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RADIONUCLIDE CONTAMINATED SOIL: LABORATORY STUDY AND ECONOMIC ANALYSIS OF SOIL WASHING

Final Report

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EXECUTIVE SUMMARY

The objective of the work discussed in this report is to determine if soil washing is a feasible method to remediate contaminated soils from the Hazardous Waste Management Facility (HWMF) at Brookhaven National Laboratory (BNL). The contaminants are predominantly Cs-137 and Sr-90. We have assumed that the target activity for Cs-137 is 50 pCi/g and that remediation is required for soils having greater activities. Cs-137 is the limiting contaminant because it is present in much greater quantities than Sr-90. This work was done in three parts, in which we:

- estimated the volume of contaminated soil as a function of Cs-137 content,
- determined if simple removal of the fine grained fraction of the soil (the material that is less than 0.063 mm) would effectively reduce the activity of the remaining soil to levels below the 50 pCi/g target,
- assessed the effectiveness of chemical and mechanical (as well as combinations of the two) methods of soil decontamination.

From this analysis we were then able to develop a cost estimate for soil washing and for a baseline against which soil washing was compared.

The volume of contaminated soil present at the HWMF first was estimated from measurements of Cs-137 in cores taken at the site. This estimate was later refined through the use of a high resolution "walk-over" survey using a shielded Nal gamma detector. Our estimate for the volume of contaminated soil (with activities greater than $50pCi/g$) was $35,600$ m³. This is discussed in detail in Appendix A.

Activities in the soil at the site vary from near background to $810,000$ pCi/g. Grain size separations for 15 stations by wet sieving show that removal of the fine grained fraction of soil from the HWMF is an effective method for decontamination of samples that contain activities to about 3,000 pCi/g. When soil is contaminated to much higher levels, Cs-137 is distributed throughout the size classes; often at activities that exceed the target. In these cases the soil may not be adequately cleaned. Surficial samples containing 2800 pCi/g have been successfully decontaminated. Samples with higher activities, particularly those from the subsurface, are more "recalcitrant" and soil washing becomes less effective as the activity increases.

Using the data generated in the laboratory studies (e.g. activity of fines, grain size to activity relationship) and costs obtained through several vendors, we have assessed the effectiveness of soil washing and bounded its limitations regarding the volume of material that probably cannot be treated with this technique. We have tried to make our cost estimate as complete as possible, including the full cycle of costs such as transportation and disposal associated with soil washing. In comparing soil washing to the baseline we have kept our assumptions and unit costs on the same basis so that the comparison is valid. The final cost estimate for the baseline is 81 million dollars. In contrast the estimate for soil washing is 53

million dollars. In reality, the bulk of the costs are not for the processing itself but for transportation and disposal of residual material and soil that cannot be decontaminated.

Soil washing provides a significant cost savings over the baseline (excavation and disposal of all soil with activities greater than the TPL). This savings varies with volume of soil requiring remediation and therefore with the inverse of the TPL (since as the TPL increases, the volume of soil decreases). At a TPL of 50 pCi/g the saving is 27 million dollars, while at a TPL of 110 pCi/g the saving is 22 million dollars.

1. OBJECTIVE

The objective of the work discussed in this report is to determine if soil washing is a feasible method to remediate contaminated soils from the Hazardous Waste Management Facility (HWMF) at Brookhaven National Laboratory (BNL). The contaminants are predominantly Cs-137 and Sr-9O. We have assumed that the target activity for Cs-137 is 50 pCi/g and that remediation is required for soils having greater activities. In the first phase of this work we estimated volumes of contaminated soil at the HWMF and defined the relationship between volume of soil and Cs-137 activities. In the second phase, we focused on:

1) better defining the relationship between volumes and activity levels of Cs-137 and,

2) determining if simple removal of the fine grained fraction of the soil (the material that is less than 0.063 mm) would effectively reduce the activity of the remaining soil - the material that is greater than 0.063 mm - to levels below the 50 pCi/g target.

In this, the third and final phase of this study we have assessed the effectiveness of various methods of soil decontamination. Chemical and mechanical methods (as well as combinations of the two) were tried. This report contains results of the soil washing treatability study and the cost analysis of soil washing that is based on the treatability study. Volume estimates from Phase I are contained in a separate document which is attached to this report as Appendix A.

2. BACKGROUND

Remediation of contaminated soil has been explored at laboratory scales for a broad range of soil and contaminant types. Removal or destruction of some organic contaminants from surficial soils has been successful, and the techniques have been applied to many sites. Removal of radionuclides from soils has not been as thoroughly examined and rarely applied at even a demonstration level. Only two studies have been conducted to remediate soil that was contaminated with Cs-137. This radionuclide (with a 30 year half-life) tends to have high partition coefficients for soils, especially those containing illite/mica minerals. Results of those two studies are reviewed below.

At the Idaho National Engineering Laboratory (INEL) four soil samples, with activities ranging from 66.8 to 1855 pCi/g, were subjected to a variety of treatments in attempts to remove Cs-137 [Cowatch et al, 1994, INEL-94/0171]. The target level for the decontaminated soil was 10 pCi/g. Processes applied included: sieving, flotation, low-energy attrition (tumbling), high energy attrition scrubbing, and brine extraction. The single most useful treatment was sieving which reduced the volume of material requiring treatment by 20 to 40 %. Brine extraction, using 0.1 M KNO₃ removed up to 21% of the activity on the fine fraction material. Sieving and attrition scrubbing of low to medium levels of contamination (about ten times the target level) were found to be the most reasonable alternatives. These two alternatives would subsequently need to be evaluated against cost, volume reduction of waste generated, risks associated with excavation and additional handling.

A bench-scale feasibility study was conducted at Hanford to assess techniques to remove radionuclides from two soil types from the "100 Area" [DOE/RL-92-51,1993]. The radionuclides were: U, Cs-137, Co-60 and Eu-152. The Cs-137 activities ranged from 0.74 to 5495 pCi/g. This soil is relatively coarse containing about *5%* fines. Attrition scrubbing for 30 minutes, reduced the activity on the soil by 80%, 28% and 61% for Co-60, Cs-137 and Eu-152 respectively. A number of parameters (e.g. impeller torque, time, energy input and percent solids) for attrition scrubbing were varied to optimize the process. A range of solution types were also examined for their effect. A combination of 30 minutes of attrition scrubbing with a 0.5M ammonium acetate solution, adjusted to $pH = 3$ with citric acid, released 79% of the Co-60, 39% of the Cs-137 and 83 % of the Eu-152.

In neither study was Cs-137 successfully removed from the soil. However, in both studies the soil was volcanic in origin and contained at least several percent of illitic minerals. This is not the case at Brookhaven where the soil, particularly the deeper material, is predominantly quartz sand.

3. METHODS

The soil contamination data, for stations not included in the survey performed by CDM, were obtained after first packing soil into standard Marinelli beakers. This geometry is calibrated against NIST standards in the same geometry. These samples were used to report the activity in the total, unsieved material. Radionuclide counting was performed by the BNL Chemistry Department (G. Harbottle).

An overview of the experimental process is given in Figure 1, which is a flow-chart of the steps taken to decontaminate the soil. This study consists of laboratory scale experiments in which several factors were examined, including:

- association of Cs-137 with different grain size fractions,
- ability of wet sieving to remove Cs-137 from the coarse (> 0.063 mm) fraction,
- ability of reagent extraction to remove Cs-137 from the coarse fraction,
- ability of attrition scrubbing with reagents to remove Cs-137 from the coarse fraction.

Soil size fraction data were obtained by wet sieving aliquots of the soil samples weighing between 100 and 150 grams. In wet sieving, standard screen sieves are used, and the soil particles are washed through the sieves with de-ionized water. The wash water is collected in a bucket along with particles that pass through the finest sieve. The sieve contents for each size fraction were transferred into plastic cups and dried in air under a heat lamp prior to weighing. The fines fraction (passes through No.230 sieve, or designated -230) was determined in two ways depending on the volume of water needed to wet sieve the sample. With a small volume the sample was dried and weighed directly. With larger volumes the sample could not be dried and weighed directly; the weight was calculated by difference. Sieve sizes are indicated in Table 1.

Radionuclide counting at the EWTC was performed using a Ge detector interfaced with a computer controlled Canberra S100 data acquisition system. Most of our analysis was used to

assess the effectiveness of the soil separation process in removing Cs-137. Measurements were made of samples in capped plastic cups. While the mass of soil varied, the basal geometry of the samples was always similar, i.e., discs with a diameter of 5 cm with thicknesses ranging from 0.1 cm to 3 cm. We determined that there is no effect of geometry on the count rate per gram as long as the samples weigh less than about 30 g. Above that weight the count rate (cpm/g) becomes lower than it should be due to distance of the bulk of the sample from the detector and to self absorption. The Cs-137 count rates of unsieved soil samples (references) in the plastic containers were correlated with the pCi/g values obtained for splits of the same samples analyzed by the BNL Chemistry Department. In this way the count rates of samples in the plastic cups were referenced to the count rates determined in the samples sent to the Chemistry Department. This calibration allowed direct calculation of activities from our count rates. It also allowed comparison of the original soil activities with those in the fine fraction of the separated samples. In addition to the collected fractions, a reference (unsieved) sample from each station and a sample of wash water, after it was passed through a $0.45 \mu m$ filter, were counted.

During Phase III of the treatability study, in which reagents and physical methods were used to decontaminate the soil, we counted 4 gram samples in plastic scintillation vials. Again, analysis was done with an intrinsic Ge gamma-detector. The scintillation vial geometry was calibrated with standards by counting them on our system and on a previously calibrated system at the BNL Chemistry Department.

3.1. Material

Phase II involved characterizing soil samples provided by Office of Environmental Restoration (OER) staff from within the RCRA area. In addition two samples (taken at stations 4 and 5) were obtained by staff from the Environmental and Waste Technology Center (EWTC) from just outside the fenced perimeter. Many of the samples examined were surface samples, containing a significant amount of organic material, i.e., roots, leaves, and other plant detritus. Other samples were obtained from a set of archived specimens taken by geoprobe. Sample numbers and their contamination levels are given in Table 2. The locations of the samples are shown in the attached map, Figure 2. From these samples, two types of surficial soil were observed: an ocher colored fine grained material with some gravel sized grains. The other soil type has less gravel and is darker brown. At depth the soil often contains less fines except when balls of clay are present. This material, by visual inspection, is dominated by yellow iron bearing weathering products and in some cases by balls of grey clay, which contribute significantly to the fines fraction. One sample (37a, the hottest sample) contained a large percentage of blue paving gravel and few fines.

Figure 1. Flow-chart of soil washing used in this study.

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Table 1. Sieve Sizes Used in Separations

Bulk density measurements were made for surface material from stations 4 and 5. This was done by carefully sampling the soil and measuring the sides and depth of the hole. The total weight and water content were determined and bulk density was calculated. Sample 4 was 1.24 g/cc and sample 5 was 1.17 g/cc. Sample 4 contained 2.4 Wt *%* of gross organic matter, consisting of leaves, stems and roots, while station 5 had 3.8 Wt %. These values do not include the small particles of organic detritus that was observed in the separated size fractions. As a rough estimate, about 4 Wt % of the surface material is organic matter.

The fact that the distal samples (stations 1-5) contain observable (albeit low) activity is probably the result of wind blown, contaminated fines being transported away from areas of higher activity and being deposited near-by. This dispersion could also be the result of plant uptake and then transport of plant matter by wind or animals. Background levels of Cs-137 from nuclear weapons fallout is about 1 pCi/g . One sample of grass and roots taken from station 3 and counted in the Chemistry department indicated that the organic matter accumulated uranium, concentrating it to 26 pCi/g (the soil in which the vegetation grew contained 1.35 pCi/g). The Cs-137 activity for this organic matter was 2.02 pCi/g while the soil contained 2.82 pCi/g.

We have calculated the area of the fenced enclosure as 62,000 square meters. Based on areas taken from a map of the site we estimate that 21% (13,000 square meters) of the area is covered with black-top, giving an area of 49,000 square meters that contain vegetation. Using a bulk density of 1.2 g/cc (measured value from two samples) and an organic matter content of 4 Wt *%,* we estimate that in the surficial 10 cm there are 2400 Kg of organic matter.

Figure 2. Map of the HWMF showing locations of stations .

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Station No.	$Cs-137$, pCi/g	U-238, pCi/g	Sr-90 pCi/g	Source of Cs-137 Data	
ı	2.84	1.71		Chem	
$\overline{2}$	5.25	1.00		Chem	
3, soil 3, grass	2.82 2.02	1.35 26.11		Chem Chem	
$\overline{\mathbf{4}}$	1.53	1.24		Chem	
5	1.60	1.32		Chem	
6	1078		0.34	EWTC	
$\overline{7}$	56.3	6.52	0.38	Chem	
12	50			EWTC	
13	2812			EWTC	
37a	810,000		1300	CDM	
37 b	190		0.25	CDM	
37 c	4,700		0.28	CDM	
37 d	6,400		0.28	CDM	
38 b	7.2		0.14	CDM	
52 c	11.0		0.16	CDM	

Table 2. OU1 Soil Sample Contamination Levels

4. PHASE H, RELATIONSHIP OF GRAIN SIZE TO SOIL VOLUME

4.1 Grain Size

Results of size determinations by wet sieving are listed in Appendix B, and compared graphically in Figure 3. The first samples analyzed were those with relatively low activities, in order to be sure our methods provided adequate contamination control. These sample were all surficial material containing grass, *etc.* The samples taken for sieving had the grass and other gross organic matter removed. Nevertheless each separated fraction from surface samples, as it dried, segregated into an upper layer of fine grained organic material (resembling peat) and the remainder of the sample containing the inorganic matter. The weight percentage of the fines, the fraction that passed through the 0.063 mm sieve (sample designation -230), varied from 22.4 Wt *%* to 66 Wt %. Samples 37b and 38b, taken at depths of 2 feet, tend to have large percentages

of fines; 45 *%* and 66 % respectively. Other samples from 4-6' and 8-10' contained lower percentages of fines averaging about 16 %. One surface sample (37a, the hottest sample) contained a large percentage of blue stone paving gravel and relatively little fines (16 %).

4.2 Radionuclides as a Function of Grain Size

Radionuclide analysis of separated size fractions (see Appendix C) indicates that most of the activity, from 81 to 92% of the Cs-137, is in the fine fraction, no matter the amount of fines present. Sample 37d is the exception to this, with the fines containing only 59 % of the activity. This is shown in Figure 4. In every case, the activity per gram of the fine fraction is the highest of any fraction. Care should be taken in using Figure 4 because it shows the percentage of total activity in each size fraction, regardless of the total activity. Consequently, for heavily contaminated samples, the coarse material may contain only a small fraction of the total activity but that small percentage may represent activities greater than the 50 pCi/g target.

Cs-137 activities for untreated samples 7, 12 and 37b were only slightly above the target of 50 pCi/g. Removal of the fines (41 *%,* 33 % and 45 % respectively) resulted in the remainder of the soil being below the target activity. Separation of fines by wet sieving samples 6, 13, 37a, 37c and 37d was not successful in reducing the activity of Cs-137 to acceptable levels . Some of the fractions greater than 0.063 mm contain significant activities of Cs-137. There does not appear to be a trend, but even some of the coarsest material (on the # 35 and 80 sieves) is contaminated to levels well above the target level and will need to be treated to lower the activity to acceptable levels.

Sr-90 was measured in size fractions of samples from stations 6, 7, 37a, 37b, 37c, and 37d. The samples with the lowest activity of Cs-137 were excluded from the analysis. The highest activity of Sr-90 measured was 24.5 pCi/g in the #100 sieve fraction at station 37a. Why this radionuclide is associated with this particular fraction is unknown. Unfortunately material from the two coarsest fractions (#8 and 35 sieves) was not available for Sr-90 analysis. Sr-90 tends to concentrate both in the fines and, for surficial samples, in the coarsest fraction. This probably represents an association with the organic matter (plant detritus) that generally dominates the #8 sieve fraction. Confirmation that there is uptake and concentration of Sr-90 in plants would be an important point for phytoremediation studies and in assessing remobilization and redistribution of Sr-90 from depth to the surface of the soil.

4.3 Activity in Wash Water

Water that had been used to wet sieve each sample of the soil was analyzed for Cs-137. Results of this analysis are given in Appendix C. In all cases but one, the activity in the wash water was below detection limits. Only in the water used to wash sample 37a was Cs-137 observed. The filtered (0.45 micron pore size) water contained about 400 pCi/g. This may indicate that station 37a is still an active source of Cs-137 for the subsurface, leaching activity with each rainfall.

Soil Particle Size

Figure 3. Weight percentages of particles size fractions from stations at the HWMF.

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5. PHASE IU, CHEMICAL AND PHYSICAL DECONTAMINATION OF SOIL

5.1 Methods

Having found that activities of Cs-137 can be greater than the target even in the coarser grained (sand and gravel) material, we examined the possibility of using physical and chemical methods to remove Cs-137 from the soil. This set of experiments was not meant to be definitive, rather it was intended to determine the feasibility of mobilizing Cs-137 from the soil.

5.1.1 Preliminary Screening

For preliminary screening of chemical and physical methods to remove Cs-137 from the coarse fractions of soil, three splits were made of each of two size fractions from station 13. Both fractions contained significant activities of Cs-137. They were the 80 mesh fraction containing about 500 pCi/g and the 230 mesh fraction containing 2250 pCi/g. One split from each was counted as a reference while the other two were used for the experiment. Four grams of the second split were placed in a centrifuge tube with 20 g of deionized water (DIW) and mixed on a Vortex-Genie for 5 minutes (at a setting of 9). The sample was then placed on a 230 mesh sieve and the newly generated fines washed through. The remaining soil sample was dried and counted. The third split was reacted with 1 M citric acid solution for 15 minutes under gentle agitation. It was then sieved, rinsed, dried and counted.

5.1.2 Reagent Extraction

Soil from the BNL/HWMF was tested to determine if it can be decontaminated by using reagents to extract Cs-137. The testing methodology was:

- 1. Wash fines (less than 63 microns) from about 75 grams of "as received" soil.
- 2. Dry the coarse fraction, weigh and mix well.
- 3. Weigh out 4g aliquots of the soil (coarse fraction);
	- A. Samples for testing go into centrifuge tubes
	- B. Samples for references go into plastic scintillation vials.
	- C. Because of variability in activity, some stations require counting the sample before and after treatment.
- 4. Add 12 g of reagent solution to soil samples.
- 5. Place on shaker table for 3 hours.
- 6. Pipette out about 4 mL of supernatant liquid, filter (0.45 microns), pipette into preweighed counting vial, reweigh.
- 7. Rinse fines from the soil through a 63 micron sieve with DIW.
- 8. Coarse fraction is washed into an aluminum pan and dried.
- 9. Coarse fraction is weighed into a scintillation vial and counted on Ge gamma-detector.
- 10. Filtered liquid is counted on Ge gamma-detector.
- 11. Activity in dissolved phase and coarse fraction are determined and compared to reference. Activity on fines is calculated by difference.

Reagents used are: water, oxalic acid, citric acid, NaOH and Na₃PO₄.

5.1.3 Attrition Scrubbing

The Methodology for the attrition scrubbing phase of the study was as follows:

- 1. Weigh about 4 grams of dried coarse fraction (> 63 microns) into blender.
- 2. Add liquid at the rate of 10 times the weight of soil.
- 3. Agitate for 6 minutes at a medium to high speed.
- 4. Rinse container walls with water and wash its contents through a 63 micron screen.
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- 5. Transfer contents of screen to aluminum pan to dry.
6. Transfer dried material to pre-weighed counting vial, reweigh and count.

Equipment used is a Hamilton Beach household blender, as recommended by USEPA.

5.2 **Results of Phase III. Soil Decontamination**

5.2.1 Preliminary Screening

Results of the preliminary assessment of the use of reagents and physical methods to decontaminate the soil, shown in Table 3, indicate that both reagents (as represented by oxalic acid) and physical attrition can be used to remove Cs-137 from the coarse fractions of the soil.

Station	Mesh	Treatment	Dry Wt	$Cs -137$ cpm/g	$\%$ Removed
13	80	Reference	5.35	24.3	---
13	80	Vortex	4.13	11.8	51.4
13	80	Citric Acid	5.19	17.9	26.3
13	230	Reference	3.14	96.3	
13	230	Vortex	1.58	64.2	33.3
13	230	Citric Acid	1.78	60.5	37.2

Table 3. Effects of Decontamination, Preliminary Tests

About one third of the activity was removed with each of the two methods used. Activities were not reduced to below the target level but neither method was optimized to provide maximum removal of Cs-137.

5.2.2 Reagent Extraction Results

Having observed that reagents and/or attrition scrubbing can be effective, a more complete assessment of reagents for use in eluting Cs-137 from the soil was then conducted as described above. A broad range of solution properties was employed in this set of experiments: acids, bases, oxidizing agent, reducing agents, complexants, and sources of exchangeable cations (e.g. potassium). These experiments were conducted on the coarse fraction of soil that remained on a 0.063mm sieve after washing with water. They involve exposure to the reagent solutions over a period of three hours on a shaker table. Results of these soil leaching experiments are shown in Tables 4 and 5, for material from the surface at Stations 6 and 13. Results are shown as both activity of Cs-137 and percentage of activity, remaining with the coarse fraction, and the percentage of activity (relative to the starting activity in the coarse fraction) in the liquid (filtered) and the fine-grained fraction. The fines that are reported in Tables 4 and 5 are the result of fines separated from large particles by the reagents. This material is in addition to the fines that were removed during the initial sieving. The objective is to minimize the activity on the coarse fraction, hopefully to below 50 pCi/g. The total activity in each sample at the start of the experiment is also given beneath each table, as a reference. Aqua Regia was used, not as a potential treatment method, but as an indicator of how effective washing with a reagent could be.

No reagent was sufficiently effective in removing Cs-137 from the soil that the target activity on the coarse fraction was achieved. Recalling that these experiments consisted of three hours exposure to the reagent on a shaker table; we conclude that no reagent, even Aqua Regia, is capable of remediating the soil with only gentle agitation. The mechanistic implication is that neither ion-exchange nor sorption on easily soluble coatings (e.g. iron oxyhydroxide) are major mechanism in retention of Cs on this soil.

5.2.3 Attrition Scrubbing

Following these experiments, we investigated the effect of attrition scrubbing using water alone as well as reagents. Attrition scrubbing is a high velocity process that is commonly used to disaggregate soils and to remove fines that are strongly bound to larger particles. The reagents were selected from among those used previously and that performed the best. Results are shown in Tables 6, and 7 for surface soil at stations 6 and 13 respectively. The "whole soil" note at the bottom of the table is the Cs-137 activity on the untreated soil. The "137Cs Before Treatment" column is the activity in the fraction of soil greater than 0.063mm after the initial wet sieving to remove the fines. In some cases we had to wet sieve another batch of soil, to get enough for all the experiments needed. As a result there are different "Before" activities. All samples were subjected to 2 -12 minutes of attrition scrubbing in either water or a reagent. Some samples were scrubbed for 6 minutes, dried, analyzed for Cs-137, and then scrubbed a second time. For these samples the attrition time is noted as $6 + 6$. The percent remaining is based on the activity of the sample compared to the activity on the coarse fraction.

5.2.4 Attrition Scrubbing with Reagents, Results with Surface Soils

Attrition scrubbing was effective in removing Cs-137 from the coarse fraction of surface soils. While scrubbing with water was moderately effective, the combination of scrubbing and 1 molar Na₃PO₄ or 1 molar NaOH was most useful, coming quite close to the target concentration of 50 pCi/g. For surface material from stations 6 (see Figure 5) and 13, removal of Cs-137 by attrition scrubbing, using either water or NaOH solution, followed a power regression curve which became asymptotic after 12 minutes of attrition scrubbing. The NaOH was more effective at removing Cs-137 than was water by about 50 pCi/g. The effectiveness of NaOH was similar to that of $Na₃PO₄$ (both are bases).

5.2.5 Attrition Scrubbing with Reagents, Results for Samples from Depth

Results for stations 27b, 27c, 37d, 39c, 40b are shown in Tables 8, 9, 10, 11 and 12 respectively. Attrition scrubbing with water removed little Cs-137. For all samples, NaOH removed more Cs-137 than water did. Oxalic acid was also effective, having results similar to those of NaOH. Samples from stations 27b and 27c (Figures 6 and 7, respectively) were decontaminated to below the target concentration, but their starting concentrations were relatively low, below 200 pCi/g. The sample from station 39c (Figure 8), also about 200 pCi/g, was decontaminated to 100 pCi/g. However, samples from stations 37d and 40b, which both contained high activities, were not decontaminated to acceptable levels. Both of these soils are termed "recalcitrant", that is, they do not respond well to attempts to decontaminate them. They are discussed in more detail below.

For station 37d the results are shown in Table 10. There was little reduction in the activity of the soil with water; only about 29 *%* of the Cs-137 was removed by 12 minutes of attrition scrubbing. Six minutes scrubbing with the NaOH solution provided somewhat better removal, but the activity was still far above the target. Oxalic acid gave no better results than did water. Because this sample was relatively coarse grained, we thought that addition of particles that were about 0.065mm would provide more attrition. Consequently 0.2 grams of corundum grit was added to 4.34 grams of sand and scrubbed for 6 minutes. No difference was observed between this sample and the water sample. The liquid to solid ratio for each experiment, so far, was 10. We reduced that ratio to 2, so that more particles were available for attrition during the 6 minute scrubbing. The result of this experiment was that about 45% of the activity was left on the soil. A large number of relatively small particles are needed to effectively implement attrition scrubbing. Consequently, some soil taken from depth would be most effectively treated if fines were not removed before the attrition scrubbing step.

For station 40b, results of attrition scrubbing are given in Table 12. These results are very scattered, showing that between 20 and 35 % of the initial activity can be removed with either water or 1 molar NaOH. For both stations 37d and 40b we noted that there are very large differences in activity for the samples tested. This was apparent even after wet sieving and homogenization as a relatively small batch of about 75 grams. The contamination of samples at depth must be a very heterogenous process that results in wide variability of Cs-137 activities on a scale of millimeters.

Figure 5. Duration of attrition scrubbing and its effect on the removal of Cs-137 from soil taken from Station 6.

 $5\overline{a}$

	Coarse Fraction	After Treatment			
	$137Cs$ Before Treatment	$137Cs$ Remaining In Coarse Fraction	$%$ ¹³⁷ Cs Remaining In:		
Reagent	pCi/g	pCi/g	Coarse	Liquid	Fines
Sat EDTA	456.0	360.0	79.0	3.0	18.0
1M Oxalic Acid	456.0	284.0	62.3	3.8	33.9
Sat $Na3PO4$	456.0	287.0	62.9	2.3	34.8
1M Acetic Acid	456.0	366.0	80.2	0.5	19.3
1MCitric Acid	456.0	381.0	83.7	1.5	14.8
1M CsCl	456.0	358.0	78.5	5.3	16.2
1M $CaCl2$	456.0	400.0	87.7	0.5	11.8
1M NH_4F	456.0	212.0	69.8	11.0	19.2
$0.5M$ NH ₄ Cl	242.0	189.0	78.3	9.7	12.0
$1M$ NH ₄ Cl	242.0	179.0	73.9	9.7	16.4
$2M$ NH ₄ Cl	242.0	192.0	79.3	9.9	10.8
$0.5M$ KCl	242.0	192.0	79.3	8.2	12.5
1M KCl	242.0	179.0	73.9	8.5	17.5
2M KCI	242.0	174.0	71.7	6.2	22.1
AquaRegia	456.0	60.5	13.3	37.6	49.1
30% H_2O_2	456.0	397.0	87.1	2.0	10.9
1M $Na2S2O4$	456.0	316.0	69.2	6.1	24.7

Table 4. Soil Washing for $137Cs$ BNL/HWMF Station 6, Surface, Shaker Table, Coarse Fraction

Starting Activity of ¹³⁷Cs in whole soil sample was 1078 pCi/g.

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	Coarse Fraction	After Treatment			
	¹³⁷ Cs Before Treatment	$137Cs$ Remaining In Coarse Fraction	$%$ ¹³⁷ Cs Remaining In:		
Reagent	pCi/g	pCi/g	Coarse	Liquid	Fines
Sat.EDTA	842.0	618.0	73.4	4.5	22.1
Oxalic	842.0	416.0	49.4	6.4	44.2
Sat $Na3PO4$	842.0	460.0	54.7	4.1	41.2
Acetic	842.0	668.0	79.4	0.7	19.9
Citric	842.0	581.0	69.0	3.2	27.7
CsCl	842.0	568.0	67.5	7.6	24.9
CaCl ₂	716.0	589.0	82.3	1.3	16.5
$Na2S2O4$	769.0	589.0	76.6	11.6	11.9
$NH_{4}F$	887.0	468.0	52.8	10.1	37.2
NH ₄ Cl	1018.0	721.0	70.8	10.8	18.3
Kcl	965.0	713.0	73.9	9.2	16.9
AquaRegia	1168.0	389.0	33.3	30.4	36.3
H_2O_2	847.0	544.0	64.2	2.7	33.1
$(CH_3)_4$ NCl	836.0	739.0	88.4	0.6	11.0
NaOH	842.0	302.0	35.9	2.6	61.5

Table 5. Soil Washing for ^{137}Cs BNL/HWMF Station 13, Surface, Shaker Table, Coarse Fraction

Starting Activity of ¹³⁷Cs in whole soil sample was 2812 pCi/g.

Table 6. Soil Washing for $137Cs$ BNL/HWMF Station 6, Surface, Attrition Scrubber, Coarse Fraction

Starting Activity of ¹³⁷Cs in whole soil sample was 1078 pCi/g.

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Table 7. Soil Washing for $137Cs$ BNL/HWMF Station 13, Surface, Attrition Scrubber, Coarse Fraction

Starting Activity of ¹³⁷Cs in whole soil sample was 2812 pCi/g.

Figure 6. Duration of attrition scrubbing and its effect on removal of Cs-137 from soil taken at Station 27b.

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Table 8. Soil Washing for ¹³⁷Cs BNL/HWMF Station 27b, 1-3 feet, Attrition Scrubber, Coarse Fraction

Starting Activity of ¹³⁷Cs in whole soil sample was 1265 pCi/g.

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Table 9. Soil Washing for ¹³⁷Cs BNL/HWMF Station 27c, 4 - 6 feet, Attrition Scrubber, Coarse Fraction

Starting Activity of ¹³⁷Cs in whole soil sample was 350 pCi/g.

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Table 10. Soil Washing for $137Cs$ BNL/HWMF Station 37d, 8 -10 feet, Attrition Scrubber, Coarse Fraction

Starting Activity of ¹³⁷Cs in whole soil sample was 6400 pCi/g.

Table 11. Soil Washing for ¹³⁷Cs BNL/HWMF Station 39c, 4 - 6 feet, Attrition Scrubber, Coarse Fraction

Starting Activity of ³⁷Cs in whole soil sample was 840 pCi/g.

Table 12. Soil Washing for $137Cs$ BNL/HWMF Station 40b, 1 - 3 feet, Attrition Scrubber, Coarse Fraction

Starting Activity of ¹³⁷Cs in whole soil sample was 2800 pCi/g as reported by CDM.

*However, intra sample variability is very large. This sample also exhibits a large anomaly between gross Beta and specific radionuclide concentrations, implying the presence of a radionuclide that was not analyzed.

Figure 8. Duration of attrition scrubbing and its effect on removal of Cs-137 from soil taken at station 37d.

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A preliminary "guess" regarding the reason for this recalcitrant behavior, at least for station 40b, is that this material seems unusually rich in the mineral muscovite (simply from visual observation of the coarse fraction). It is known that Cs sorbs readily onto this type of micaceous mineral (illite especially).

To explore the issue of preferential sorption of Cs by minerals more completely, the gravel componants of samples 37d, 39c and 40b, were separated into several fractions based oh visual appearance of the grains. Most of the grains were quartz, but there were other minerals as well. Results are given in Table 13. One type of gravel, for want of a mineralogical/petrographic description, we have called "black gravel" concentrates Cs-137 preferentially over other grains from the same samples. In station 37d this concentration factor is greater than 1700. Energy dispersive X-ray analysis was done on two samples of the "black gravel" material. One sample consisted of Si, Al, Fe, and Mn. The second sample contained: Mg, Al, Si, K, Ca, and Fe.

Sample	Black	White		Brown Coated Rock Fragment
37d	113,880	65.8	----	
39c	434	13.2		
40b	5,165	47.3	O.	---

Table 13 ¹³⁷Cs Activity (pCi/g) on Gravel Samples from the HWMF

During separation of fines derived by attrition scrubbing, we noticed that the color of fines released from the coarse particles was light gray, compared to the ochre color of the fines from the wet sieving of the "whole" sample. Samples of both types of material were analyzed by X-ray diffraction for mineralogy. Results for both samples are shown in Figure 9.

Both materials are similar, quartz is the dominant mineral with muscovite, kaolinite, chlorite, orthoclase and plagioclase feldspar minerals also present. These results are similar to those obtained by Faust (USGS Bulletin 1156-B, 1963). The sample obtained after attrition scrubbing contains greater quantities of muscovite and the feldspars indicating that these minerals are associated with the larger grains and contain significant Cs-137 activities.

Figure 9. X-ray diffraction scans, showing the mineralogy of the fine (less than 63 μ m) fractions of soil from the "whole" soil and from the material that was removed from coarse grains by attrition scrubbing. The material that was removed by attrition scrubbing is enriched in feldspars and Muscovite. Muscovite preferentially sorbs Cs-137.

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6. DISCUSSION

6.1 Activity/Volume Analysis

The volume of soil as a function of activity was estimated in Pardi, Fuhrmann and Brower, March 13, 1996, (Appendix A). Figure 10 is a semi-log plot indicating that the most contaminated material, with activities greater than 10,000 pCi/g, comprises about 2,000 cubic meters. The very high activities and potential for dispersion of this material may make removal with standard earth moving techniques problematic. As a result, use of new techniques of in-situ solidification with polymers may be appropriate. After small sections are solidified in the ground they can be removed with much less risk than if the soil were dispersible.

We found that we can decontaminate surficial soil with activities of about 3000 pCi/g. The volume of contaminated soil containing between 3000 pCi/g and the target of 50 pCi/g is illustrated on Figure 10. We believe this is the regime in which soil washing will be most effective. If so, the volume of contaminated soil to be washed will be about 30,300 cubic meters. This is calculated by subtracting the volume of soil that is greater than 3000 pCi/g $(5,300 \text{ m}^3)$ from the total volume of soil that requires remediation $(35,600 \text{ m}^3)$. In this activity range, based on our grain size analysis, we found that the average fines content was 35.5 Wt %; therefore about 19,700 cubic meters of decontaminated soil can be returned to the site. The remaining 10,600 cubic meters, the concentrate, will require disposal. This material will be a mixture of fines (e.g. clay minerals) and water clarification media, with a water content of about 50 Wt %. If activities are greater than $3,000$ pCi/g, the concentrate may require solidification prior to disposal.

Analysis of activity and volumes for the samples that have been examined in detail, are shown in Tables 14 a, and b. Here we have calculated the fraction of two surficial soil samples that could be returned to the site (based on a kilogram sample), assuming that the target is between 50 and 100 pCi/g. These tables compare the mass of sample remaining after each treatment and the activity associated with that mass, after wet sieving and two cycles of attrition scrubbing in $Na₃PO₄ (NaOH would be similar) solution. From this analysis, about 60 Wt % of the surface soil$ can be returned to the site, having met the activity target.

Soil from depth, however, appear to be more difficult to decontaminate than those from the surface. This observation may be an artifact of the samples that were available for analysis because there was a wide range of activities. As a result, most of the "hotter" samples were above the 3000 pCi/g limit for application of soil washing. However, even some lower activity samples such as 39c were not decontaminated as readily as we had hoped for.

¹³⁷Cs Activity (pCi/g)

Figure 10. The volume of soil at the HWMF as a function of Cs-137 activity, as calculated with activities in soil cores that were contoured with a kriging process and then adjusted for conformance with a site "walk-over" survey with a NaI gamma detector.

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To achieve the level of decontamination required, the use of reagents is necessary. Reagents that can be used most effectively are $Na₃PO₄$ and NaOH. Both have similar efficiency. The cost of NaOH is significantly less than that of $Na₃PO₄$, so in later experiments we used NaOH. While oxalic acid had similar results for some samples it was ineffective for others.

The relationship between soil activity and the activity at which the target is set (we have assumed 50 pCi/g) is a powerful driver of volume. Figure 11 shows this relationship for both untreated soil and soil that was washed. Above the assumed cut-off of 50 pCi/g the total volume requiring treatment is about 35,000 cubic meters. Raising the cut-off provides significant reduction in soil volumes. For example changing the cut-off to 100 pCi/g reduces the volume to about 30,000 cubic meters. To get the curve for "after soil washing", the volume/activity curve for the untreated soil was multiplied by a constant percentage of 35 *%* which is the average percentage of fines in the 50 -1000 pCi/g activity range of the soil.

Description	Weight Remaining (g)	Activity (pCi/g)
Whole Soil	1000	1078
Wet Sieved $(63 \mu m)$	670	455
$1st$ Attrition Scrub/Na ₃ PO ₄	580	78
$2nd$ Attrition Scrub/Na ₃ PO ₄	560	54

Table 14a. Estimate of Soil Washing Effectiveness: Station 6, Surface.

Table 14b. Estimate of Soil Washing Effectiveness: Station 13, Surface

Description	Weight Remaining (g)	Activity (pCi/g)	
Whole Soil	1000	2812	
Wet Sieved $(63 \mu m)$	721	842	
$1st$ Attrition Scrub/ $Na3PO4$	692	136	
$2nd$ Attrition Scrub/Na ₃ PO ₄	674	74	

7. COST ANALYSIS

To estimate costs of soil washing we have used estimates of soil volumes derived from Figure 10 showing:

- total volume with activities greater than the assumed target of 50 pCi/g,
- volume with activities greater than $3000 \frac{\text{pCi}}{\text{g}}$.

Other factors that are important in determining cost are:

- \bullet cost per m³ of soil washing,
- \bullet cost per m^3 of transportation to the disposal facility (Hanford),
- cost per m³ for disposal (there are surcharges for high activity waste, but we have ignored them since only a small fraction of the waste should be over these activities),
- cost of containers (assuming B-25 boxes),
- cost of solidification of material that exceeds activities of 3,000 pCi/g. (Includes soil and sludges containing fines removed from the soil).

Details of the analysis are given in Appendix D, as are details of the analysis for the baseline against which soil washing is compared. The baseline consists of disposal of all soil having activities greater than 50 pCi/g. It also includes solidification of soils with activities greater than 3,000 pCi/g. The volume of high activity soil is the same in both scenarios. Costs for various parameters were obtained from vendors or personnel in the BNL Waste Management group.

As described in Appendix D, the cost of the baseline scenario is approximately 80 million dollars, driven primarily by the costs of transportation and disposal. We have assumed that the soil is all shipped to Hanford by truck, and that it is packaged in B-25 boxes. Solidification, with its requisite increase in volume, adds to the cost.

Appendix D also contains analysis of soil washing as a means of reducing the volume of contaminated soil that must be disposed of from the HWMF. As discussed in Appendix D, the cost of both soil washing and the baseline scenario are dependant on the volume of soil that needs to be remediated, and that volume in turn, is dependant on the Target Performance Level (TPL). The TPL value is the activity of Cs-137 above which remediation is required. While the TPL is not known exactly for the HWMF, we have used 50 pCi/g. The final value may be more or less, depending on the future land-use of the site. Consequently, we have plotted cost as a function of the TPL in Figure 12, for both soil washing and the baseline.

The cost for the soil washing option is approximately 53 million dollars at the 50 pCi/g TPL. With higher values of TPL the volume, and therefore the cost, decrease. At a TPL of 110 pCi/g, for example, the cost is 48 million dollars. In contrast, the cost for disposal without soil washing is 81 million dollars at 50 pCi/g and 70 million dollars at 110 pCi/g.

The cost savings of soil washing over the baseline is due to the smaller volume of soil that must be shipped for disposal. Some cost components are similar for both scenarios. These include excavation costs and solidification costs for soils with activities that are greater than 3,000 pCi/g. This is necessary because some soil contains more Cs-137 than can be adequately decontaminated; consequently in both cases, it would need to be solidified and sent to disposal.

Evidence from the "activity vs soil volume" work indicates that much of the activity is on a relatively small volume of soil; about $5,300 \text{ m}^3$ are over 3000 pCi/g . Given the apparent difficulty in cleaning the Cs-137 from the coarser fraction of the hottest samples, and given the very high activity in a few locations, consideration should be given to solidifying some of the soil in-place and subsequently removing the most contaminated material as a solid block. This would help reduce potential exposure to workers during remediation. Applying soil washing to the remainder of the material would then be easier because the "hot spots" will have been removed.

Both scenarios discussed in this report have very large costs associated with them. The data generated in this study also may be used to support another concept, specifically: removal of "hot spots" with subsequent capping of the site with clean soil. The laboratory study performed to obtain basic information on the effectiveness of different reagents and mechanical decontamination also shows that essentially all of the activity remains fixed on solid phase materials. We observed Cs-137 in wash water only at the location with the greatest activity. In all other stations we found none in solution. When reagents were applied, Cs-137 was observed in solution. For example, at station 6 (see Table 4), between 0.5 and 38% of the activity that was originally present on the soil, was moved to solution. The ability of some of the reagents to mobilize Cs-137, even to a limited extent, may be taken as an indicator of potential bioavailability. On the other hand, considerable effort, in the form of attrition scrubbing, was required to remove particulates that adhered to larger particles. This material is essentially non-transportable. Indeed, in some cases only a fraction of the activity was removed by exposure to Aqua Regia. Since Cs-137 generally is not able to transfer to the aqueous phase without reagents, a strong case can be made that the contaminated soil can be left in place and the Cs-137 allowed to decay without endangering the groundwater. This analysis is based on data for Cs-137. Since Sr-90 activities were so much lower than Cs-137 in all samples analysed, and given the dilution factor for soil washing, we determined that Sr-90 would be below detection limits in liquid extracts from all samples but 37a. This one sample did show Cs-137 in the wash water. Since Cs has much higher K_d values than Sr, a small quantity of Sr-90 would be expected in the aqueous phase.

8. CONCLUSIONS

We have completed grain size separations for 15 stations by wet sieving a broad range of samples with activities varying from near background to 810,000 pCi/g. Results indicate that removal of the fine grained fraction of soil from the HWMF is an effective method for decontamination of samples that contain activities to about 3,000 pCi/g. A combination of reagents and attrition scrubbing is needed to implement removal of fines that are associated the larger particles. When soil is contaminated to much higher levels, Cs-137 is distributed throughout the size classes; often at activities that exceed the target. In these cases the soil may not be adequately cleaned. Surficial samples containing 2800 pCi/g have been successfully decontaminated. Samples with higher activities, particularly those from the subsurface, are more "recalcitrant" and soil washing becomes less effective as the activity increases.

Using the data generated in the laboratory studies (e.g. activity of fines, grain size to activity relationship) and costs obtained through several vendors, we have assessed the effectiveness of soil washing and bounded its limitations regarding the volume of material that probably cannot be treated with this technique. We have tried to make our cost estimate as complete as possible, including the full cycle of costs such as transportation and disposal associated with soil washing. In comparing soil washing to the baseline (excavation and disposal of all soil with activities greater than the TPL) we have kept our assumptions and unit costs on the same basis so that the comparison is valid. The final cost for the baseline is 81 million dollars. In contrast the estimate for soil washing is 53 million dollars. In reality, the bulk of the costs are not for the processing itself but for transportation and disposal of residual material and soil that cannot be decontaminated.

Soil washing provides a significant cost savings over the baseline. This savings varies with volume of soil requiring remediation and therefore with the inverse of the TPL (since as the TPL increases, the volume of soil decreases). At a TPL of 50 pCi/g the saving is 27 million dollars, while at a TPL of 110 pCi/g the saving is 22 million dollars.

Figure 11. The volume of contaminated soil from the HWMF as a function of the Target Performance Level for Cs-137.

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APPENDIX A

Estimate of the Volume of Contaminated Soil at OU-1

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ESTIMATE OF THE VOLUME OF CONTAMINATED SOIL AT OU-1

March 13, 1996

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γġ James Brower

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This report presents estimates of the volume of contaminated soil at the Hazardous Waste Management Facility (HWMF) at Brookhaven National Laboratory. The estimates are based on:

- analysis of $137Cs$ and $90Sr$ concentrations in soils taken from a series of geoprobe borings,
- contours of concentrations around those borings as calculated with a Kriging routine,
- contours around the same points (at the ground surface) from a radiological survey using a shielded Nal gamma spectrometer (conducted by RadChem).

Data Sets

The data used for these analyses are for surface (0.00 to 0.50') samples taken from 53 borings within HWMF and from 6 additional surface samples collected by hand during the summer of 1995. The location of the borings can be found on a BNL map entitled "Proposed Soil Borings: Operable Unit - lHWMF:/home3/OUl/HWMF-SOIL-BORING" DATED 6/1/94. Most of the original set of borings also include samples taken down to a depth of 10'. Volume estimates reported here are for the surface horizon (0-0.5 feet) and the total site. The estimate for the whole site is based on cores taken to 10'. Considering the levels of ¹³⁷Cs at 10' at several locations, we can assume that the soil is contaminated to greater depths. Additional sampling will be required to define the depth to which the contaminant has traveled.

Local Coordinate System

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The analyses reported here use a local coordinate system employed by BNL & CDM Corporation. The axes of this local system closely parallel the fence line of the HWMF. In the figures that follow_y values for the north-south and east-west axes have been converted to meters from the English units given on the BNL-HWMF map, and the north-south coordinate values have been inverted from the map orientation by first subtracting the plotted coordinate from 700' and then converting the difference to meters. This conversion was required by the gridding software.

The coordinates (in meters after the above conversion) of all 59 samples used in these analysis and the measured concentration of ^{137}Cs in pCi/g are given in Table 1. The positions of the borings and hand-collected surface samples are shown in Figure 1.

Methods

The analyses were performed using Surfer Version 4 by Golden Software. The input data of geographic coordinates and ¹³⁷Cs values was first converted to a regular grid of computed values. The computed grid had a spacing of 5 meters in both the N-S and E-W directions. The dimensions chosen correspond closely to the dimensions of the fenced-in area of HWMF, namely 275 meters east-west and 215 meters north south, with the northern boundary along .the centerline of Brookhaven Ave.

The borings and surface samples on which ¹³⁷Cs measurements have been made were not taken at locations defined by some regular grid pattern. Instead, their location was chosen specifically to define areas of maximum or minimum contamination. As a consequence of this sampling scheme the distribution of the data is, not surprisingly, statistically non-normal both in space and level of contamination. Figure 2 is a histogram of the common log of ¹³⁷Cs frequency. The data are only roughly log-normally distributed. The fact that the data are not stochastic has important implications for procedures used to estimate contaminant volumes.

In order to estimate the volume of surface material that is contaminated at any given range of ¹³⁷Cs level, it is first necessary to extrapolate, from the given data set, a matrix of regularly-spaced grid elements. Extrapolation from given locations to a regular grid requires the calculation of estimated values based on levels at points that surround the grid point. This regular grid has been used to generate a series of contour maps and 3D diagrams, on which the basic characteristics of the distributions can be observed.

Several different schemes have been developed to perform this type of estimate, and, as discussed below, Surfer provides the capability of using either Inverse-square, Kriging, or Minimum Curvature methods of generating the grid from which volumes are calculated. In addition, Surfer permits both the search radius (the distance over which the routine will look for measured points in order to produce the grid value), and number of nearest neighbors to be specified. A log transformation of data also was suggested because of the distribution noted above. In order to determine the sensitivity of the volume estimates to variations in these three parameters and, also, the gridding methodology, a number of analyses were performed that are described below.

Search Radius

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The default value for search radius (450 meters) used by Surfer is calculated to include all data points. From all those points a number of nearest neighbors can be selected up to and including all data points. It was found that varying the search radius down to about 100 meters had very little effect on the calculation of the grid points. Significant differences could be seen in the topology of the gridded surface at a search radius of 50 meters, while below a search radius of 25 meters, each method of gridding had difficulty calculating values and closing contours. Since the average distance between measured points was between 30 and 40 meters, a search radius of 35 meters was selected for the analyses that follow.

Table 1.

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Locations and Activities of ¹³⁷Cs used in this Analysis

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Table 1. (Continued)

Locations and Activities of ¹³⁷Cs used in this Analysis

Number of Nearest Neighbors

Surfer will include a variable number of nearest neighbors in the grid point calculation up to some specified number, if those points fall within the search radius. This number was varied from 10 to 5 to 2. As the number of nearest neighbors decreased the average slope of the gridded surface about each of the original data points increases. Despite the marked difference in appearance among the three maps, volumes calculated from either 10 or 2 nearest neighbors were negligibly different. Results are shown in Figures 3 and 4 (a 3-D plot) using a 35 meter search radius and 2 nearest neighbors.

Methodology

In order to evaluate the effect that different methodologies would have on the results of the volume calculations two fundamentally different methods were compared: Inverse Square Distance weighting, and Kriging. The Inverse Distance method is a simple, straightforward technique that estimates values at grid points by weighting the contribution of a specified number of surrounding data points within a search radius based on some power function of that distance. A square distance approach was taken here. Kriging bases its weighting function on a statistical analysis of a number of surrounding data points by employing a scheme termed a variogram. Essentially the weight given to surrounding values is a function of the local variance among those values. The results of Kriging are shown in Figures 5 $\&$ 6 and are fairly similar in appearance to the map shown in Figure 3 for the Inverse Distance method with 2 nearest neighbors. This implies that the high variance within the input data set limits the Kriging method to approximately two nearest neighbors.

Log **Transformation**

As was discussed earlier, the data are, at best, (distributed in a log-normal fashion. This suggests that a log transformation of the data might decrease the influence of the few very high values and render a more reasonable distribution of data across the site. The results of a log transformation are shown in Figures 7 & 8, which clearly shows a suppression of the influence of high values around the Oil/Water Separator and other spills in that area. The log transformation also suggests that lower-level contamination of the site is more widespread than would appear from the contouring of non-transformed data. The 50 pCi/g target activity appears on the plot as log 1.7.

Samples at Depth

The methods presented above were used on surface samples, however we know that contamination extends to depth at some locations at the site. Fewer samples are available for this analysis. Moreover the sampling was not continuous, with samples taken only at the following intervals: $0-0.5$ ', $1-3$ ['], $4-6$ ', and $8-10$ '. As a result, measured activities for about 35% of the soil column is missing. Estimates of volumes as a function of activity per gram were made based on this limited data. Because of the few samples available the search radius had to be set at a higher value than that at the surface, as a result these estimates may be high.

Figure 9 a-d is a set of 3-D plots showing the distribution of ¹³⁷Cs over the HWMF at the four levels described above. Note that the scales on the z-axes are different for each plot. The area around the Oil/Water Separator is contaminated to significant activities from the surface to the bottom of the sampling intervals (at 10'). There are areas at depth where contamination apparently is not supported by surficial contamination. For example, ¹³⁷Cs activities at $1 - 3'$ are lower in many locations around the Oil/Water Separator than they are in the 4 - 6 ' and the 8 - 10' samples. This appears to be most pronounced east of the main peak in activity.

Figure 10 a-d is a set of 3-D plots showing 90 Sr activities. These levels are about 500 times lower than the ¹³⁷Cs activities, but in general they mirror the distribution of ¹³⁷Cs. At the surface ${}^{90}Sr$ is a little more spread out from the Oil/Water Separator than is ${}^{137}Cs$. At the 1 - 3' interval activities are low, with all samples below the 50 pCi/g limit. However at the 4 - 6' interval activities are slightly increased above those of 1- 3', especially to the north-east. It is noteworthy that the location of the greatest contamination in the 0.5 , $4 - 6$ and $8 - 10'$ intervals is almost background in the 1 - 3' interval. The cause for this is unknown.

Comparison with" Walk-Over" Survey Data

The estimate of volumes at depth is fraught with uncertainty and this uncertainty is intensified by the spread in samples both laterally and vertically. As a result the portion of the site that represents by far the greatest volume is the least characterized. A significant issue is the validity of using a data set that was purposefully biased to "hotter" samples. This in turn bias our estimates of activity vs volume. Aside from embarking on a new sampling and analysis program using a random pattern or a grid pattern, the next best approach was to try to use data from a detailed surface activity survey to normalize the estimate of activity distribution around the sampled locations.

In order to better estimate the volume of contaminated soil present at the site, it was necessary to calibrate the contours done by the Kriging program with some measure of activity distribution around the "hot spots". This would help relieve some of the sampling bias toward the "hot spots". Figure 11 shows data from the "walk-over" survey of the HWMF, done by ChemRad, using a shielded Nal gamma-ray detector. We compared this plot to Figure 7 , which is the results of Kriged data from the geoprobe survey (combined with our surface samples). The locations of most "hot spots" match on the two figures. The major difference between the two figures is that the contours on the "walk-over" survey are much closer together. This means that ¹³⁷Cs activity falls to low-levels much more rapidly based on the "walk-over" survey than it does in the Kriged data. Because the "walk-over" is based on a very large number of surface count rate measurements, we believe that its trend of showing that the contamination is spatially more discrete (compared to the Kriging) is valid.

To assess the difference in volume between the two approaches, we applied a simple approach, necessitated by the lack of electronic data files for the "walk-over" survey. We copied that figure showing the contours for the "walk-over". From one copy we cut out the

section containing the entire HWMF, which is an area of $62,000$ m². From the other copy we cut out the sections with count rates greater than log 4.5. Both were weighed, and from the weights a ratio was determined. This applies only to the surface. The contaminated area was 20.3% of the total area or $12,600$ m². Multiplying by 0.15 (since the surface samples were taken in the top 15 cm), we get a volume of 1890 $m³$. This is 0.25 to 0.33 of the volume estimated from the physical samples. From the Kriged data we estimated about $110,000$ m³ of soil that was contaminated above 50 pCi/g. Applying the factors of 0.25 and 0.33 to this estimate for the total volume of contaminated material we get between 27,500 and 36,300 $m³$ of contaminated soil. Considering the closer contours on the "walk-over" survey, we believe this estimate to be reasonable. One concern with this approach is the use of the log 4.5 contour. We do not know how this correlates to pCi/g in the soil. However CDM has recently provided an analysis of count rate from the Nal detector as a function of soil activity. Based on a linear relationship (y = $1.787X - 5.644$, where y is in pCi/g and x is the count rate from the Nal detector), the log 4.5 contour corresponds to 250 pCi/g.

Results and Discussion

Results shown in Figures 4 and 6, (which have linear Y-axes) shows contours for surface contamination only in the upper left area of the HWMF, around the Oil-Water Separator. This is an artifact of the very broad range of activities found at the site and the contours selected by the program; the lowest contour is 40,000 pCi/g. Lower activities can be seen in Figures 7 and 8 which use log scaling for the y-axes. In fact, of the 59 surface samples taken in the area, half exceed the 50 pCi/g target.

The surfaces generated from the gridding routines were subjected to a volume analysis. This analysis consisted of taking "slices" through the gridded volumes at specified levels of ¹³⁷Cs contamination, calculating the volume assuming uniform contamination across the entire 0.5' depth of the surface layer, and plotting the results as volume vs. level. These curves give the volume that would have to be treated to reach any specified level of contamination between 10⁶ and 1 pCi/g. Curves of the volume relationship for non-transformed, log-transformed, and volume estimates calculated from the Kriging Analyses are shown in Figures-12,13 and 14 for surface material. From these figures, comparing the volume of soil for a cleanup level of, say, 50 pCi/g, both the non-transformed volume curve and the Kriging curve give an estimate that 7000 cubic meters out of the site total (to a depth of 0.5 feet) of about 9000 cubic meters would have to be treated to reach this level. The log-transformed analysis reduces this estimate somewhat to approximately 6000 cubic meters. Note, however, that the steep slope of the log-transform volume curve above 1000 pCi/g suggests that, despite the limitations of all these gridding schemes, the volume of highly contaminated surface soil is relatively small (about 2500 cubic meters) for the top 0.5' of the site.

In addition to surface samples, an analysis was done on samples taken at depth $(1 - 3)$, 4 - 6' and 8 - 10'). At the 50 pCi/g target the estimated volume is about 70,000 cubic meters. Based on the spread of the depth intervals at which samples were taken, this estimate includes only about 65 % of the total volume. Accounting for the missing 35 %, the total volume of

contaminated soil requiring remediation or disposal would then be about 110,000 cubic meters. Adjusting this value with the factor of 3 obtained by comparing the Kriged data to that in the "walk-over" survey, the volume is reduced to a range between 27,500 and 36,300 m³ . **Our estimate for the total volume of contaminated soil at OU-1 is 35,000 m³ .** This high end of the range is taken because we know that there must be contaminated soil below the lowest sampling depth. These results are summarized in Figure 15 which shows the total volume of soil as a function of activity, for both the Kriged data and the data that was adjusted with the walk-over survey information. Using the 50 pCi/g cutoff for soil-washing/separation as the lower limit and 3000 pCi/g as the upper limit for effective soil-washing, **we estimate about 30,000 cubic meters of soil could be treated by soil-washing.**

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Stations at which samples were taken at the BNL HWMF. Stations labeled with an "S" are surface samples only. Figure 1. Stations labeled with a "B" are geoprobe borings from a survey by CDM.

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Distribution of ¹³⁷Cs in Surface Soil BNL OU-1

Figure 2. Histogram showing the log-distribution of ^{137}Cs for surface samples from the Hazardous Waste Management Facility.

Figure 5. Contours of ¹³⁷Cs using a 35 meter search radius and 10 nearest neighbors, with Kriging gridding.

Figure 7. Contours of ¹³⁷Cs using a log scale for the contours, showing more detail.

Figure 8. Three-D plot of contours from figure 7. The 50 pCi/g cutoff is 1.7 on the log scale, showing that much of the site is above that cutoff. This is for the surface 0.5' only.

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Figure 9 A-D. 3-D plots showing activities of $137Cs$ at four depth intervals.

Figure 10 A-D. 3-D plots showing activities of ^{90}Sr at four depth intervals.

Figure 11. Contour chart of data from the "walk-over" survey conducted by ChemRad using a shielded Nal gamma-detector. The contours are closer together than on Figure 7 (kriged data from cores), illustrating that concentrations of ¹³⁷Cs decrease more rapidly, spatially, than indicated by Figure 7.

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Figure 12. Estimate of soil volumes as a function of contamination for the surface 0.5' only. Based on non-transformed data.

Estimate of soil volumes as a function of contamination for the surface Figure 13. 0.5' only. Based on log-transformed data.

Figure 14. Estimate of soil volumes as a function of contamination for the surface 0.5' only. Based on Kriging analysis.

Figure 15. Total volume of soil at the HWMF as a function of contamination, including all samples at depth. The treatable volume is delineated by the 50 pCi/g line and the 1000 pCi/g line.

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APPENDIX B

Grain Size Analysis of Soils from the BNL Hazardous Waste Management Facility

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APPENDIX C

Cs-137 and Sr-90 in Soil from the BNL Hazardous Waste Management Facility

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Cs-137 and Sr-90 in Soil from the BNL Hazardous Waste Management Facility

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Cs-137 and Sr-90 in Soil from the BNL Hazardous Waste Management Facility

C5

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APPENDIX D

Economic Analysis of Soil Washing

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ECONOMIC ANALYSIS OF HWMF OU-1 TREATMENT ALTERNATIVES

The following spreadsheet details the cost estimates analysis performed for soil washing at the Hazardous Waste Management Facility compared with the cost of simply removing and disposing of the contaminated soils. A written explanation of the assumptions and calculations is provided below.

A key parameter in this analysis is the Target Performance Level (TPL) which is the specific activity of the soil below which the soil does not need to be remediated. The TPL is based (among a number of other factors) on the future land use of the site as well as the acceptability of allowing an interim period of institutional control pass before the TPL must be reached. In effect this allows radioactive decay to help attain acceptable concentrations. The TPL for the HWMF has not been decided yet, although 110 pCi/g has been calculated as a TPL by using 75 mRem/yr under the Suburban Residential scenario.

SOLIDIFICATION VOLUME ANALYSIS:

The objective of soil washing is to return the largest possible volume of soil to the site as "clean" material, while generating the smallest volume of concentrated residuals. "Clean" as used here denotes an activity that is below the TPL. As a result, estimates of solidification and disposal volumes are key to providing accurate cost information for the two treatment alternatives being considered. These volumes can be extrapolated from the volume versus activity data detailed in the report "Estimate of the Volume of Contaminated Soil at OU-1". Key assumptions and experimental approximations include:

- Volume of soils to be excavated is approximated to be all soils with activity levels greater than the TPL plus 3000 cubic meters of soil. This attempts to account for "clean" soils (below TPL) that would to be removed due the accuracy limitations of excavating equipment.
- A 3000 pCi/gram Upper Activity Limit is also taken as the limit above which soils will require solidification. This activity is also the limit above which we found that soil washing did not provide adequate decontamination.
- Percentage of concentrated fines generated from soil washing is approximated to be 35% based on bench-scale studies of soil samples taken from the site.
- Ratio of activity in the "whole" soil sample versus activity in the concentrated fines is taken to be 2.25. This number is based on laboratory bench scale studies of soil samples taken from the site (see Table D.I).

Based on the above approximations, a plot of volume of fines as a function of Cs-137 activity in the fines was determined from volume and activity data from the plot of "whole" soil as a function of total Cs-137 activity (see Figure D.I). This is accomplished by calculating 35% of

the total volume at each data point to determine the volume of concentrated fines and multiplying its corresponding activity by 2.25, which gives the activity of the fines. Estimates of solidification and/or disposal volumes for each remediation alternative can be extrapolated from these two curves. From the whole soil plot in Figure D.I, the total volume of soils above the $3,000$ pCi/gram upper activity limit was found to be $5,300$ m³. This volume represents the total volume requiring solidification if soil washing is not considered. If soil washing is performed, an additional volume of material requiring solidification is generated within the concentrated residuals that are separated from the coarse (lower activity) material. The volume of additional material, that requires solidification, can be estimated from the fines curve in Figure D.I. The total volume of concentrated fines above the 3,000 pCi/gram activity limit is $3,000$ m³, which represents the amount of fines for the entire site that would require solidification. However, since soil with activities greater than 3,000 pCi/g cannot be treated by soil washing, the fines in this fraction will not be separated but will be solidified with the "whole" soil having activities greater than $3,000$ pCi/g.. This fraction is 1,900 m³ (calculated by multiplying 5,300 by 0.35) and needs to be excluded from the total volume of fines by subtraction from the original $3,000$ m³ of fines. Therefore, the amount of concentrated soil washing residuals requiring solidification is $1,100 \text{ m}^3$ and the total volume requiring solidification after soil washing is estimated at $6,400 \text{ m}^3$. Note that the estimated volumes requiring solidification are independent of the Target Performance Level.

Disposal volumes can vary depending on the TPL. For varying TPL's of 200,110, 70, 50, 20 and 10 pCi/gram the volumes of contaminated soil which are not solidified but require disposal are calculated and shown in the disposal volume table. The total soil volume above each TPL is read off the whole soil plot in Figure D.1. Subtracting $5,300 \text{ m}^3$, as the volume of soil for which soil washing is not feasible, results in the effective soil-washing volume. This volume is also the total volume requiring disposal (without solidification) if soil washing is not considered. The volume of concentrated fines after soil washing represents 35% of the effective soil-washing volume. Of these concentrated fines we determined that $1,100 \text{ m}^3$ would require solidification. Subtracting this solidification volume yields the volume of soil requiring disposal (without solidification) after soil washing. These solidification and disposal volumes are applied to the cost of treatment, solidification, transportation and disposal *to* provide an estimate of the total project cost for each alternative.

ECONOMIC ANALYSIS:

The detailed cost estimates shown for each of the two treatment options are based on a 50 pCi/gram TPL. Treatment costs for both alternatives are extrapolated from soil washing cost data taken from Alternative Remedial Technologies Inc. (ART Inc.) for a typical site sized at $38,200$ m³ (50,000 yd³). The transportation and disposal costs are based on a capacity of 40,000 lbs.per truckload and a 1.4 tons/cubic yard bulk density. Using HWMF's estimates of transportation costs at \$10,000 per truck load, and a $$22/ft³$ disposal cost, an estimated \$1,695/m³ (\$48/ft³) was calculated for transportation and disposal at the Hanford Site. Container costs can also be significant, considering the large volumes of soils requiring disposal. The cost of new B-25 containers, as provided by Scientific Ecology Group (SEG), is

\$500 per container. Cement solidification costs were estimated at \$70/m³ of dry soil with an 100% estimated volume increase.

Baseline, Disposal without Soil Washing

Option 1. Without Soil Washing: The itemized treatment costs shown were adjusted from ART's soilwashing data to reflect the added expense of remediating a site with high activity. Also reflected in the treatment cost data are the reduced cost of analysis and office expense compared to that required by soil washing. Each itemized cost was summed and the total divided by 38,200 m^3 (50,000 yd³) to arrive at a \$98 treatment cost per cubic meter. Note that this is not completely valid since not all of the itemized costs scale with the volume of material to be remediated. However, the error of scaling all treatment costs to the total site volume is small enough as not to effect the results significantly while simplifying calculations. A total disposal volume of 40,900 m^3 is estimated by adding the volume of solidified waste to the volume of soil being disposed (without solidification). Total costs for simple excavation and disposal are:

For a **Total Project Cost of \$81.000.000**

Soil Washing

Option 2. Soil Washing: The itemized treatment costs shown were adjusted from ART's soilwashing data to reflect the added expense of remediating a site with high activity, resulting in an estimated cost of \$272 per cubic meter. A total disposal volume of $22,300$ m³ was estimated by summing the volume of solidified waste and the volume of soil being disposed (without solidification). Total costs for soil washing are:

For a **Total Project Cost of \$52.800.000**

Cost estimates for a 200, 110, 70, 20 and 10 pCi/g TPL were also performed with the results shown in Tables D.2 and D.3, entitled "Without Soil Washing Summary" and "Soil Washing Summary". The data from these two tables are plotted as cost versus TPL in Figure 12, in the body of the report.

Table D.I

Activity on Whole Soil Samples and on Fines Separated From Them

Figure D.I. Estimates of the volume of "whole soil" and of its associated fines, as a function of Cs-137 activity.

Table D.2

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Cost Estimates as a Function of Target Performance Level Baseline: Without Soil Washing

Table D.3

Cost Estimates as a Function of Target Performance Level

Soil Washing

OUI - Economic Analysis of Treatment Alternatives

Volumes Analysis:

KEY ASSUMPTIONS

SOLIDIFICATION, VOLUME ESTIMATES

DISPOSAL VOLUME ESTIMATES

Economic Analysis:

KEY_ASSyMPTIQNS

Volume

- Based on calculations detailed in the above Volume Analysis for a TPL = 50 pCi/gram

Treatment Cost

Treatment Cost numbers extrapolated from soil washing cost estimates taken from Alternative Remedial Technologies Inc. and are based on a total site volume of:

Transportation and Disposal DATA

Option 1: Without volume reduction through soil washing

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Treatment Costs

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75 (\$/cubic yards)

TOTAL Volume Estimates

Total Project Cost

Option 2: With Soil Washing

Treatment Costs

TOTAL Volume Estimates

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Total Project Cost

