



Laboratory treatability testing of soils contaminated with lead and PCBs using particle-size separation and soil washing

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Abstract

A soil treatability study was conducted using particle-size separation and soil washing to reduce the volume of material contaminated with polychlorinated biphenyls (PCBs) and lead at a Superfund site. Soil washing using surfactant was effective at removing 95% of PCBs into fine material and residual wash water. Results indicate that almost 80% of the material contaminated with up to 140 mg/kg PCBs could be treated to concentrations below 10 mg/kg using soil washing with surfactant. There did not appear to be a difference in lead removal using either particle size separation or soil washing, although the lead data have high uncertainty because of soil heterogeneity. Lead concentrations in soil were reduced from as high as 1700 to ≤ 150 mg/kg and from 560 to ≤ 220 mg/kg in about half of the material using particle size separation. © 1999 Elsevier Science B.V. All rights reserved.

Keywords: Soil; Particle-size separation; PCBs; Lead; Washing; Treatability

1. Introduction

This paper presents a case study of a treatability testing program for remediation of soils contaminated with lead and PCBs at a Superfund site. Soils at the site were contaminated from metal recycling and salvage activities including handling and stockpiling of various ferrous materials, PCB transformers, and lead acid batteries. Soil washing and particle-size separation technologies were selected as potential clean-up

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methods at the site because of their effectiveness at concentrating contaminants into a relatively small mass of fine-grained material and therefore reducing the volume of contaminated material needing further treatment or disposal. In addition, these methods can be used to treat a wide variety of contaminants which was considered important at this site containing both PCBs and lead.

The goal of this treatability study was to determine if preliminary treatment goals for site soils could be achieved from particle size separation, a simple separation of material into coarse and fine fractions, or from soil washing, a process involving high energy contact between the soil and an aqueous wash solution. The contaminant removal efficiencies and potential volume reduction of both methods was evaluated. The effectiveness of soil washing was tested using water alone, water with surfactant, and water with lowered pH. Surfactants have been found to be effective in soil washing for removing organic contaminants including PCBs, pentachlorophenol, and creosote at concentrations of 0.2 to 1.0% in wash water [1,2]. Lowering the pH of water has shown to be effective at extracting lead from soils [3–6]. The soil treatability study was conducted following EPA's guidance for conducting soil washing studies under CERCLA [7].

2. Experimental procedures

2.1. Sample preparation

A compositing scheme was devised to create three composite soil samples to represent low, average, and high concentrations of PCBs and lead within the area being investigated. First, thirteen soil samples were collected at the site. PCB immunoassay testing was conducted on six samples; those results and field observations were used to select seven samples for laboratory analysis of PCBs, lead, and grain size distribution. Based on the analytical results of the seven soil samples, the soils were composited into three samples—Samples L, A, and H. Concentrations of PCBs in the three samples ranged from 52 to 670 mg/kg and concentrations of lead ranged from 560 to 5200 mg/kg (Table 1). The grain size of the original samples was also considered in creating the composite scheme so the samples would have grain-size ranges that have been determined to be suitable for soil washing [7]. Prior to compositing, all soil samples

Table 1
Concentrations of PCBs and lead in composite soil samples

	PCBs (mg/kg)	Lead (mg/kg)
Sample L	52	560
Sample A	140	1700
Sample H	670	5200
Sample H (duplicate)	700	na

na: Not analyzed.

were prescreened over a US 1 in. sieve. Grain size and specific gravity of Samples L, A, and H are shown in Table 2.

2.2. Particle size separation

For the separation procedure, 5000 g of each sample (L, A, and H, plus one duplicate of Sample A) was washed through a set of sieves using 20 l of wash water consisting of carbon-filtered and deionized water at a 1:2.5 ratio. The wash water pH was adjusted to 8.0–8.2 using ammonium hydroxide to approximate the pH of water at the site. Each sample was separated into five particle size ranges as follows: > 4.75 mm, 4.75–2.0 mm, 2.0–0.425 mm, 0.425–0.074 mm, and < 0.074 mm. The four coarser size fractions were air-dried and then subsampled for chemical analysis. The fine material less than 0.074 mm, including the wash water, was allowed to settle for 6 h. The supernatant and fine-grained material were then subsampled for chemical analysis. All samples were analyzed for PCBs and lead. In addition, three particle size fractions were analyzed for specific gravity to evaluate its potential relationship with the concentration of lead. The material > 4.75 mm was examined visually for the presence of particulate lead.

2.3. Sample prescreening for soil washing

Six samples, two each of L, A, and H, were prepared for soil washing testing. The soil samples were prescreened to remove fines from the soil prior to washing as the fines

Table 2
Grain size and specific gravity of composite soil samples

	Percent of sample	Specific gravity
<i>Sample L</i>		
> 4.75 mm	37	na
4.75–2.0 mm	13	na
2.0–0.425 mm	19	na
0.425–0.074 mm	16	na
< 0.074 mm	15	na
<i>Sample A</i>		
> 4.75 mm	39	na
4.75–2.0 mm	14	2.65
2.0–0.425 mm	25	2.72
0.425–0.074 mm	14	2.66
< 0.074 mm	8	na
<i>Sample H</i>		
> 4.75 mm	35	na
4.75–2.0 mm	15	2.67
2.0–0.425 mm	21	2.75
0.425–0.074 mm	17	2.74
< 0.074 mm	12	na

na: not analyzed.

interfered with the operation of the soil washing unit. The samples were prescreened by agitating 2000 g of material with approximately 8 l of wash water prepared as described previously for the particle size separation step. The agitation was performed to aid in the breakdown of soil agglomerates and to remove some of the fine material and organic matter. Material was then passed through a sieve to remove fine material less than 0.425 mm diameter for Samples A and H and less than 0.125 mm diameter for Sample L. The wash water from the agitation of each sample was settled for approximately 4 h. The settled fines and sludge were saved to combine with fines produced later during the soil washing step.

2.4. Soil washing procedure

The apparatus used to conduct the soil washing was a pilot-type rotating trommel washer. The soil was fed through the hopper and dropped into the wash chamber. In the wash chamber, wash water was recirculated continuously to the soil mix through a spray bar with four nozzles extending the length of the chamber. As material passed through the wash chamber, additional spray nozzles assisted in the removal of finer material passing through a series of slotted screen sections ranging in size from 0.005 to 0.10 cm. The finer materials passing through the screen sections was collected on shallow trays equipped with a No. 320 screen. This allowed water and fines to pass through a filter housing unit equipped with a 0.025- μm -rating welded liquid filter bag. The filtered wash water was recirculated to the spray bar through a 50 gpm pump. The coarser material (approximately 1 mm and larger) passed out the end of the trommel barrel into a collection container. The material passing through selected trommel screen sections could be removed for analysis or composited with the coarse material exiting the end of the trommel barrel. The water pressure of the spray bars was set at 30–40 psi and the water temperature at 38°C. For each wash, 60 l of wash water were added to the trommel reservoir and reused throughout the wash cycle. The duration of the wash cycle was 50 min.

2.5. Water wash

One 2000 g subsample of each of the three soil samples L, A, and H was subjected to soil washing using water only. The wash water was prepared combining carbon-filtered and deionized water at a 1:2.5 ratio and adjusting the pH to 8.0–8.1 with ammonium hydroxide to represent site water.

Samples L and H were each subjected to one cycle of water washing (Fig. 1). Sample A was subjected to three cycles to evaluate the effect of multiple cycles. For Samples L and H, soil, residual fine material and residual water samples were collected after one wash cycle. For Sample A, soil was collected after each of the three cycles and analyzed separately. Fine material was collected after each cycle, and composited into one sample for analysis. Residual water was collected for analysis after the first cycle. The fine material samples from L, A, and H were combined with their respective fine material

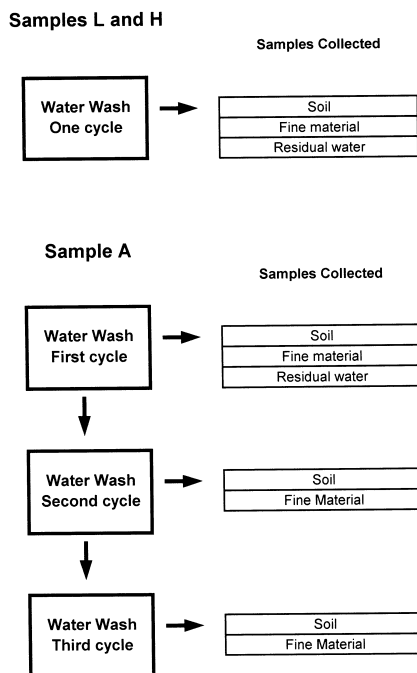


Fig. 1. Schematic diagram of sample treatment and collection for water washing of soil.

samples collected from the prescreening step. All samples were analyzed for lead and PCBs. Toxicity Characteristic Leaching Procedure (TCLP) extractions for lead were performed on soil samples collected at the end of each test, and in all fine material samples.

2.6. Surfactant and pH-adjusted wash

One 2000 g subsample of each of the three soil samples L, A, and H was subjected to a surfactant wash followed by a pH-adjusted wash. The surfactant wash water consisted of a 0.5% by weight solution of Witconol 1206 in carbon-filtered/deionized water prepared as described previously. The pH-adjusted water consisted of the wash water adjusted to a pH of less than 4 by adding nitric acid.

Samples L and H were each subjected to one cycle of surfactant washing followed by one cycle of pH-adjusted washing (Fig. 2). Sample A was subjected to three cycles of surfactant washing followed by three cycles of pH-adjusted washing. For Samples L and H, residual water samples were collected after each surfactant and pH-adjusted cycle. Soil samples were collected after the pH-adjusted cycle. For Sample A, residual water samples were collected after the first surfactant and first pH-adjusted cycles and soil samples were collected after the third surfactant and third pH-adjusted cycles. Two soil subsamples were crushed and analyzed to evaluate the effect of laboratory subsampling

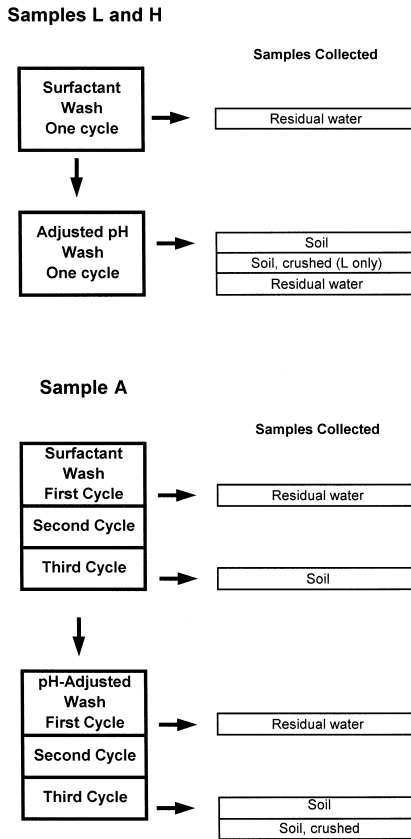


Fig. 2. Schematic diagram of sample treatment and collection for chemically enhanced soil washing.

bias. Fine material was not collected. All samples were analyzed for PCBs and lead. Soil samples collected in the final cycle of each test were analyzed for TCLP-lead.

3. Results and discussion

3.1. Particle size separation

Concentrations of PCBs were higher in the finer size fractions than in the coarser size fractions for all of the samples (Fig. 3), with the exception of the 2.0–0.425 mm range for Sample H. Concentrations of lead also showed a general increasing trend with finer size fractions with the exception of the 2.0–0.425 mm range for Samples A and L, where concentrations of lead were higher than in the two smaller size fractions (Fig. 4). Concentrations of lead and PCBs in residual water increased with higher soil concentrations (Table 3).

The higher concentrations of PCBs in smaller particle size fractions is likely due to the higher surface area-to-volume ratio in soils with finer grain size. Finer grained soils

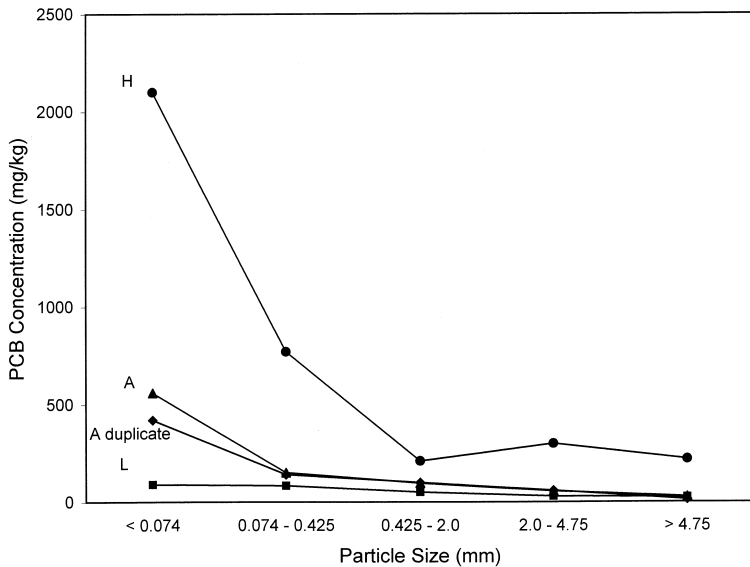


Fig. 3. Concentration of PCBs in soil after particle size separation.

also generally have a higher organic matter content which can result in higher adsorption of PCBs. The specific gravity data indicate that the high lead concentrations in the 2.0–0.425 mm size range in Samples A and H may be due to the presence of particulate lead. Visual observations revealed tire ballast weights and other miscellaneous scrap metal in the soil, also indicating particulate lead. Particulate lead is heavier than soil

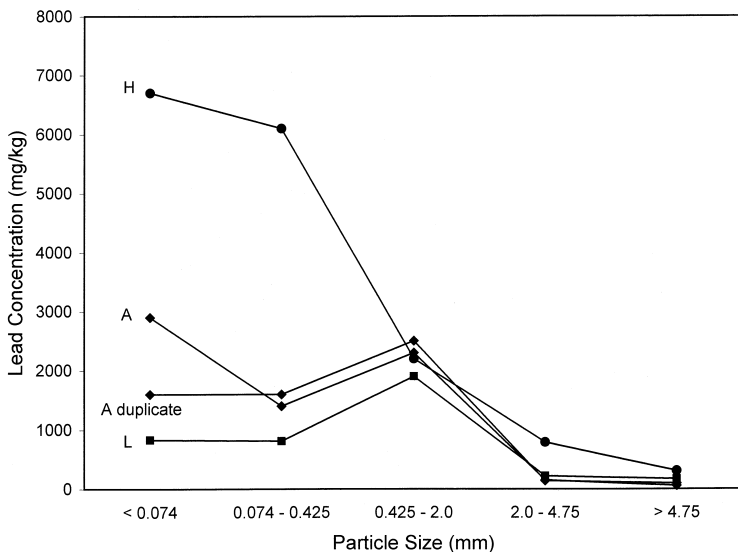


Fig. 4. Concentrations of lead in soil after particle size separation.

Table 3
Concentrations of PCB and lead in residual water from the particle-size separation process

	PCBs ($\mu\text{g}/\text{l}$)	Lead (mg/l)
Sample L	42	8.2
Sample A	110	17
Sample A (duplicate)	210	13
Sample H	550	34

minerals, and the specific gravity was highest in the 2.0–0.425 mm size range for Sample A (Table 1). Specific gravity was not measured on Sample L.

The preliminary treatment goal of 10 mg/kg PCBs was not attained in any of the soil fractions from particle-size separation. The concentration of lead was lower than the preliminary treatment goal of 500 mg/kg in all soil fractions > 2.0 mm for Samples L and A, and in all soil fractions > 4.75 mm for Sample H (Fig. 4). These fractions comprised 50, 53, and 35% of the soil mass for Samples L, A, and H, respectively (Table 1).

3.2. Effects of soil washing on PCBs

The efficiency in removing PCBs from soil to fine material or water after one water wash ranged from 27 to 70%, with an average of 55% for the three soil samples (Table 4). Adding three water washes for Sample A increased the removal efficiency from 70 to 75%. Average PCB removal efficiencies for the surfactant and pH-adjusted water wash were higher than for the water wash alone, ranging from 71 to 95%, and averaging 78% among the three soil samples (Table 4; Fig. 5). The surfactant wash had a greater effect on removal of PCBs than the pH-adjusted wash, as shown by the very small increase in PCB removal from 94 to 95% in Sample A.

The preliminary treatment goal of 10 mg/kg PCBs was attained in Sample A (7 mg/kg) and was almost met in Sample L (15 mg/kg). The higher concentration of PCBs in the screened Sample L is likely because Sample L included particles > 0.149 mm and was only washed through one cycle, whereas Sample A contained only particles > 0.425 mm and was washed three times. The results indicate that this method is applicable for treating soils at the site contaminated with PCBs at concentrations up to 140 mg/kg.

Concentrations of PCBs were generally two to four times higher in residual fine material collected after one water wash than in the untreated bulk soil samples (Table 5). Concentrations of PCBs were substantially higher in the water collected after one surfactant wash than in water collected after one water wash or one pH wash, indicating that the surfactant step has the greatest effect on mobilizing PCBs into the aqueous phase.

3.3. Effect of soil washing on lead

Results for lead were highly variable, indicating a heterogeneous distribution of lead in the soils as evidenced by higher lead concentrations after washing than before

Table 4

PCBs, lead and TCLP-lead in soils before and after washing procedures and removal efficiencies

	Concentration in soil			Removal efficiency	
	PCBs (mg/kg)	Lead (mg/kg)	TCLP-lead (mg/kg)	PCBs	Lead
<i>Sample L (> 0.149 mm)</i>					
Before washing	52	560	na		
After one water wash	38	720	2.4	27	0
After surfactant and pH-adjusted water wash	18	670	1.5	65	0
After surfactant and pH-adjusted water wash (duplicate)	12	220	na	77	61
Average for surfactant and pH-adjusted water washes	15	445	na	71	30
<i>Sample A (> 0.425 mm)</i>					
Before washing	140	1700	17		
After one water wash	42	1600	na	70	6
After three water washes	35	10000	27	75	0
After three surfactant washes	9	950	na	94	44
After three surfactant and pH-adjusted water washes	5	740	36	96	56
After three surfactant and pH-adjusted water washes (duplicate)	4	2200	na	97	0
After three surfactant and pH-adjusted water washes (crushed)	11	495	5.5	92	71
Average for surfactant and pH-adjusted water washes	7	1145	na	95	42
<i>Sample H (> 0.425 mm)</i>					
Before washing	700	5200	67		
After one water wash	230	2800	710	67	46
After one water wash (duplicate)	190	7100	na	73	0
Average for water washes	210	4950	na	70	23
After surfactant and pH-adjusted water wash	220	2100	150	69	60

washing in some samples (Table 4). These variable data suggest the presence of particulate lead, such as the concentration of 10 000 mg/kg measured in Sample A after three water washes. Overall, the chemically enhanced water wash improved the removal efficiencies when compared to the water wash only (Table 4). The increase in average removal efficiencies for lead from the water wash to the chemically enhanced water wash were from 0 to 30% for Sample L, from 3 to 42% for Sample A, and from 23 to 60% for Sample H. Average removal efficiencies appeared to increase with increasing concentrations of lead in the soils (Fig. 6). Neale et al. [5] also observed the greatest removal of lead and cadmium at the highest initial concentrations in soil, suggesting that lower concentrations of metals in soils may be more tightly bound and more difficult to extract.

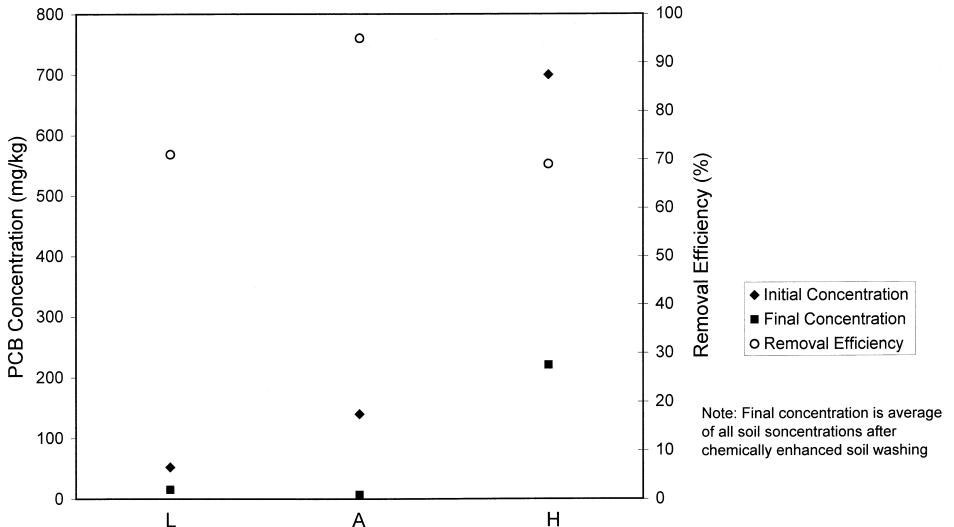


Fig. 5. Concentrations of PCBs in soils after chemically enhanced soil washing.

The addition of the pH-adjusted water wash for Sample A showed a high variability in the results, with an increase in the removal efficiency of lead in the sample from 44 to 56% but no increase in the duplicate sample. Other studies have shown that pH is an important factor in removing lead from soils, with greater removals observed at a lower

Table 5
Concentrations of PCBs and lead in residual fine material and water from chemically enhanced soil washing

	Residual water		Fine material	
	PCBs ($\mu\text{g}/\text{l}$)	Lead (mg/l)	PCBs (mg/kg)	Lead (mg/kg)
<i>Sample L</i>				
After one water wash	4	0.13	140	1200
After one surfactant wash	65	0.21		
After one pH-adjusted wash	7.5	0.13		
<i>Sample A</i>				
After one water wash	58	0.26	320	3500
After one surfactant wash	210	1.0		
After one pH-adjusted wash	1.8	0.29		
<i>Sample H</i>				
After one water wash	50	0.46	2700	8500
After one surfactant wash	540	1.5		
After one pH-adjusted wash	14	0.35		

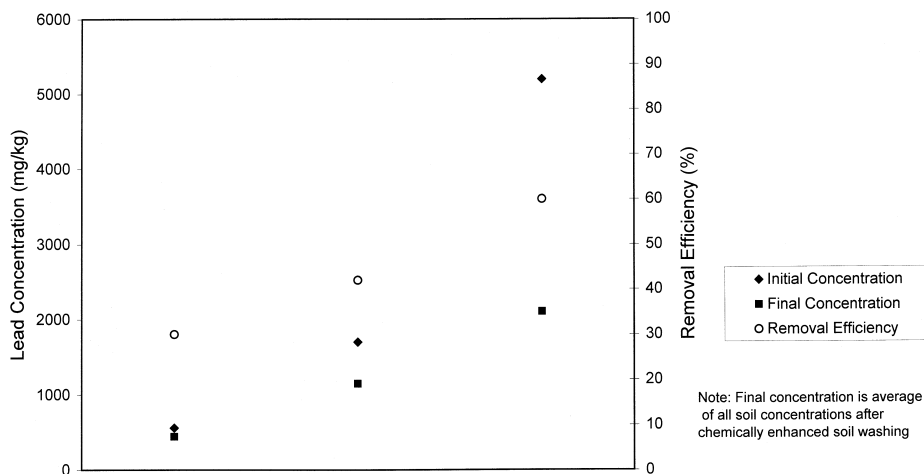


Fig. 6. Concentrations of lead in soils after chemically enhanced soil washing.

pH [4,6]. Grasso et al. [4] also observed that the number of soil washing stages needed for lead removal decreased with lowered pH. In that particular study using Connecticut soils, a pH of 3.25 required five equilibrium stages [4]. These data indicate that a lower pH used with this method or an increased number of wash cycles may improve the removal of lead. In addition, increased residence time of the solution with the soil to allow time for equilibrium may improve the removal efficiency.

TCLP results showed that washed soil from Sample L met the EPA limit of 5 mg/l after both the water wash and chemically enhanced wash (Table 4). TCLP-lead concentrations in washed soil from Samples A and H exceeded the EPA limit, which would require these materials to be regulated as a hazardous waste and subject to land disposal restrictions. The results were highly variable and showed higher concentrations after washing than before washing in Samples A and H.

Chemically enhanced washed soil from Sample L contained lower concentrations of lead (445 mg/kg) than the preliminary treatment goal (500 mg/kg), but washed soil from Samples A and H did not meet the treatment goal. These data indicate a practical limiting concentration of about 560 mg/kg for treatment of lead-contaminated soils to concentrations below 500 mg/kg without prior particulate lead removal. If particulate metals such as lead are present in soils, a specific gravity separation step should precede any enhanced chemical leaching step to maximize metal leachability and to reduce total concentrations remaining in the soil.

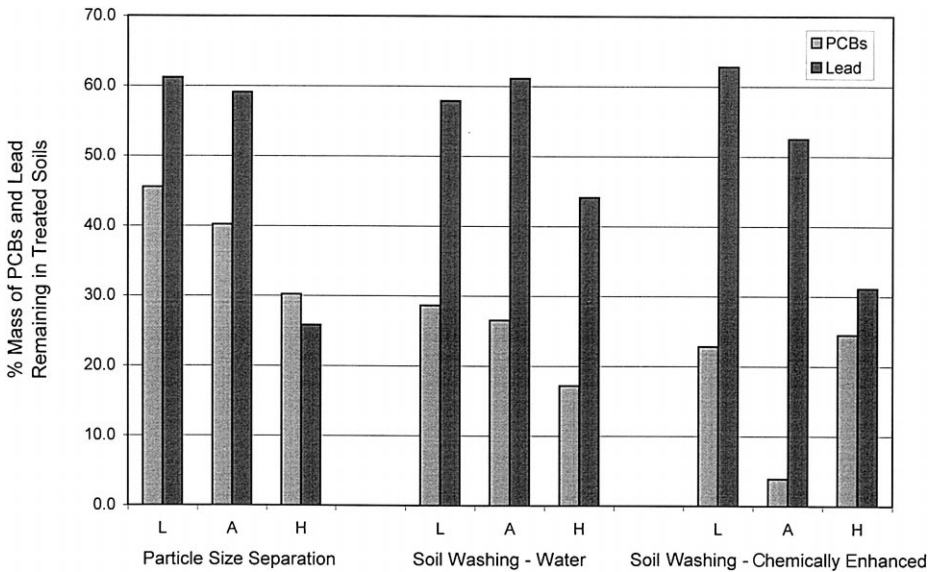
Concentrations of lead were generally about two times higher in residual fine material than in the untreated bulk soil samples (Table 5). Concentrations of lead were slightly higher in the water collected after one surfactant wash than in water collected after one water wash or one pH wash. Lead concentrations were much lower in residual water collected after the chemically enhanced wash than water collected from the particle-size separation process (Tables 3 and 5).

3.4. Comparison of particle-size separation and chemically enhanced washing

The relative removal of chemical mass from coarser soil particles is shown for the three different soil washing procedures in Fig. 7. For all samples, the percentage of PCBs removed from soil was generally lowest with particle size separation and highest with chemically enhanced soil washing. The three cycles of chemically enhanced soil washing had a positive effect on removing PCBs from Sample A, compared to the results observed for Samples L and H which only had one cycle. For lead, soil washing with either water or additives did not improve contaminant removal from the soil. In general, the most highly contaminated soil, Sample H, had the highest amount of contaminant removed using each treatment method.

Table 6 compares the lowest final contaminant concentrations reached for the particle size separation and the chemically enhanced soil washing along with the percentage of total material the soil comprises. Particle-size separation reduced the amount of material above the lead preliminary treatment goal by 35 to 53%. In the chemically enhanced soil washing which only separates material into fines and soil, the lead preliminary treatment goal was only achieved in the soil with the lowest initial concentration of lead.

The chemically enhanced soil washing method using three surfactant washes was successful at cleaning 78% of the material to concentrations below the preliminary treatment goal for Sample A. Surfactant cleaning of Sample L reduced soil PCB concentrations to 15 mg/kg, very close to the preliminary treatment goals. Those results indicate that Sample L would likely reach the treatment goal of 10 mg/kg with



Note: Soils are in the particle size range >0.425 mm, except Sample L after soil washing, which contains particles >0.149 mm.

Fig. 7. Mass of PCBs and lead remaining in soils after particle size separation and soil washing.

Table 6

Final soil treatment concentrations and volumes using particle size separation in comparison to soil washing methods

	Lead			PCBs		
	L	A	H	L	A	H
<i>Particle size separation</i>						
Initial concentration (mg/kg)	560	1700	5200	52	140	700
Final concentration (mg/kg)	170–220	56–150	310	24–29	13–59	210–300
Percent of material	50	53	35	50	53	71
Grain size of material (mm)	> 2.0	> 2.0	> 4.75	> 2.0	> 2.0	> 4.75
<i>Surfactant / pH wash</i>						
Initial concentration (mg/kg)	560	1700	5200	52	140	700
Final concentration (mg/kg)	445	1145	2070	15	7	220
Percent of material	79	78	71	79	78	71
Grain size of material (mm)	> 0.149	> 0.425	> 0.425	> 0.149	> 0.425	> 0.425

Concentrations in bold are less than the preliminary treatment goals.

additional screening of fines and more surfactant washes. Particle size separation without chemical enhancement was not successful at reducing PCBs to preliminary treatment goals in any of the soils.

4. Conclusions

The chemically enhanced soil washing procedure was the most effective of the methods tested for removing PCBs from the coarser soil fractions, with removal efficiencies as high as 95% for the sample treated with three wash cycles. The surfactant appeared most effective at mobilizing PCBs into the residual wash water. It is estimated that close to 80% of the material contaminated with up to 140 mg/kg PCBs could be treated to concentrations below 10 mg/kg using three wash cycles of surfactant wash water.

Particle size separation and soil washing with water or surfactant and pH-adjusted water appeared about equally successful at removing lead from the soil fraction into the fine material and residual water fraction. The highest proportion of lead was removed from the most highly contaminated soil using all methods. It should be noted that the lead results contain uncertainty because of the heterogenous nature of the soil which produce high variability in the data and could also make quality control difficult for a soil washing remedy. Prior removal of particulate lead using flotation or gravity settling is recommended for these soils before washing to improve removal efficiencies. Lead contamination was reduced from concentrations as high as 1700 to 56–220 mg/kg in about half of the material in Samples L and A using particle size separation and from 560 to 445 mg/kg in 79% of the material in Sample L using chemically enhanced soil washing. Greater removal efficiencies might be obtained using lower pH in the wash water and a higher number of wash cycles.

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