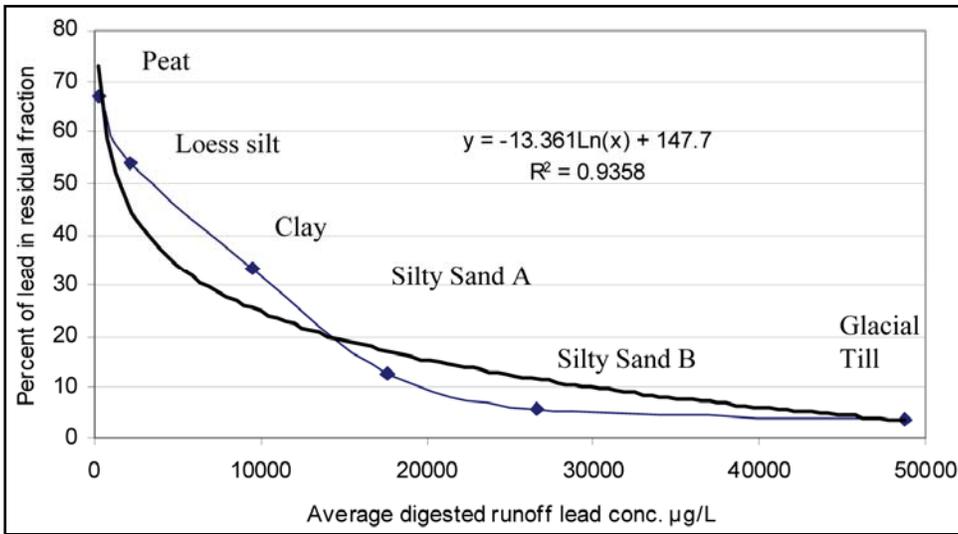


Figure 17. Correlation of average digested runoff lead concentration with percent of lead in residual fraction of the sequential extraction test

Lead partitioning

As expected, the relative concentrations of lead in the digested leachate and runoff generally followed the inverse of the partition coefficient for all of the soils except Glacial Till (Table 22). The K_d for Glacial Till was larger than those for Silty Sand A, Clay, and Silty Sand B; however, the Glacial Till lead concentrations were much higher than that for any of these soils. This result may be caused by the predominance of larger particle sizes and the small amount of Clay particles in the Glacial Till. Also, even though the K_d for the Loess soil is much higher than that of the Peat soil, the Loess Silt soil had a higher average lead concentration.

Soil	Average Digested Leachate Conc., mg/L	Average Digested runoff Conc., mg/L	Partition Coefficient K_d, L/kg
Clay	1,150	9,450	320
Silty Sand A	980	17,600	254
Glacial Till	8,680	48,800	696
Silty Sand B	423	26,600	157
Peat	102	267	2,670
Loess	261	2,110	36,000

Comparison of Other Metals Lost with Sequential Extraction Results

Copper

The soils enriched with copper from the bullets produced a calculated total concentration of approximately 6,000 mg/kg. Copper was unique in the metals analyzed as it was found predominantly in the particle size fractions of 1 mm or larger as seen in see Figures 5 and 6. Most of the copper was found in unavailable forms as shown in Table 8. About 95 percent of the copper was found in the residual and organic/sulfide fractions of the sequential extracts in 4 of the 6 soils; the percentages in these fractions in the other two soils, Silty Sand B and Silty Sand A, were around 65 percent (see Table 8). Less than 1 percent of the copper was lost to leachate and runoff during the testing.

As expected from the sequential extraction results, the three soils with the highest copper concentrations in both effluents were Silty Sand A, Glacial Till, and Silty Sand B (Table 23). These soils also contained the highest copper levels in the smaller size fractions, especially the Glacial Till. Very little copper was found in the filtered effluents, indicating very low copper solubility (Table 23).

For most of the metals, leachate concentrations were a small fraction of the runoff concentrations. HAP addition was generally effective in reducing the copper concentrations in the effluents, reducing the loss of copper in the effluents by 70 percent or more in 8 of the 24 effluents (Table 24).

Table 23
Average Effluent Copper Concentrations Over 16-Week Test Period

Soil and Treatment	Average Cu Concentration, µg/L							
	Leachate				Runoff			
	Filtered		Digested		Filtered		Digested	
	Avg	Std	Avg	Std	Avg	Std	Avg	Std
Clay	<0.6	0.53	59.8	35.7	1.34	1.16	363	236
Clay + 5 percent HAP	<0.6	0.03	14.9	7.2	<0.6	0.21	49	24
Silty Sand A	10.4	7.9	164.3	73.4	9.52	6.60	1,570	771
Silty Sand A + 5 percent HAP	0.6	0.5	18.5	7.0	3.21	2.84	538	286
Glacial Till	5.2	1.4	274.6	153.5	6.71	3.37	1,460	590
Glacial Till + 5 percent HAP	<0.6	0.5	50.8	26.6	0.82	0.61	694	469
Silty Sand B	15.3	7.0	52.3	17.6	49.2	29.26	1,360	875
Silty Sand B + 5 percent HAP	9.5	3.8	27.7	7.7	3.43	2.49	299	176
Peat	6.1	16.1	23.2	21.2	1.6	1.38	22	10
Peat + 5 percent HAP	3.9	2.4	16.9	6.2	<0.6	1.29	13	8
Loess Silt	0.8	1.4	39.2	18.9	0.84	0.78	294	207
Loess Silt + 5 percent HAP	<0.6	0.92	33.4	18.4	<0.6	0.24	256	106

Avg = Average value.
Std = Standard deviation.

Table 24
Percent Reduction in Average Copper Concentrations in Effluents

Soil and Treatment	Percent Reduction in Lead Concentration by HAP Treatment			
	Leachate		Runoff	
	Filtered	Digested	Filtered	Digested
Clay	Negligible change	75	Negligible change	86
Clay + 5% HAP				
Silty Sand A	94	89	66	66
Silty Sand A + 5% HAP				
Glacial Till	Negligible change	82	88	53
Glacial Till + 5% HAP				
Silty Sand B	38	47	93	78
Silty Sand B + 5% HAP				
Peat	35	27	Negligible change	41
Peat + 5% HAP				
Loess Silt	Negligible change	15	Negligible change	13
Loess Silt + 5% HAP				

Antimony

Antimony concentrations from most lysimeters were higher in the filtered leachates than in the filtered runoff (Table 25). The three soils (Table 9) with the highest percentage of antimony in the exchangeable and carbonate fractions, of the sequential extracts, (Silty Sand A, 29 percent; Silty Sand B, 31 percent; and Glacial Till, 63 percent) produced leachates and runoffs with the highest

antimony concentrations. Peat and Loess soils, for which no antimony was found in these fractions, lost very little antimony.

Of all the metals analyzed, antimony had the largest fraction of its losses in the filtered samples. The physical and chemical interactions of specific metals with soils such as association with inorganic and organic materials, complexation reactions with inorganic soil constituents, substitution reactions, and specific contaminant reactions sometimes referred to as chemisorption may have contributed to higher concentrations in the <0.45- μm filtered samples. Filtered samples averaged 16 percent of the digested leachates and 86 percent of the digested runoff (Table 26). Also unique to antimony, HAP addition increased the losses of antimony in all filtered leachates and runoffs, in all digested leachates, and in 5 of 6 digested runoffs, in many cases by several times. Antimony concentrations in the digested runoff were reduced slightly in 4 of the 6 soils by HAP addition. Evidently, HAP addition had an effect on antimony's solubility even though there was very little effect of HAP addition on the pH values of the samples.

The average concentration of antimony in both the filtered and digested runoff had a significant negative correlation (Figures 18 and 19) with the percent of antimony found in the residual fraction of the sequential extraction test. Higher antimony in the residual fraction resulted in lower losses to the runoff. The relation of these parameters did not hold for the antimony levels in the filtered and digested leachates due largely to the low concentration of antimony in the leachates from the Silty Sand B (which also had low percentage in the residual fraction).

Table 25
Average Effluent Antimony Concentrations Over 16-Week Test Period

Soil and Treatment	Average Sb Concentration, $\mu\text{g/L}$							
	Leachate				Runoff			
	Filtered		Digested		Filtered		Digested	
	Avg	Std	Avg	Std	Avg	Std	Avg	Std
Clay	27.1	8	47.4	28	47.2	17	174	76
Clay + 5% HAP	95.9	24	64.3	28	48.6	18	53	21
Silty Sand A	93.9	39	103.6	21	45.5	16	317	122
Silty Sand A + 5% HAP	288	140	261.6	116	151	75	176	82
Glacial Till	375	50	546.6	121	222	96	1,230	414
Glacial Till + 5% HAP	770	104	657.4	110	844	442	1,100	572
Silty Sand B	98.9	30	88.4	38	149	61	521	244
Silty Sand B + 5% HAP	272	92	201.2	92	171	79	204	91
Peat	<20	8	<20	9	<20	2	<20	2
Peat + 5% HAP	35.7	16	24.8	15	<20	11	<20	9
Loess Silt	42.8	6	28.7	12	36.8	14	48	20
Loess Silt + 5% HAP	67.9	25	42.7	29	79.8	21	87	38

Avg = Average value.
Std = Standard deviation.

Table 26
Comparison of Percent of Antimony in Effluents and Percent Reduction in Average Antimony Concentrations

Soil and Treatment	Filtered as % of Digested		Percent Reduction in Sb Conc. After HAP Treatment			
			Leachate		Runoff	
	Leachate	Runoff	Filtered	Digested	Filtered	Digested
Clay	57	27	-255	-1322	-3	70
Clay + 5% HAP	14	92				
Silty Sand A	8	14	-207	-156	-233	45
Silty Sand A + 5% HAP	10	86				
Glacial Till	5	18	-105	4	-279	10
Glacial Till + 5% HAP	11	77				
Silty Sand B	11	29	-176	-145	-15	61
Silty Sand B + 5% HAP	12	84				
Peat	15	191	negligible	negligible	negligible	negligible
Peat + 5% HAP	12	247				
Loess Silt	16	76	-59	-40	-117	-79
Loess Silt + 5% HAP	18	92				

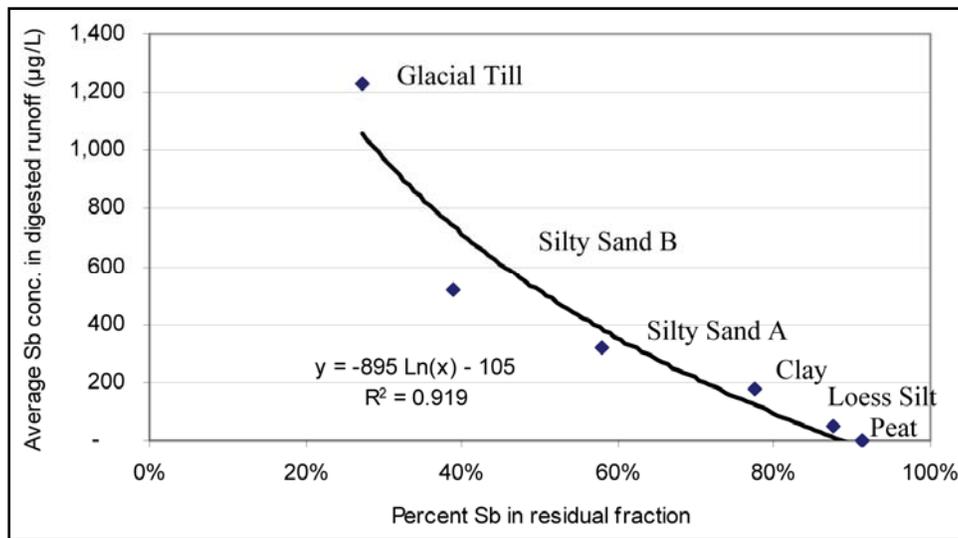


Figure 18. Plot comparing percent of antimony in sequential extraction residual fraction with average antimony concentration in digested runoff

Chromium

Over 98 percent of the chromium was found in the residual fraction of the sequential extracts from all soils except Silty Sandy A, which had 25 percent of the chromium in the Fe-Mn oxide fraction (Table 10). Chromium concentrations are shown in Table 27 and reductions in concentrations after HAP treatment in Table 28. Very low concentrations of chromium were found in both leachates and runoff from the lysimeters (Table 27).

Table 27
Average Effluent Chromium Concentrations Over 16-Week Test Period

Soil and Treatment	Average Cr Concentration, µg/L							
	Leachate				Runoff			
	Filtered		Digested		Filtered		Digested	
	Avg	Std	Avg	Std	Avg	Std	Avg	Std
Clay	<0.6	0	8.83	5	<0.6	0	43.5	27
Clay + 5% HAP	<0.6	0	1.05	1	<0.6	0	7.20	4
Silty Sand A	1.15	1	8.12	5	0.975	2	21.0	14
Silty Sand A + 5% HAP	<0.6	0	0.85	0.8	<0.6	0	9.29	6
Glacial Till	<0.6	0	5.21	3	<0.6	0	29.0	12
Glacial Till + 5% HAP	<0.6	0	1.96	2	<0.6	0	15.8	12
Silty Sand B	2.11	1	3.42	3	25.4	19	44.6	27
Silty Sand B + 5% HAP	<0.6	0	0.95	0.9	<0.6	0	9.90	5
Peat	<0.6	0	1.23	2	<0.6	0	5.30	2
Peat + 5% HAP	<0.6	0	<0.6	1	<0.6	0	1.50	2
Loess Silt	<0.6	0	5.13	3	<0.6	0	29.9	12
Loess Silt + 5% HAP	<0.6	0	3.42	3	<0.6	0	25.7	24

Avg = Average value.
Std = Standard deviation.

The filtered and digested leachate and runoff chromium concentrations were either low or below the method detection limit. In general, the same patterns seen in the lead results were apparent with chromium in that the majority of the chromium was found in the particulate (digested) fractions. After HAP treatment, reductions in chromium in the digested effluents ranged from 14 to 89 percent (Table 28). Again, a very small fraction of the total chromium in the soils was lost during the test period.

Nickel

A substantial portion of the nickel in the soils (14 to 78 percent) was found in the exchangeable plus carbonate sequential extraction fractions (Table 11). HAP addition had a negligible effect on the nickel concentrations from the various soils, especially the digested leachates and filtered runoff (Table 29). Nickel concentrations for the filtered leachate samples were inadvertently not determined. Although no data are reported for these samples, the pattern of much lower metal concentrations in the filtered runoff compared to the digested runoff was the same as the results for the other metals except for antimony.

Table 28 Percent Reduction in Average Chromium Concentrations in Effluents				
Soil and Treatment	Percent Reduction in Chromium Concentration by HAP Treatment			
	Leachate		Runoff	
	Filtered	Digested	Filtered	Digested
Clay	Negligible change	88	Negligible change	83
Clay + 5% HAP				
Silty Sand A	Negligible change	89	Negligible change	56
Silty Sand A + 5% HAP				
Glacial Till	Negligible change	62	Negligible change	46
Glacial Till + 5% HAP				
Silty Sand B	Negligible change	72	Negligible change	78
Silty Sand B + 5% HAP				
Peat	Negligible change	Negligible change	Negligible change	72
Peat + 5% HAP				
Loess Silt	Negligible change	33	Negligible change	14
Loess Silt + 5% HAP				

Table 29 Average Effluent Nickel Concentrations Over 16-Week Test Period								
Soil and Treatment	Average Ni Concentration, µg/L							
	Leachate				Runoff			
	Filtered		Digested		Filtered		Digested	
	Avg	Std	Avg	Std	Avg	Std	Avg	Std
Clay	no data	no data	23.0	15.7	<1.6	0.52	79.7	45.2
Clay + 5% HAP	no data	no data	20.6	25.7	<1.6	0.39	18.5	8.2
Silty Sand A	no data	no data	12.1	8.3	<1.6	0.61	18.9	11.7
Silty Sand A + 5% HAP	no data	no data	11.5	11.2	<1.6	1.01	15.2	7.9
Glacial Till	no data	no data	14.9	9.6	<1.6	0.24	36.9	13.2
Glacial Till + 5% HAP	no data	no data	8.38	6.0	<1.6	0.00	23.7	13.7
Silty Sand B	no data	no data	14.2	17.1	2.86	2.04	27.1	18.4
Silty Sand B + 5% HAP	no data	no data	12.7	7.6	<1.6	0.46	10.6	8.1
Peat	no data	no data	12.9	12.1	<1.6	0.60	11.5	9.6
Peat + 5% HAP	no data	no data	10.9	5.5	<1.6	0.73	6.0	3.2
Loess Silt	no data	no data	12.8	4.8	<1.6	0.42	63.1	20.7
Loess Silt + 5% HAP	no data	no data	11.9	7.4	<1.6	0.65	56.2	41.2

Avg = Average value.
Std = Standard deviation.

The addition of HAP had a lesser effect upon the average nickel concentrations, lowering the digested leachate and runoff average concentrations by 23 and 38 percent, respectively (Table 30).

Table 30
Comparison of Percent Nickel in Effluents and Percent Reduction
in Average Nickel Concentrations

Soil and Treatment	Filtered Runoff as % of Digested	Percent Reduction in Ni Conc. After HAP Treatment		
		Digested Leachate	Runoff	
			Filtered	Digested
Clay	1.1	Negligible change	Negligible change	77
Clay + 5% HAP	2.1			
Silty Sand A	1.8	5	Negligible change	20
Silty Sand A + 5% HAP	2.6			
Glacial Till	0.3	Negligible change	Negligible change	36
Glacial Till + 5% HAP	0.0			
Silty Sand B	10.5	Negligible change	Negligible change	61
Silty Sand B + 5% HAP	4.6			
Peat	5.4	16	Negligible change	48
Peat + 5% HAP	11.6			
Loess Silt	1.1	7	Negligible change	11
Loess Silt + 5% HAP	0.75			

Zinc

The highest percentages of zinc were found in the residual fraction of the sequential extraction test for all the soils (Table 12). The percentages ranged from 35, 44, and 73 percent for the Silty Sand B, Silty Sand A, and Peat soils, respectively, to over 90 percent for the Clay, Glacial Till, and Loess Silt soils. With the exceptions of the Silty Sand B and Clay-filtered leachates, the average zinc concentrations were higher in all leachate and runoff samples than in the HAP-treated samples (Table 31). As expected, most of the zinc appeared in the runoff particulate fractions. Disregarding the Silty Sand B and Clay-filtered leachates, reductions in zinc concentrations after HAP treatment ranged from 13 to 60 percent (Table 32). In most instances, the addition of HAP to the soils had a moderately reductive effect on the zinc concentrations in leachates and runoffs relative to the untreated controls.

**Table 31
Average Effluent Zinc Concentrations Over 16-Week Test Period**

Soil and Treatment	Average Zn Concentration, µg/L							
	Leachate				Runoff			
	Filtered		Digested		Filtered		Digested	
	Avg	Std	Avg	Std	Avg	Std	Avg	Std
Clay	18.5	19.3	76.9	43.0	<3	5.07	323	222
Clay + 5% HAP	21.7	16.7	38.3	30.8	<3	0.43	130	111
Silty Sand A	47.6	54.1	91.0	113.1	8.58	13.60	355	174
Silty Sand A + 5% HAP	12.7	11.7	50.9	62.0	8.23	25.92	176	583
Glacial Till	7.5	7.2	99.0	75.7	6.97	10.22	352	210
Glacial Till + 5% HAP	4.9	8.8	40.1	45.3	<3	0.68	167	102
Silty Sand B	29.9	18.2	48.8	30.4	42.1	22.40	404	216
Silty Sand B + 5% HAP	54.9	60.2	38.4	74.7	3.34	4.85	185	161
Peat	65	25.1	32.8	24.5	6.49	5.39	60	35
Peat + 5% HAP	34.4	48.5	28.5	33.4	4.21	23.93	53	91
Loess Silt	30.6	36.5	59.6	83.6	<3	6.44	233	80
Loess Silt + 5% HAP	5.65	7.9	36.0	31.2	<3	2.87	196	222

Avg = Average value.
Std = Standard deviation.

**Table 32
Percent Reduction in Average Zinc Concentrations in Effluents**

Soil and Treatment	Percent Reduction in Lead Conc. by HAP Treatment			
	Leachate		Runoff	
	Filtered	Digested	Filtered	Digested
Clay	Negligible change	50	Negligible change	60
Clay + 5% HAP				
Silty Sand A	Negligible change	44	Negligible change	Negligible change
Silty Sand A + 5% HAP				
Glacial Till	Negligible change	60	Negligible change	53
Glacial Till + 5% HAP				
Silty Sand B	Negligible change	21	Negligible change	54
Silty Sand B + 5% HAP				
Peat	Negligible change	13	Negligible change	Negligible change
Peat + 5% HAP				
Loess Silt	Negligible change	40	Negligible change	Negligible change
Loess Silt + 5% HAP				

Iron

Although over 95 percent of the iron was found in the residual fraction for all of the soils except Silty Sandy A (refer to Table 13), relatively large concentrations of iron were also found in the digested leachates. The average iron concentrations in the effluents over the test period are shown in Table 33. On

average, HAP treatment lowered the iron concentrations in the filtered effluents by around 70 to 55 percent, but lowered the concentrations in the digested effluents by only 4 to 84 percent (Table 34).

Table 33
Average Effluent Iron Concentrations over 16-Week Test Period

Soil and Treatment	Average Fe Concentration, µg/L							
	Leachate				Runoff			
	Filtered		Digested		Filtered		Digested	
	Avg	Std	Avg	Std	Avg	Std	Avg	Std
Clay	38.5	45.8	10,080	4,770	1,420	1,470	53,370	30,900
Clay + 5% HAP	<6	3.4	1,640	1,130	64	60	8,810	4,390
Silty Sand A	715	447.9	6,000	2,150	2,750	2,550	20,600	11,630
Silty Sand A + 5% HAP	12.3	13.0	979	530	818	697	9,510	5,480
Glacial Till	18.2	15.8	4,710	2,600	248	188	22,100	9,120
Glacial Till + 5% HAP	<6	3.2	1,770	980	<6	8	12,700	8,970
Silty Sand B	1,130	700	2,670	1,490	15,870	10,700	24,300	14,480
Silty Sand B + 5% HAP	144	94.2	923	526	1,210	1,190	5,950	3,300
Peat	13.7	21.6	1,590	1,400	664	622	4,400	2,080
Peat + 5% HAP	11.2	55.6	502	419	85	114	1,590	1,500
Loess Silt	40.1	25.7	6,490	3,810	333	334	43,180	16,380
Loess Silt + 5% HAP	6.77	11.5	5,520	3,660	113	147	41,800	29,690

Avg = Average value.
Std = Standard deviation.

Table 34
Percent Reduction in Average Iron Concentrations in Effluents

Soil and Treatment	Percent Reduction in Fe Concentration by HAP Treatment			
	Leachate		Runoff	
	Filtered	Digested	Filtered	Digested
Clay	Negligible change	84	96	84
Clay + 5% HAP				
Silty Sand A	Negligible change	84	70	54
Silty Sand A + 5% HAP				
Glacial Till	Negligible change	62	Negligible change	43
Glacial Till + 5% HAP				
Silty Sand B	87%	65	92	76
Silty Sand B + 5% HAP				
Peat	Negligible change	68	Negligible change	64
Peat + 5% HAP				
Loess Silt	Negligible change	15	Negligible change	4
Loess Silt + 5% HAP				

Manganese

The distribution of manganese varied throughout the different fractions of the sequential extraction test (Table 14). Less than 2 percent of the manganese was found in the exchangeable fraction from any of the soils. Manganese concentrations in the effluents are shown in Table 35. The data do not follow typical patterns as were noted for the majority of the other metals. Effluent concentrations ranged from values at or near the method detection limit of 0.3 µg/L after blank correction for the untreated and treated samples to about 1,900 µg/L for the untreated Clay soil.

Table 35
Average Effluent Manganese Concentrations Over 16-Week Test Period

Soil and Treatment	Average Mn Concentration, µg/L							
	Leachate				Runoff			
	Filtered		Digested		Filtered		Digested	
	Avg	Std	Avg	Std	Avg	Std	Avg	Std
Clay	0.3	0	264	121	2.91	3	1,890	1251
Clay + 5% HAP	1.9	3	53	38	<0.3	0	296	158
Silty Sand A	4.8	3	112	46	3.96	2	489	265
Silty Sand A + 5% HAP	0.5	1	34	18	1.39	1	197	94
Glacial Till	56.9	138	222	136	3.38	2	595	239
Glacial Till + 5% HAP	0.3	1	59	35	0.42	0	348	259
Silty Sand B	4.5	3	37	20	8.78	4	293	188
Silty Sand B + 5% HAP	29.0	55	48	47	0.60	0	109	67
Peat	2.1	5	58	54	1.44	1	125	69
Peat + 5% HAP	153	230	180	216	2.17	8	50	47
Loess Silt	1.0	1	168	89	1.02	1	14,00	581
Loess Silt + 5% HAP	<0.3	0	149	120	<0.3	0	1,360	997

Avg = Average value.
Std = Standard deviation.

The addition of HAP also produced quite variable results for the different soils and effluents. As evidenced by the percent reduction values in Table 36, low concentration values made slight differences much more prominent.

Vanadium

As shown in Table 15, over 30 percent of Silty Sand A's vanadium was found in the Fe-Mn oxide fraction, but for all other soils over 85 percent was found in the residual sequential extraction fraction. Vanadium was found at higher concentrations in the digested runoff than in the digested leachate (Table 37). Again, the majority of the vanadium was found in the particulate fraction as seen by the differences between the filtered and digested effluent concentrations. HAP addition had a small and variable effect on vanadium concentrations.

Table 36 Percent Reduction in Average Manganese Concentrations in Effluents				
Soil and Treatment	Percent Reduction in Manganese Concentration by HAP Treatment			
	Leachate		Runoff	
	Filtered	Digested	Filtered	Digested
Clay	Negligible change	80	Negligible change	84
Clay + 5% HAP				
Silty Sand A	Negligible change	70	65	60
Silty Sand A + 5% HAP				
Glacial Till	Negligible change	74	88	42
Glacial Till + 5% HAP				
Silty Sand B	Negligible change	-30	93	63
Silty Sand B + 5% HAP				
Peat	Negligible change	Negligible change	Negligible change	60
Peat + 5% HAP				
Loess Silt	Negligible change	-13	Negligible change	3
Loess Silt + 5% HAP				

Table 37 Average Effluent Vanadium Concentrations Over 16-Week Test Period								
Soil and Treatment	Average V Concentration, µg/L							
	Leachate				Runoff			
	Filtered		Digested		Filtered		Digested	
	Avg	Std	Avg	Std	Avg	Std	Avg	Std
Clay	<0.3	0	25.7	11	1.41	1.24	86	51
Clay + 5% HAP	<0.3	0	6.7	3	1.13	1.19	17	8
Silty Sand A	1.02	2	20.8	6	1.26	1.82	54	32
Silty Sand A + 5% HAP	<0.3	1	5.2	2	0.94	0.93	26	15
Glacial Till	<0.3	0	14.2	7	0.57	0.87	60	24
Glacial Till + 5% HAP	<0.3	0	7.5	3	0.45	0.70	35	24
Silty Sand B	1.20	2	11.5	5	8.44	5.66	86	50
Silty Sand B + 5% HAP	1.50	3	9.0	3	1.38	1.25	20	11
Peat	2.12	5	7.2	3	1.71	1.37	8.7	4.5
Peat + 5% HAP	9.62	28	9.5	2	1.64	1.86	4.3	3.0
Loess Silt	0.83	2	14.1	7	1.74	1.40	74	53
Loess Silt + 5% HAP	<0.3	1	11.9	6	1.66	1.51	68	28

Avg = Average value.
Std = Standard deviation.

Reductions in concentrations after HAP treatment ranging from 8 percent to 80 percent were observed for digested runoff samples while the digested leachate from the Peat soil was negatively impacted by HAP addition (Table 38).

Table 38 Percent Reduction in Average Vanadium Concentrations in Effluents				
Soil and Treatment	Percent Reduction in V Concentration by HAP Treatment			
	Leachate		Runoff	
	Filtered	Digested	Filtered	Digested
Clay	Negligible change	74	Negligible change	80
Clay + 5% HAP				
Silty Sand A	Negligible change	75	Negligible change	51
Silty Sand A + 5% HAP				
Glacial Till	Negligible change	47	Negligible change	41
Glacial Till + 5% HAP				
Silty Sand B	Negligible change	22	84	76
Silty Sand B + 5% HAP				
Peat	Negligible change	-32	Negligible change	51
Peat + 5% HAP				
Loess Silt	84	16	5	8
Loess Silt + 5% HAP				

TCLP

The soils were subjected to the TCLP before and after lead enrichment and after the completion of the LLP. TCLP results were compared to other test results in the study.

Clean soils

A summary of the results for the clean soils is shown in Table 39. Lead levels from all six TCLP leachates from the unenriched soil samples were below the 5.0-mg/L action level for lead. For comparison, the target goal for the UTS of 0.75 mg/L (ITRC 2003) was also considered. Of the six soils used to prepare lead-enriched soils for the current study, only one (Silty Sand A) produced a TCLP leachate containing lead at a concentration that exceeded 0.75 mg/L.

Untreated lead-enriched control and lead-enriched HAP-treated soils before LLP testing

The average TCLP results from the six untreated lead-enriched control soils and the six lead-enriched HAP-amended soils for eight metals of interest before LLP testing are listed in Table 40. Individual results from triplicate analyses are listed in Appendix A, Table A9. For the untreated soils, the untreated Clay soil had the highest TCLP lead concentration (705 mg/L) and the lowest (20.6 mg/L) was from the Peat soil. HAP addition lowered the TCLP lead concentrations in all soils. With the exception of the Clay and Glacial Till soils, lead

concentrations in the TCLP leachates were lowered to below the 5-mg/L action level immediately following HAP amendment.

Table 39
TCLP Results for the Six Clean Soils

Metal	Results of TCLP on Soils Screened over a 26.5-mm Sieve, mg/L					
	Clay	Silty Sandy A	Glacial Till	Silty Sandy B	Peat	Loess Silt
Pb	0.012	2.26	0.02	0.012	0.018	0.01
Cr	0.01	0.01	0.01	0.01	0.01	0.01
Cu	0.01	0.027	0.01	0.01	0.01	0.01
Ni	0.03	0.01	0.01	0.01	0.01	0.01
Zn	0.10	0.10	0.10	0.10	0.10	0.10
Fe	0.339	0.298	0.271	0.292	0.593	0.640
Mn	0.156	0.143	0.492	0.272	0.074	0.084
Mo	0.01	0.01	0.01	0.01	0.01	0.01
V	0.011	0.01	0.01	0.01	0.012	0.012
Sb	0.03	0.03	0.03	0.03	0.03	0.03

Table 40
Average TCLP Metal Concentrations of Untreated, Lead-Enriched Control Soils and Lead-Enriched, HAP-Treated Soils Before LLP Testing

Metal	TCLP Metal Concentrations, mg/L					
	Clay		Silty Sand A		Glacial Till	
	Control	5% HAP	Control	5% HAP	Control	5% HAP
Pb	705	10.3	371	1.6	528	324
Cr	0.01	0.01	0.01	0.01	0.01	0.01
Cu	0.97	0.49	8.23	1.17	6.49	3.24
Ni	0.061	0.01	0.01	0.01	0.01	0.01
Zn	0.70	0.07	1.22	0.097	0.92	0.663
Fe	0.48	0.03	0.03	0.03	0.37	3.61
Mn	11.3	7.32	2.18	1.07	1.14	1.95
Sb	0.52	0.83	1.36	0.66	2.24	1.58
	Silty Sand B		Peat		Loess Silt	
	Control	5% HAP	Control	5% HAP	Control	5% HAP
Pb	337	3.7	20.6	0.29	131	0.36
Cr	0.01	0.01	0.01	0.01	0.01	0.01
Cu	3.33	0.54	0.211	0.112	1.4	0.17
Ni	0.01	0.01	0.01	0.01	0.01	0.01
Zn	0.85	0.065	0.18	0.01	0.20	0.01
Fe	2.3	0.03	0.03	0.03	0.03	0.03
Mn	2.26	1.04	1.58	1.25	2.68	0.57
Sb	2.05	1.48	0.17	0.08	0.37	0.114

Untreated, lead-enriched control and lead-enriched, HAP-treated soils after LLP testing

The TCLP results from the six untreated lead-enriched control soils and the six lead-enriched HAP-amended soils after LLP testing are shown in Table 41.

Table 41 Average TCLP Metal Concentrations of Untreated, Lead-Enriched Control Soils and Lead-Enriched, HAP-Treated Soils After LLP Testing						
Metal	TCLP Metal Concentrations, mg/L					
	Clay		Silty Sand A		Glacial Till	
	Control	5% HAP	Control	5% HAP	Control	5% HAP
Pb	481	30.5	535.1	1.2	923.6	134.4
Cr	0.070	0.001	0.001	0.002	0.001	0.001
Cu	1.53	0.410	11.3	1.21	7.83	3.05
Ni	0.053	0.0245	<0.0016	0.0218	0.001	0.0228
Zn	0.51	0.103	1.21	0.100	1.14	0.648
Fe	1.35	<0.006	4.23	0.0196	0.35	0.663
Mn	9.17	3.91	2.673	0.891	1.01	1.53
Sb	0.391	0.684	1.12	0.421	1.836	1.101
Metal	Silty Sand B		Peat		Loess Silt	
	Control	5% HAP	Control	5% HAP	Control	5% HAP
	Pb	489.5	6.9	54	0.22	159.2
Cr	3.58	0.002	0.001	0.001	0.001	0.001
Cu	3.83	1.241	0.14	0.086	0.94	0.06
Ni	0.89	0.0129	<0.0016	0.0108	<0.0016	0.0228
Zn	0.71	0.146	0.21	0.0168	0.22	0.01
Fe	0.89	0.007	<0.006	0.0136	<0.0016	<0.0016
Mn	1.848	0.620	1.62	0.257	2.38	0.703
Sb	2.048	0.909	0.20	0.0793	0.57	0.235

The percent reduction in TCLP lead for the six HAP-treated lead-enriched soils before and after LLP testing is shown in Figure 19. The Hamilton Air Force Base (HAFB) soil that was used in a previous study (Larson et al. 2004) is shown here for comparative purposes only. With the exception of the Glacial Till soil, after HAP treatment and before the LLP, all soils showed reductions of over 98 percent in TCLP leachate lead concentrations. Before the LLP, the reduction of the lead concentration in the TCLP leachate from the Glacial Till soil was only 38.6 percent. The same comparison was made for untreated and treated soil pairs following the LLP. Among the post-LLP soils, the percent reduction in TCLP leachate lead concentration of the HAP-treated soil compared to the untreated soil was lower than the percent reduction observed for the treated and untreated, pre-LLP soils for only one of the soils (Clay). The percent reduction in the TCLP leachate lead concentration of the HAP-treated soil compared to the untreated soil following LLP was either greater or unchanged for five of the soils (Silty Sand A, Silty Sand B, Glacial Till, Peat, and Loess Silt) when compared to the percent reduction for treated and untreated soils prior to LLP.

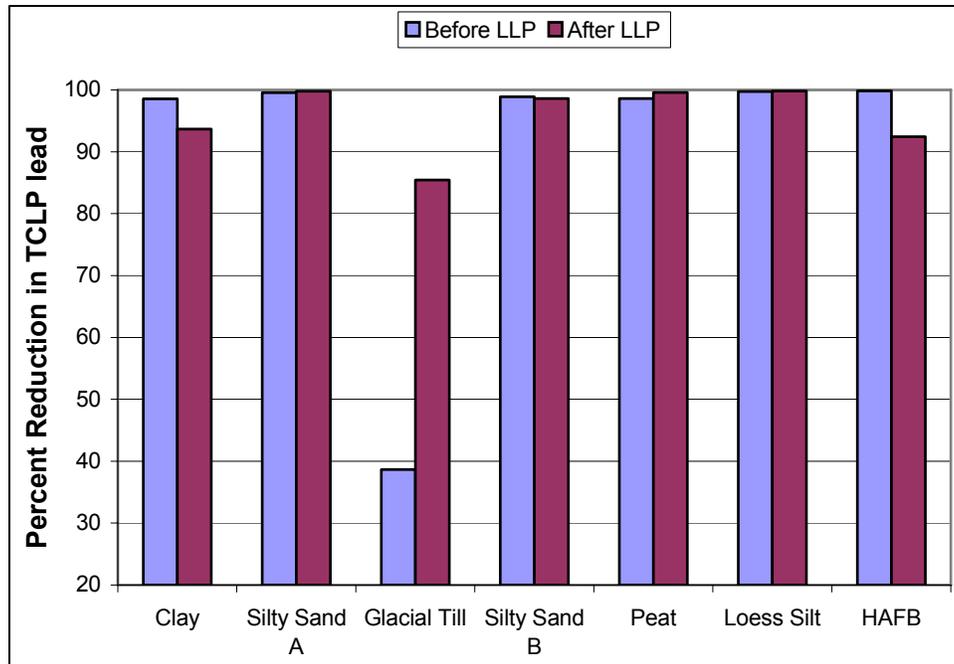


Figure 19. Percent reduction in TCLP lead by soil type before and after LLP

Table 42 shows the trend in the concentration of lead in the TCLP leachates for the untreated, lead-enriched control and the 5-percent HAP-treated soils for the six soils in the present study along with the HAFB findings from Larson et al. (2004) before and after LLP. The table also includes the pass/fail results for the TCLP lead concentration for the soils treated with HAP before and after subjecting the soils to the LLP. Two different standards were used to evaluate the results: the 5-mg/L RCRA standard and the more stringent 0.75-mg/L UTS.

Soil and Treatment	TCLP Trend Pre/Post LLP		RCRA Standard (5 mg/L)		UTS Standard (0.75 mg/L)	
	Untreated Control	5% HAP-Treated	Before LLP	After LLP	Before LLP	After LLP
Clay + 5% HAP	Decrease	Increase	fail	fail	fail	fail
Silty Sand A + 5% HAP	Increase	Negligible change	pass	pass	fail	fail
Glacial Till + 5% HAP	Increase	Decrease	fail	fail	fail	fail
Silty Sand B + 5% HAP	Increase	Increase	pass	fail	fail	fail
Peat + 5% HAP	Increase	Negligible change	pass	pass	pass	pass
Loess Silt + 5% HAP	Increase	Negligible change	pass	pass	pass	pass
HAFB + 5% HAP	Decrease	Increase	pass	fail	fail	fail

For the untreated control soils, the processes of removal of TCLP-leachable lead by runoff and leaching could be expected to contribute to a decrease in the amount of TCLP-leachable lead following LLP, and the process of corrosion and weathering could be expected to contribute to an increase the amount of TCLP-

leachable lead following LLP. These two processes combined to produce soils following the LLP that generated TCLP leachate solutions with either an increased or decreased lead concentration when compared to leachates from the soil prior to LLP. Two of the soils listed in Table 42 (Clay and HAFB) showed a decrease in the amount of lead in the TCLP leachate for the post-LLP soil compared to the pre-LLP soil. In these two soils the process of removal of TCLP-leachable lead by runoff and leaching can be thought to be dominant over the process of production of TCLP lead by weathering and corrosion. Five of the soils listed in Table 42 (Sandy Soil A, Glacial Till, Sandy Soil B, Peat, and Loess Silt) showed an increase in the amount of lead in the TCLP leachate for the post-LLP soil compared to the pre-LLP soil. In these five soils the process of removal of TCLP-leachable lead by runoff and leaching can be thought to be small relative to the process of production of TCLP lead by weathering and corrosion.

For the 5-percent HAP-treated soils, three of the post-LLP soils (Clay, Silty Sand B, and HAFB) produced TCLP leachate solutions with higher lead concentrations than that produced by the pre-LLP soil. Four of the 5-percent HAP-treated soils produced TCLP leachate solutions with lower lead concentrations than that produced by the pre-LLP soil (Silty Sand A, Glacial Till, Peat, and Loess). For the treated soils, the two processes discussed above still contribute to the trend in the relative amount of TCLP-leachable lead for pre-and post-LLP soils. A third process associated with the potential reduction in the amount of TCLP-leachable lead (chemical stabilization) is also expected. This process involves the reaction between the HAP and ionic lead produced during corrosion and weathering of the system lead over the 16 weeks of the LLP. The observed results suggest that the chemical stabilization process has reduced effectiveness for the reduction of TCLP-leachable lead during aging in certain soil types.

The RCRA legislation of 1976 for the 5-mg/L TCLP lead concentration criterion for classification as non-hazardous waste also states that the non-hazardous classification for the waste must be irreversible over time. For the treatment applied to the soils in the current study to be classified as successful, the soil treatment process must demonstrate permanency over time. Three of the soils (Silty Sand A, Peat, and Loess Silt) in Table 42 passed the RCRA standard both before and after the LLP. Two of the soils (Clay and Glacial Till) failed the standard both before and after the LLP. Two the soils (Silty Sand B and HAFB) initially passed the standard prior to the LLP but failed it after the LLP. Only two of the seven soils in Table 42 (Peat and Loess Silt) passed the UTS criterion for both the pre-LLP and post-LLP process, whereas the remaining five soils failed the goal both before and after the LLP.

There appears to be a soil dependence on the permanence of the stabilization process. For the Silty Sand B soil in this study and the HAFB soil (Larson et al. 2004), the LLP process resulted in the production of treated soils for which the TCLP lead concentration initially passed the TCLP test and then failed after 4 months and 16 artificial rain cycles (equivalent to about 46.7 cm rainfall, annually).

Phosphate Loss in Lysimeters with HAP Amendments

Prior to placement in the lysimeters, the soils were amended by adding a total of 5,500 g HAP, equivalent to 3,122 g phosphate, to 110 kg of the soils for a concentration of 50 g HAP/kg soil or 28.4 g phosphate/kg soil on a dry weight basis. The average phosphate concentrations in the leachates and runoffs from each phosphate treated soil are shown in Table 43. Phosphate concentrations were highest in the Peat and Silty Sand A leachates and less than 1 mg/L in the remaining leachates and runoffs. As shown in Table 44, phosphate losses from the HAP-treated soils over the 16-week test period were negligible, averaging less than 0.08 percent. The major contributing factor to very low phosphate losses is the very low solubility of HAP ($K_{sp} = 1 \times 10^{-36}$ at 25 °C) in pH 7 water. Phosphate losses would have been much higher if water-soluble phosphate compounds had been used; however, these types of compounds negatively impact water sources and contribute to eutrophication of lakes and streams (Larson et al. 2004).

Soil and Treatment	Leachate, mg/L	Runoff, mg/L
Clay + 5% HAP	0.13	0.48
Silty Sand A + 5% HAP	0.22	0.96
Glacial Till + 5% HAP	0.16	0.20
Silty Sand B + 5% HAP	5.9	0.60
Peat + 5% HAP	21.3	0.85
Loess Silt + 5% HAP	0.1	0.18

Soil and Treatment	Phosphate in Leachates, g	Phosphate in Runoffs, g	Total Phosphate, g	Percent Phosphate Remaining in Lysimeters
Clay + 5% HAP	0.021	0.040	0.061	99.998
Silty Sand A + 5% HAP	0.041	0.076	0.117	99.996
Glacial Till + 5% HAP	0.026	0.017	0.042	99.999
Silty Sand B + 5% HAP	1.041	0.040	1.081	99.965
Peat + 5% HAP	3.936	0.060	3.996	99.872
Loess Silt + 5% HAP	0.014	0.016	0.031	99.999

4 Conclusions and Recommendations

The efficacy of many treatment systems is typically judged by analysis of filtered and unfiltered leachate samples taken from the site and offsite tests conducted on materials removed from the site. This protocol is prescribed by the TCLP in assessing the potential for loss of toxic components from test materials. The test material is mixed with a mildly acidic solution for a period of time, and the material is then filtered to remove particulates. The filtrate is analyzed for specific components. The TCLP was originally designed for measuring the potential for migration of toxic materials from landfills through dissolution in mildly acidic landfill leachate. The TCLP has also been used as a surrogate test for measuring the success and permanence of treatment processes in containing toxic materials in contaminated soils. Several other leaching tests also filter the tested mixture before analysis. Any materials lost in or sorbed to particulate solids are not measured by any of these test methods. The TCLP approach is based on the assumption that dissolved compounds are much more bioavailable than those associated with the soil matrix.

In this study, an LLP using large berm lysimeters was designed to measure all modes of loss of materials from an SAFR site including leachates passing through the test soil and runoff from the surface. In addition, analysis of the metal content of the leachates and runoffs, both before filtration and after filtration of the samples, allowed for determination of the relative amounts of metals lost in solution as well as the total amount lost, including that which is sorbed to particulate materials.

This procedure has shown that, for most metals and soil types, the great majority of the metals were lost in the particulate fraction of both leachates and runoff from the lysimeters. This finding was true for all of the metals in this study. This finding complicates the field determination of metal migration from contaminated sites, as simple leaching tests using filtered samples do not determine losses in the particulates. Actual runoff volumes and composition are difficult to determine at SAFRs due to surface modifications and vegetation. The soils used in this study, like most SAFR berm soils, were not vegetated, thus permitting collection of maximum runoff volumes.

The effectiveness of HAP to lower lead concentrations in the effluents varied widely between the different soil types: from a 11-percent reduction in the digested leachate from the Loess soil to over 90-percent reduction in digested

leachates from Clay and Silty Sandy A soils. Percent reductions in the digested runoff varied from 6 percent for the lysimeter containing Loess Silt soil to 84- and 63-percent reduction for the Clay and Silty Sandy A lysimeters, respectively.

As would also be expected, the effects of HAP addition varied greatly between different metals. For each soil type for leachate and runoff samples, HAP treatment reduced lead and iron concentrations in all effluents; had varying effects on copper, chromium, nickel, zinc, manganese, and vanadium concentrations; and in almost all effluents actually increased the losses of antimony by several fold. HAP addition also did not affect metal concentrations in a consistent manner: it appeared to be most effective in reducing losses from the Clay, Silty Sandy A, and Glacial Till soils and less so in the Peat and Loess Silt soils. A part of the effectiveness of HAP addition may result from it reducing the TSS in both the leachates and runoffs. TSS reductions were highest in the leachates and runoff from the Clay soil: 82 and 79 percent respectively. HAP increased the TSS losses from the Loess Silt soils. All others had reduced TSS levels roughly between 40 and 60 percent of the control levels.

With the exception of the digested leachate sample from the Glacial Till soil, the filtered leachate, the digested leachate, and the filtered runoff samples from the lysimeters both before and after HAP treatment had lead concentrations below the 5-mg/L RCRA standard. Very different results were observed for the unfiltered, digested runoffs from the lysimeters. Before HAP treatment, only the digested runoffs from the Peat and Loess Silt soils passed the standard. Before HAP treatment, digested runoff from the Clay, Silty Sand A, Glacial Till, and Silty Sand B soils exceeded the standard by 1.8, 3.52, 9.7, and 5.3 times, respectively. HAP amendment was ineffective in reducing lead in the digested runoffs to below the 5-mg/L action level in the Silty Sand A, Glacial Till, and Silty Sand B soils. Only the Clay soil passed the 5.0-mg/L criterion after HAP treatment.

The effectiveness of the 5-percent HAP treatment for the stabilization of lead can be seen in terms of reducing the lead present in the leachates and runoffs collected during the LLP. The TCLP was used to evaluate the permanency of the soil treatment process. There appears to be a soil dependence on the permanency of the stabilization process. For the Silty Sand B soil in this study and the HAFB soil in an earlier study (Larson et al. 2004), the TCLP lead concentrations initially passed the 5.0-mg/kg TCLP test but failed the test after the soils were subjected to the LLP for 4 months and 16 artificial rain cycles (equivalent to about 46.6 cm/year of rainfall).

The sequential extraction results showed that fine-grained soils, such as clays and silts, tend to tightly bind lead and antimony and would be expected to produce less contaminated leachate. In contrast, the granular soils, sands and gravels, demonstrated weaker bonds with the heavy metals and would be expected to pose a greater risk of dispersion of contaminated leachate. However, granular soils with significant amounts of fine particles, such as the Glacial Till, tended to produce stronger bonds with the heavy metals and reduced leachate quantities. Thus, the soil type and its characteristics directly influenced the quantity of leachate and the bonding of soil with heavy metals. The proportion of metals in the different fractions of the sequential extraction procedure may serve

as a moderate predictor of the potential of the soils to lose metals in leachates and runoffs. All of the soils loaded into the lysimeters had bullets fired into them resulting in lead and copper concentrations in the soil of 6,600 and 10,000 mg/kg, respectively. By examining lead concentrations in a number of soil particle-size ranges, an assessment of the results of bullet impacts on the soils was made. The relative metal mass in the < 1.0-mm size fraction gave an indication of the shattering and disintegration of the bullets on impact. The results of this analysis and the amount of lead and antimony lost in digested control effluents are compared in Table 45.

Table 45 Comparison of Percent Lead and Antimony in the <1.0 mm-Soil Fractions with the Total Amount of Lead and Antimony Lost in Effluents from Lead-Enriched Control Soils					
Control Soil	Percent of lead in <1.0-mm fraction	Total Lead in Digested Effluents, mg		Total Antimony in Digested Effluents, mg	
		Leachate	Runoff	Leachate	Runoff
Glacial Till	107	1,810	2,660	1,490	67
Clay	86	222	540	9.1	10
Silty Sand A	59	179	1,390	217	25
Silty Sand B	59	69	2,200	146	43
Peat	39	21	13	24	0.07
Loess Silt	19	40	137	42	3.1

The amount of lead lost from the six soils in both digested leachates and digested runoff followed nearly the same pattern as the percent of lead in the <1.0-mm fraction: Glacial Till, Clay, Silty Sandy A, Silty Sand B, Loess Silt, and Peat. The only discrepancies were the lower Clay values in the digested runoff, and the reversal of Peat and Loess Silt, which lost very little metal.

This same general pattern holds for most of the digested control effluents as the bulk of the metals was found in the particulate fraction. Antimony concentrations also followed this relationship. The amount of destruction of the bullets by the soil particles could be a predictor of the potential for losses of lead and antimony from the soils.

This pattern was not nearly as consistent for the total amount of metals lost in the filtered effluents. The total metal lost from the soils showed mostly an inconsistent and negative correlation with the partition coefficient (Table 46). The solubility of the metals appears to be the determining factor.

Control Soil	Partition Coefficient, K_d, L/kg	Total Lead in Filtered Leachates, mg	Total Lead in Filtered Runoff, mg
Silty Sand B	157	96	4,570
Silty Sand A	254	37	347
Clay	320	2	100
Glacial Till	696	37	347
Peat	2,670	1	105
Loess Silt	36,000	0.17	23

Specific recommendations for future research that can be drawn from this study are as follows:

- Evaluate lower and higher levels of HAP treatment to assess amendment effectiveness in reducing lead mobility in SAFR soils.
- Estimate possible infiltration and surface runoff risks at SAFR sites of interest. If runoff dominates, it might be easier and less costly to capture the runoff for treatment and disposal than to excavate and treat the soil.
- Estimate cost of excavation, capping, and other alternatives for prevention of infiltration into and runoff from SAFR sites.

References

- American Public Health Association. (1998). *Standard methods for the examination of water and wastewater*, Method 2540D. Andrew. D. Eaton, Lenore S. Clesceri, Arnold E. Greenberg, Mary Ann H. Franson, ed., 20th ed., prepared and published jointly by American Public Health Association, American Water Works Association, Water Environment Federation, Washington, DC.
- American Society for Testing and Materials. (2001). "ASTM D422, D854, and D2487," West Conshohocken, PA.
- Berti, W. R., and Cunningham, S. D. (1997). "In-place inactivation of Pb in Pb-contaminated soils," *Environ. Sci. Technol.* 31, 1359.
- Interstate Technology and Regulatory Council. (2003). "Characterization and remediation of soils at closed small arms firing ranges," Small Arms Firing Range Team, Blacksburg, VA. www.itrcweb.org/SMART-1.pdf
- Laperche, V., Logan, T. J., Gaddam, P., and Traina, S. J. (1997). "Effect of apatite amendments of plant uptake of lead from contaminated soil," *Environ. Sci. Technol.* 31, 2745.
- Larson, S. L., Tardy, B. A., Beverly, M., Hearn, A., Thompson, M., and Williams, G. (2004). "Topical application of phosphate amendments to lead-contaminated small arms firing range soils," ERDC/EL TR-04-15, U.S. Army Engineer Research and Development Center, Vicksburg, MS.
- Ma, L. Q., and Gade, R. N. (1997). "Effect of phosphate rock on sequential chemical extraction of lead in contaminated soils," *J. Environ. Qual.* 26, 788.
- Ma, Q. Y., Traina, S. J., Logan, T. J., and Ryan, J. A. (1993). "In situ lead immobilization by apatite," *Environ. Sci. Technol.* 27, 1803.
- Ma, Q. Y., Logan, T. J., and Traina, S. J. (1995). "Lead immobilization from aqueous solutions and contaminated soils using phosphate rocks," *Environ. Sci. Technol.* 29, 1118.
- Martin, T. A., and Ruby, M. V. (2004). "Review of in situ remediation technologies for lead, zinc and cadmium in soil," *Remediation*, Wiley Periodicals, Inc. www.interscience.wiley.com

- Middleton, J. (2000). "Elimination of toxic heavy metals from small caliber ammunition," SERDP report (Project PP/1057/78).
- Sims, R. (1990). "Soil remediation techniques at uncontrolled hazardous waste sites: A critical review," *J. of the Air And Waste Management Assoc.* 40, 704.
- Tardy, B. A., Bricka, R. M., and Larson, S. L. (2003). "Chemical stabilization of lead in small arms firing range soils," ERDC/EL TR-03-20, U.S. Army Engineer Research and Development Center, Vicksburg, MS.
- Tessier, A., Campbell, P. G. C., and Bisson, M. (1979). "Sequential extraction procedure for the speciation of particulate tract metals," *Anal. Chem.* 51, 844-851.
- U.S. Environmental Protection Agency. (1998). "Test methods for evaluating solid waste, physical/chemical methods," SW-846, Washington, DC.
- _____. (1999). "Understanding variation in partition coefficient, K_d , values," EPA 402-R-99-004A, Washington, DC.
http://www.earthwardconsulting.com/library/epa_kdvoll.pdf
- _____. (2001). "Best management practices for lead at outdoor shooting ranges," EPA/902-B-01-001, RCRA Compliance Branch, New York, NY.
- _____. (2002). "Lead in drinking water regulation: Public education guidance," EPA 816-R-02-010, Washington, DC.
<http://www.epa.gov/safewater/lcrmr/implement.html>
- Zhang, P., and Ryan, J. A. (2000). "Soil lead remediation: Is removal the only option?" USEPA Risk Reduction Engineering Laboratory, Cincinnati, OH.

Appendix A

Analytical Data

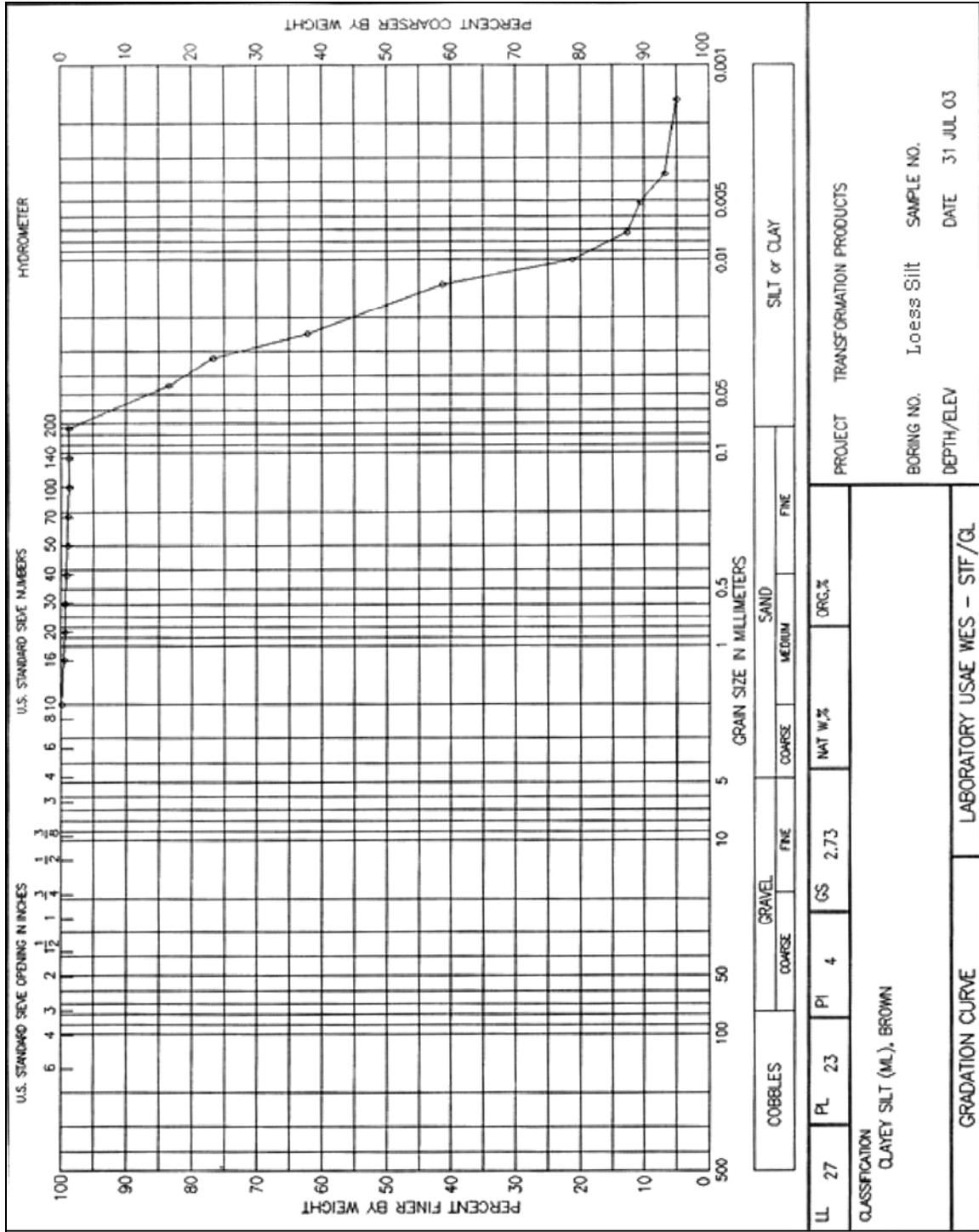


Figure A1. Grain-size distribution for Loess Silt soil

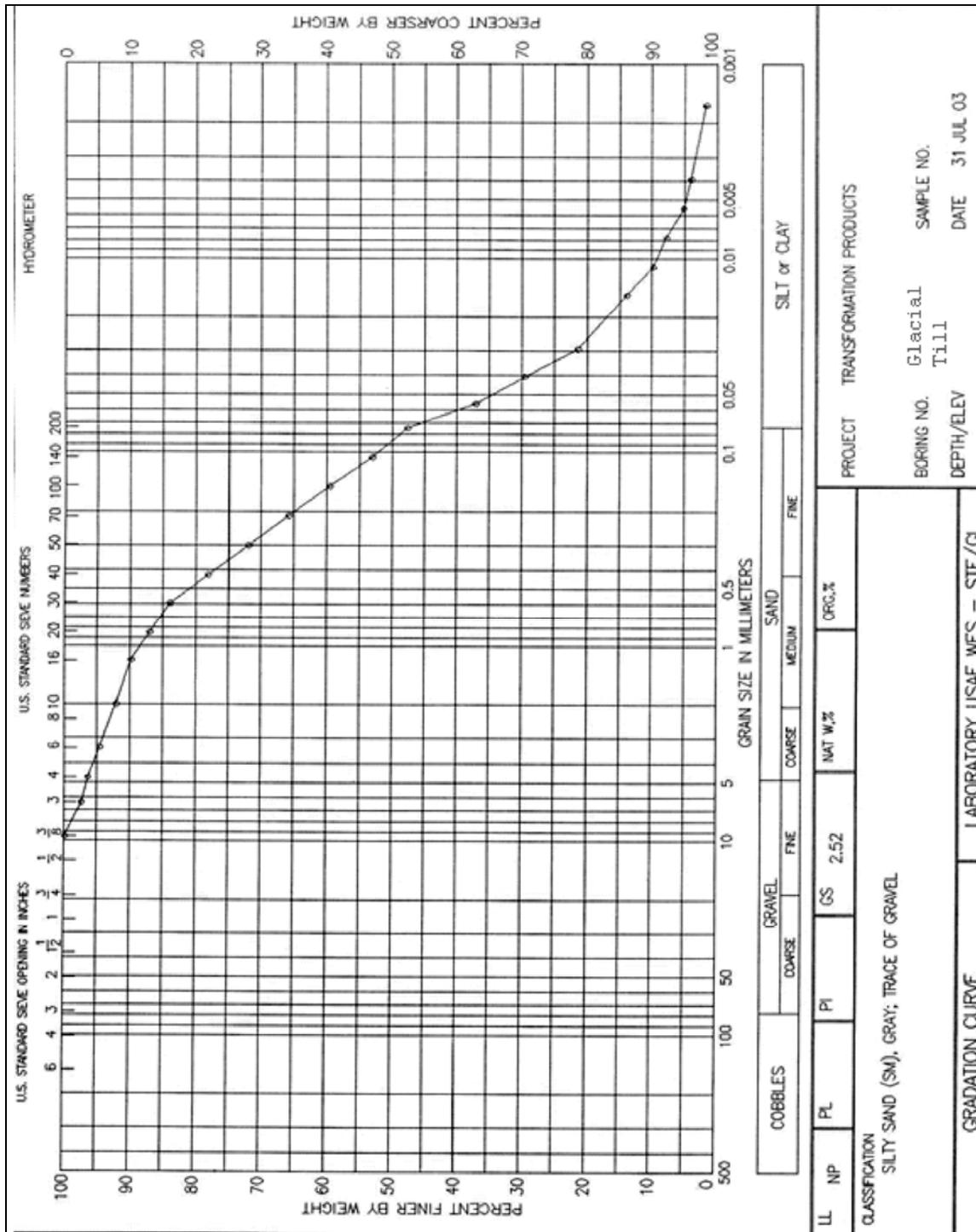


Figure A3. Grain-size distribution for Glacial Till soil

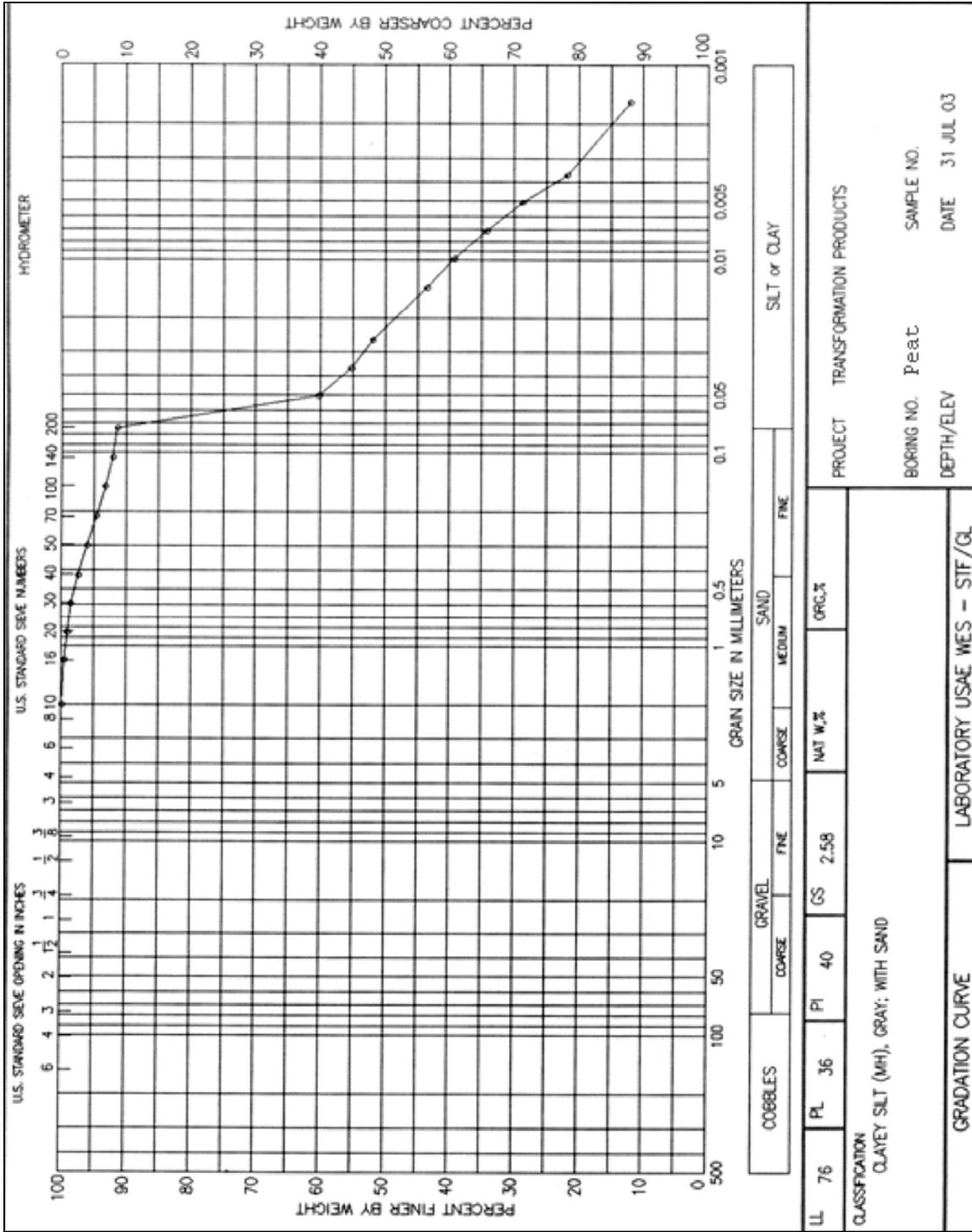


Figure A4. Grain-size distribution for Peat soil

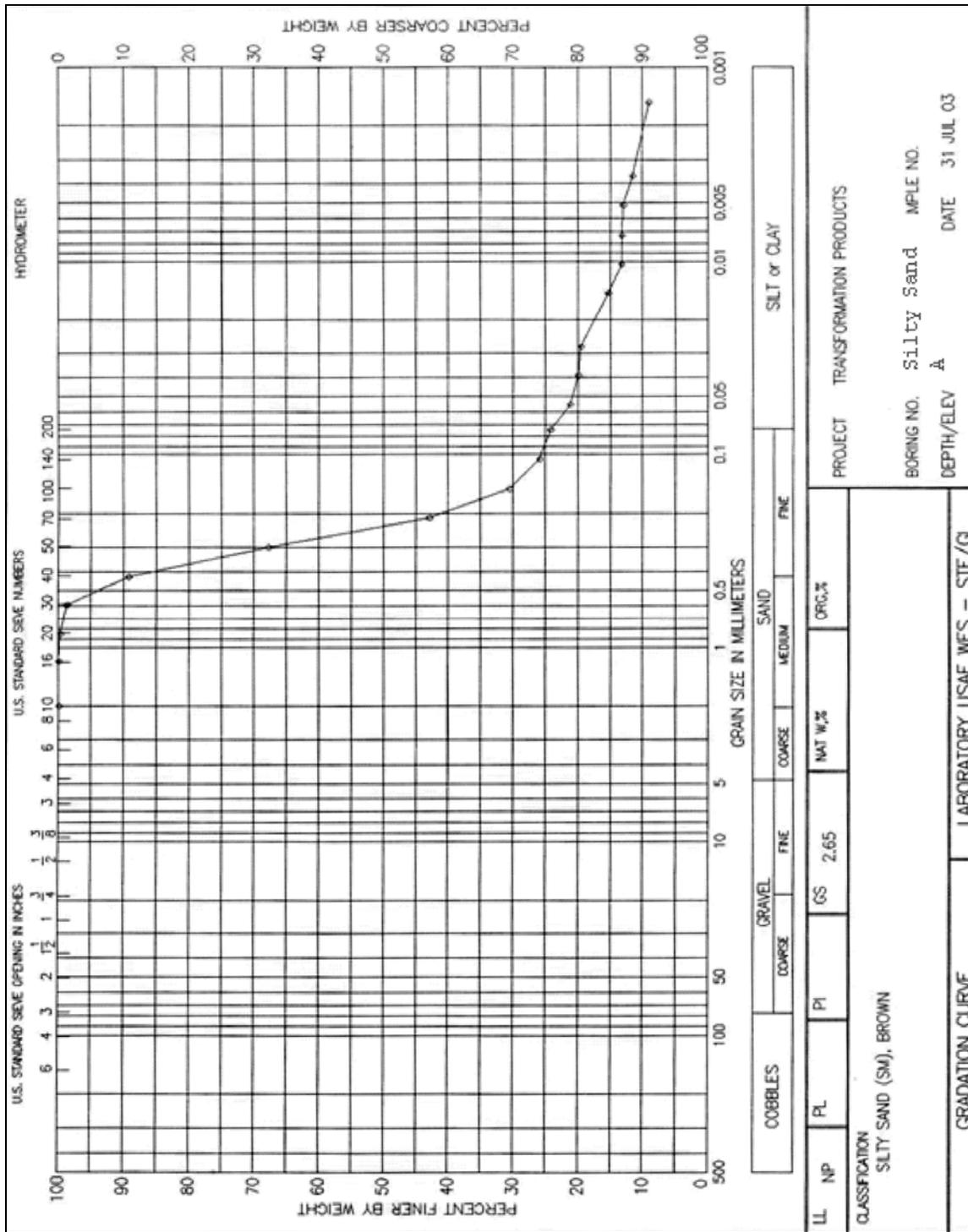


Figure A5. Grain-size distribution for Silty Sand A soil

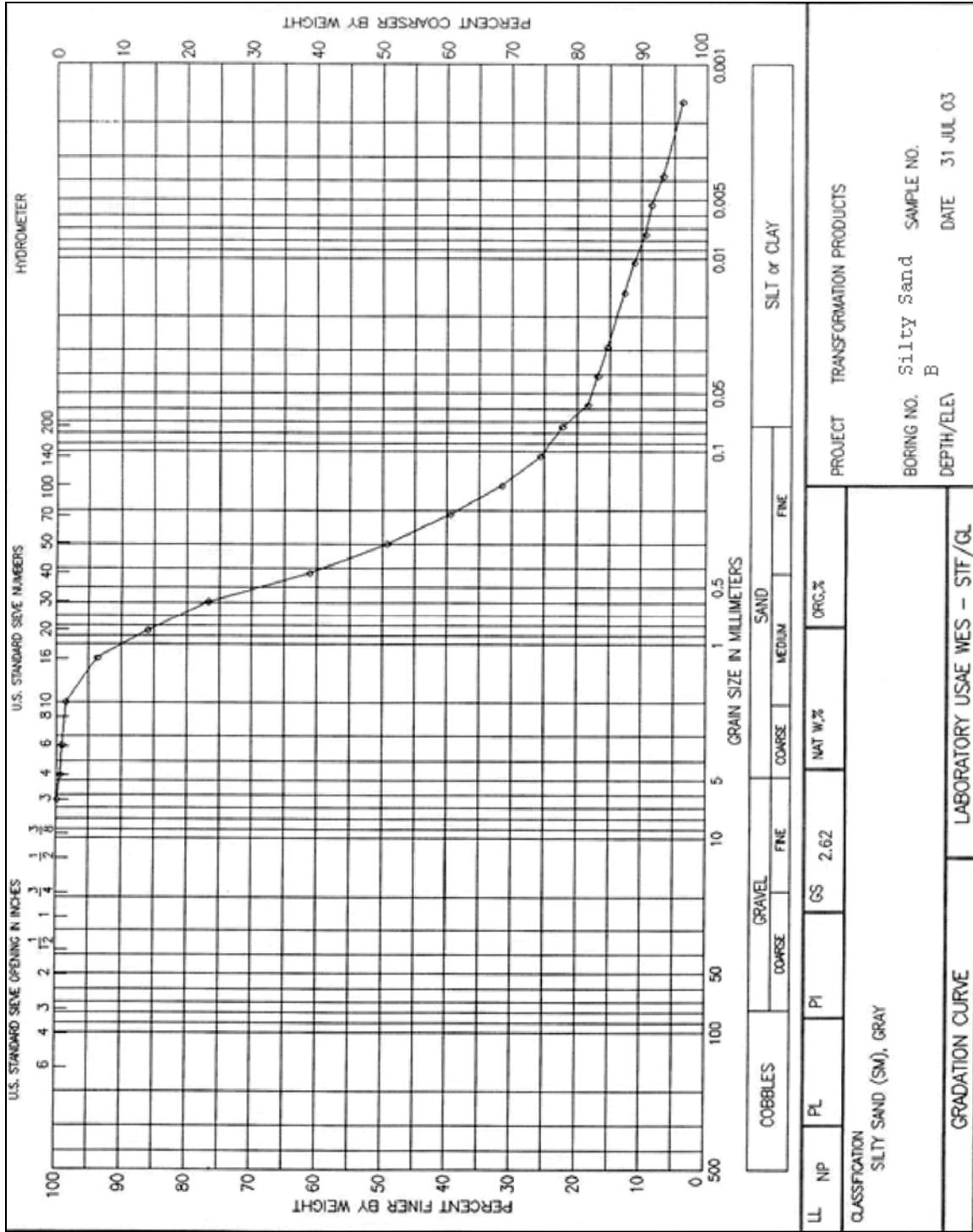


Figure A6. Grain-size distribution for Silty Sand B soil

**Table A1
Sequential Extraction Procedure**

Materials and Methods	Reagents	Conditions
Step 1 Exchangeable Pb	1 M MgCl ₂	1. Weigh 80 g of soil into a 1,000-mL centrifuge bottle
		2. Add 800 mL of 1M MgCl ₂ solution at pH 7
		3. Shake for 1 hr
		4. Centrifuge at 2,000 rpm for 30 min
		5. Filter using 0.45-μ filter
		6. Collect supernatant, label "Exchangeable"
Step 2 Lead Carbonates	1 M NaOAc	1. To residue add 800 mL of 1 M NaOAc solution at pH 5
		2. Shake for 3 hr
		3. Centrifuge at 2,000 rpm for 30 min
		4. Filter using 0.45-μ filter
		5. Collect supernatant, label "Carbonates"
Step 3 Fe-Mn Oxides	0.04 M NH ₂ OH•HCl in 25% (v/v) HOAc	1. To residue add 800 mL of 0.04 M NH ₂ OH•HCl in 25% (v/v) HOAc
		2. Heat in water bath at 95 °C for 3 hr with occasional agitation
		3. Centrifuge at 2,000 rpm for 30 min
		4. Filter using 0.45-μ filter
		5. Collect supernatant, label "Fe-Mn Oxides"
Step 4 Organic Matter and Sulfides	0.02 M HNO ₂ 30% H ₂ O ₂ 3.2 M NH ₄ OAc in 20% HNO ₃	1. To residue add 240 mL of 0.02 M HNO ₃ and 240 mL of 30% H ₂ O ₂ at pH 2
		2. Warm in water bath at 85 °C for 2 hr
		3. Add 240 mL of 30% H ₂ O ₂ at pH 2
		4. Warm in water bath at 85 °C for another 3 hr with intermittent agitation
		5. Cool and add 400 mL of 3.2 M NH ₄ OAc in 20% HNO ₃
		6. Dilute to 1600 mL and shake for 30 min
		7. Centrifuge at 2,000 rpm for 30 min
		8. Collect supernatant, label "OM and Sulfides"
Step 5 Residual	1 M HNO ₃ 1 M H ₃ PO ₄	1. Remove residue from centrifuge bottles and weigh
		2. Dry residue for 24 hr
		3. Weigh dried sample and grind for 10 min
		4. Microwave samples using method 3050B (method modified by using 8 mL of HNO ₃ and 2 mL of H ₃ PO ₄ to 2 g of dried sample)
		5. Filter samples and label "Residual"

**Table A2
Metal Concentration Results for Clay, Silty Sand A, and Glacial Till Lead-Enriched Soils,
mg/kg**

Metal	Rep 1	Rep 2	Rep 3	Rep 4	Rep 5	Rep 6	Rep 7	Rep 8	Rep 9	Avg	STD	% STD
Clay												
Pb	5,230	4,250	6,170	5,470	7,920	7,130	22,700	37,640	5,210	11,302	11,383	100.7
Cr	47.9	44.1	45.3	40.0	42.2	42.3	47.0	43.2	48.5	44.5	2.89	6.49
Cu	51.7	44.9	62.7	51.8	96.4	73.7	55.4	177	57.5	74.6	41.4	55.48
Ni	37.9	36.3	36.9	37.0	36.4	36.9	36.2	36.0	41.7	37.3	1.76	4.73
Zn	99.0	95.4	93.3	95.9	99.2	97.5	90.4	107.0	99.1	97.4	4.65	4.77
Fe	58,900	26,700	26,400	27,300	26,400	28,800	30,700	26,100	28,900	31,133	10,524	33.80
Mn	754	712	668	771	706	783	733	724	761	735	36.43	4.96
Mo	1.80	1.80	2.20	1.79	1.60	1.79	1.99	1.79	1.99	1.86	0.17	9.28
V	78.4	76.8	70.9	70.1	76.0	74.7	72.5	74.5	80.8	75.0	3.48	4.64
Sb	90.8	79.2	115	96.1	143.0	122.0	390	653	95	198.2	195.6	98.7
Silty Sand A												
Pb	8,990	4,540	5,170	7,650	5,240	4,630	9,950	4,010	3,140	5,924	2,362	39.867
Cr	10.8	11.7	11.5	12.2	12.1	10.5	11.7	21.3	10.5	12.5	3.4	27.0
Cu	204	236	266	267	436	212	301	625	245	310	136.7	44.1
Ni	6.99	7.14	5.96	6.99	6.94	6.36	6.76	6.96	5.97	6.67	0.46	6.87
Zn	33.2	35.1	37.1	40.0	56.5	33.8	40.3	70.8	34.6	42.4	12.8	30.3
Fe	5,220	5,820	5,700	5,760	5,300	5,100	5,940	5,650	5,070	5,507	333.35	6.05
Mn	83.5	89.5	85.4	91.3	86.9	75.7	91.0	90.8	79.4	85.9	5.54	6.44
Mo	1.60	1.39	1.59	1.80	1.79	1.59	1.79	3.98	1.59	1.90	0.79	41.56
V	17.2	18.3	17.9	18.4	18.8	16.3	18.5	18.7	16.7	17.9	0.92	5.12
Sb	200	107	126	182	124	110	230	97	78	139	52.03	37.36
Glacial Till												
Pb	23,800	32,900	23,900	24,700	25,900	27,000	25,400	29,100	21,400	26,011	3,367	12.94
Cr	33.1	31.1	44.9	37.2	38.6	32.6	40.4	29.3	33.9	35.7	4.99	13.98
Cu	861	1,980	1,330	1,370	4,740	976	1,320	1,820	2,550	1,883	1,192.36	63.32
Ni	29.9	29.9	27.9	28.3	28.3	28.1	27.8	29.9	28.5	28.7	0.90	3.13
Zn	129	235	163	173	527	136	162	211	289	225	124.12	55.17
Fe	21,300	21,400	21,100	21,500	21,400	20,800	20,200	20,900	20,400	21,000	463.68	2.21
Mn	349	370	348	372	366	363	353	352	353	358.4	9.32	2.60
Mo	1.40	1.59	1.99	1.39	1.39	1.40	1.40	1.60	1.60	1.53	0.20	12.98
V	57.8	55.8	59.8	61.7	53.1	60.5	55.8	57.6	57.1	57.7	2.669	4.63
Sb	453	629	451	498	509	503	56	606	459	463	165.28	35.72

Rep 1 – Rep 9 = Replicate sample number.
Avg = Average.
STD = Standard deviation.

**Table A3
Metal Concentration Results for Silty Sand B, Peat, and Loess Silt Lead-Enriched Soils,
mg/kg**

Metal	Rep 1	Rep 2	Rep 3	Rep 4	Rep 5	Rep 6	Rep 7	Rep 8	Rep 9	Avg	Std	% Std
Silty Sand B												
Pb	6,760	5,530	7,120	5,320	6,080	4,580	7,040	6,560	4,380	5,930	1,032	17.40
Cr	18.8	17.7	18.9	28.0	16.7	18.7	19.7	18.5	20.6	19.7	3.29	16.67
Cu	254	234	267	392	357	134	251	285	166	260	81.40	31.31
Ni	9.7	9.0	9.6	8.8	8.4	9.4	10.0	9.6	12.8	9.7	1.27	13.16
Zn	48.0	44.1	45.6	38.2	37.5	33.1	43.0	46.7	41.4	42.0	4.88	11.64
Fe	7,000	6,020	7,220	6,860	6,200	6,690	6,870	7,740	6,850	6,828	510.46	7.48
Mn	107	101	95	101	96	99	101	100	101	100	3.54	3.54
Mo	1.4	1.2	1.4	1.4	1.2	1.2	1.4	1.4	1.6	1.4	0.134	9.89
V	26.8	23.9	27.5	24.6	22.9	25.7	25.5	26.9	27.6	25.7	1.65	6.42
Sb	169	137	167	130	147	117	166	151	114	144	21.12	14.65
Peat												
Pb	5080	6590	2050	3152	5610	2920	2680	4100	2530	3857	1577	40.88
Cr	39.6	40.4	38.9	37.8	36.4	41.1	36.9	39.6	41.0	39.1	1.72	4.40
Cu	183.0	28.5	26.4	27.4	30.4	60.4	67.7	73.5	32.8	58.9	50.20	85.23
Ni	32.5	31.5	29.3	28.8	32.2	32.2	30.2	37.8	36.6	32.3	3.06	9.47
Zn	160	149	150	141	144	145	152	156	155	150	6.20	4.13
Fe	38200	38200	38100	36600	37200	37400	36300	37800	39600	37711	987	2.62
Mn	1,160	1,240	1,290	1,060	1,140	1,065	1,060	1,130	1,090	1,137	82.05	7.21
Mo	0.796	0.595	0.595	0.596	0.397	0.596	0.397	0.400	0.398	0.530	0.14	26.48
V	46.2	49.1	47.2	45.3	44.3	48.9	44.9	48.8	49.8	47.2	2.07	4.38
Sb	2.8	3.0	1.8	1.6	2.0	1.0	1.0	1.0	0.8	1.7	0.81	48.68
Loess Silt												
Pb	1380	1570	1450	1840	2520	1370	2770	2660	1700	1918	573	29.85
Cr	32.0	23.0	26.6	27.1	26.7	26.3	31.6	27.2	29.2	27.7	2.80	10.10
Cu	77.1	65.6	73.7	99.9	84.3	85.9	102.0	113.0	98.1	88.8	15.42	17.35
Ni	18.9	18.1	17.5	18.8	19.7	18.7	19.1	18.8	18.7	18.7	0.61	3.29
Zn	51.3	48.3	50.1	53.5	60.2	53.6	54.3	56.5	55.1	53.7	3.54	6.60
Fe	16,300	10,100	17,000	17,000	17,800	16,900	17,500	17,500	17,800	16,433	2424	14.75
Mn	412	412	424	421	434	422	430	464	445	429	16.67	3.88
Mo	4.57	1.59	1.59	1.39	1.79	1.79	1.99	1.78	1.59	2.01	0.98	48.57
V	40.3	38.3	43.1	43.2	42.4	43.7	46.8	44.0	47.3	43.2	2.82	6.53
Sb	32.8	36.2	34.6	39.0	55.8	37.5	64.1	55.7	61.3	46.3	12.61	27.22

Rep 1 – Rep 9 = Replicate sample number.
Avg = Average.
Std = Standard deviation.

Table A4 Total Mass of Metals in Lysimeters						
Metal	Metal Content of Enriched Soils, g					
	Clay	Sandy Clay	Glacial Till	Sand	Peat	Loess
Lead (Pb)	1243	651.6	2861.1	652.3	424.3	211
Chromium (Cr)	4.9	1.4	3.9	2.2	4.3	3.0
Copper (Cu)	8.2	34.1	207.1	28.6	6.5	9.8
Nickel (Ni)	4.1	0.7	3.2	1.1	3.6	2.1
Zinc (Zn)	10.7	4.7	24.8	4.6	16.5	5.9
Iron (Fe)	3421	606	2310	751	4147	1804
Manganese (Mn)	80.9	9.4	39.4	11.0	125.1	47.2
Molybdenum (Mo)	0.2	0.2	0.2	0.1	0.1	0.2
Vanadium (V)	8.3	2.0	6.3	2.8	5.2	4.8
Antimony (Sb)	21.8	15.3	50.9	15.8	0.2	5.1

Table A5 Leachate pH Results												
Week Number	Leachate pH											
	Clay		Silty Sand A		Glacial Till		Silty Sand B		Peat		Loess Silt	
	Control	5% HAP	Control	5% HAP	Control	5% HAP	Control	5% HAP	Control	5% HAP	Control	5% HAP
1	6.85	7.21	7.00	6.81	7.12	6.89	6.46	6.31	6.78	6.66	6.53	7.05
2	7.17	6.93	7.01	6.88	7.10	7.25	6.72	6.16	6.50	6.47	6.66	7.28
3	7.00	7.05	7.04	6.80	6.95	6.89	6.52	6.23	6.74	6.76	7.06	7.25
4	7.63	7.23	7.18	6.92	6.73	6.81	6.67	6.44	6.56	6.52	6.89	7.53
5	7.66	7.28	7.13	6.91	7.18	7.15	6.94	6.73	6.90	6.77	7.24	7.64
6	7.48	7.13	7.09	6.78	7.08	6.94	6.81	6.66	6.78	6.75	6.98	7.92
7	7.29	7.22	7.97	6.98	7.16	7.11	7.06	6.92	6.87	6.93	7.29	7.99
8	6.89	7.24	7.34	7.06	7.12	6.99	7.01	6.85	6.85	6.81	7.17	8.09
9	7.23	7.24	6.87	7.02	7.14	7.05	6.99	6.81	6.80	7.06	6.19	8.12
10	7.81	7.57	7.25	7.14	7.17	7.05	7.23	7.51	7.11	6.63	7.48	7.64
11	6.73	6.68	6.29	6.89	6.86	6.71	6.83	6.85	7.16	7.10	7.46	8.33
12	7.48	7.69	7.33	7.37	7.27	7.43	7.38	7.29	7.36	7.10	7.53	7.96
13	6.85	6.79	7.02	6.98	6.82	6.85	7.02	7.03	7.35	7.09	7.12	7.83
14	7.58	7.58	7.45	7.39	7.22	7.56	7.45	7.39	7.38	7.27	7.69	7.99
15	7.23	7.34	6.22	7.20	7.18	7.23	7.02	6.99	7.30	6.45	6.46	7.43
16	7.33	7.29	7.12	7.08	7.13	7.10	6.96	6.29	7.25	6.47	6.00	6.92

Control = Lead-enriched soil samples without HAP amendment.
5% HAP = Lead-enriched soil samples with 5% HAP amendment.

**Table A6
Runoff pH Results**

Week Number	Runoff pH											
	Clay		Silty Sand A		Glacial Till		Silty Sand B		Peat		Loess Silt	
	Control	5% HAP	Control	5% HAP	Control	5% HAP	Control	5% HAP	Control	5% HAP	Control	5% HAP
1	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	7.52	8.28
2	7.03	7.40	6.89	7.13	7.07	7.80	6.76	6.81	6.31	6.53	8.74	8.76
3	7.27	7.39	7.03	7.10	7.32	7.55	7.55	6.82	6.52	6.57	8.34	8.46
4	7.18	6.93	7.02	6.81	6.91	7.32	6.70	6.62	6.48	6.34	9.07	8.98
5	7.56	7.12	7.13	7.22	7.19	7.35	7.20	ns	6.98	6.97	8.16	7.88
6	7.64	7.30	7.30	7.06	7.33	7.29	7.10	7.07	6.73	6.97	7.95	8.59
7	7.37	7.36	7.00	7.10	7.22	7.13	7.34	6.87	6.76	7.16	8.08	8.59
8	7.05	7.30	7.30	7.22	7.30	7.32	7.25	6.93	7.16	6.81	8.43	8.70
9	7.24	7.32	6.64	7.14	7.14	7.26	6.74	6.80	6.49	6.79	8.28	8.32
10	7.73	7.49	6.80	7.24	7.05	7.40	7.21	6.89	6.63	6.76	8.40	8.80
11	6.67	6.34	6.50	6.53	6.50	6.83	6.60	6.63	6.63	6.75	8.51	8.75
12	7.69	7.74	6.98	7.46	7.29	7.49	7.48	7.03	7.02	7.00	8.43	9.01
13	7.11	6.65	6.63	6.74	6.58	7.18	6.77	7.13	6.65	6.28	8.55	9.10
14	7.93	7.74	7.38	7.66	7.47	7.77	7.65	6.60	7.70	7.51	8.19	8.81
15	7.61	7.11	7.06	6.20	7.14	7.24	7.05	6.12	6.16	6.37	7.93	8.29
16	7.37	7.62	7.09	7.17	6.74	7.22	5.99	6.55	6.41	6.71	7.84	6.76

ns = no sample.
Control = Lead-enriched soil samples without HAP amendment.
5% HAP = Lead-enriched soil samples with 5% HAP amendment.

**Table A7
Leachate TSS**

Week Number	TSS in Leachates, mg/L											
	Clay		Silty Sand A		Glacial Till		Silty Sand B		Peat		Loess Silt	
	Control	5% HAP	Control	5% HAP	Control	5% HAP	Control	5% HAP	Control	5% HAP	Control	5% HAP
1	85	20	0	22	15	73	2	7	78	3	209	20
2	154	98	105	32	347	171	60	25	207	84	271	480
3	194	141	174	31	316	88	37	27	174	25	127	145
4	253	81	335	13	261	105	43	38	98	96	95	256
5	291	25	84	35	139	89	58	21	47	37	144	126
6	187	9	117	38	129	48	89	46	31	6	164	95
7	254	27	112	24	222	55	40	35	42	28	131	178
8	39	24	174	59	144	68	53	28	13	24	146	85
9	421	16	38	214	225	88	69	11	41	57	93	90
10	351	33	47	232	176	112	9	11	18	16	164	123
11	274	43	90	265	158	86	25	21	6	19	83	305
12	305	6	67	9	115	18	36	18	17	9	73	162
13	432	105	83	99	341	129	36	19	41	18	84	65
14	267	27	102	6	127	47	29	14	57	30	96	65
15	232	18	127	19	82	8	54	18	40	26	75	130
16	160	21	41	9	78	28	24	4	17	14	57	117

Control = Lead-enriched soil samples without HAP amendment.
5% HAP = Lead-enriched soil samples with 5% HAP amendment.

**Table A8
Runoff TSS**

Week Number	TSS in Runoff, mg/L												
	Clay		Silty Sand A		Glacial Till		Silty Sand B		Peat		Loess Silt		
	Control	5% HAP	Control	5% HAP	Control	5% HAP	Control	5% HAP	Control	5% HAP	Control	5% HAP	
1	nd	nd	ns	nd	nd	nd	nd	nd	nd	nd	nd	27	1653
2	1734	2640	2098	1541	5765	4726	1083	1370	448	193	3997	11369	
3	1453	864	1822	738	2126	1340	2017	720	490	81	3403	6411	
4	1220	611	1086	542	2536	1105	2240	722	424	179	4881	12078	
5	2295	367	2818	1234	2081	799	1510	0	221	93	3750	2419	
6	1708	250	2574	245	778	667	3412	658	138	68	1873	5110	
7	2345	500	2402	189	727	705	1975	612	42	42	2908	3167	
8	1842	240	1361	439	770	504	1322	234	101	44	2091	2516	
9	1988	273	736	721	735	580	906	462	86	30	1877	1758	
10	1966	319	510	739	723	521	689	267	57	0	2248	6154	
11	2503	288	1935	741	1045	344	1016	200	47	15	974	3989	
12	1211	100	485	303	318	312	675	46	36	27	890	1217	
13	4322	518	1229	875	968	795	2134	460	88	16	1469	1537	
14	5620	395	653	336	906	598	2029	903	189	15	2688	4027	
15	925	114	287	356	141	318	410	593	96	15	262	1482	
16	5004	100	1078	341	508	439	1257	364	70	27	1518	1320	

nd = not determined.

Control = Lead-enriched soils without HAP amendment.

5% HAP = Lead-enriched soils with 5% HAP amendment.

**Table A9
TCLP Lead Concentration Results, mg/L**

Soil and Treatment	Rep 1	Rep 2	Rep 3	Average	Standard Deviation
Before LLP Rain Simulations					
Clay	603	835.4	676.1	705	118.8
Clay + 5% HAP	10.7	11.5	8.6	10.3	1.5
Silty Sand A	343.3	405.1	363.3	370.6	31.5
Silty Sand A + 5% HAP	2.3	1.1	1.5	1.6	0.63
Glacial Till	690.2	554.2	338.5	527.6	177.3
Glacial Till + 5% HAP	334.6	350	286.1	323.5	33.3
Silty Sand B	406	285.1	318.9	336.7	62.4
Silty Sand B + 5% HAP	3.9	2.9	4.4	3.7	0.73
Peat	38.5	14.7	8.7	20.6	15.8
Peat + 5% HAP	0.39	0.24	0.23	0.29	0.09
Loess Silt	161.5	106	126.2	131.2	28.1
Loess Silt + 5% HAP	0.75	0.20	0.14	0.36	0.34
After LLP Rain Simulations					
Clay	532.4	488.4	421.3	480.7	55.9
Clay + 5% HAP	23.1	51.8	16.5	30.5	18.8
Silty Sand A	586.5	587	431.7	535.1	89.5
Silty Sand A + 5% HAP	1.69	0.772	1.005	1.16	0.477
Glacial Till	972	964	835	924	76.57
Glacial Till + 5% HAP	132.6	134.7	136	134.4	1.7
Silty Sand B	474	584.4	410.1	489.5	88.2
Silty Sand B + 5% HAP	17	1.8	1.9	6.9	8.7
Peat	70.8	41.5	49.6	54	15.1
Peat + 5% HAP	0.237	0.143	0.269	0.216	0.066
Loess Silt	168.7	130.5	178.5	159.2	25.4
Loess Silt + 5% HAP	0.467	0.234	0.153	0.285	0.163

