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Pacific Northwest
National Laboratory
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Radionuclide Activities in Contaminated Soils: Effects of Sampling Bias on Remediation of Coarse-Grained Soils in Hanford Formation

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Executive Summary

The Remedial Action Sampling and Analysis Plan (RASAP) for the Hanford Site's 100 Area soils requires sampling and analyzing the soil material for contaminants of concern (COC). However, the RASAP does not include a protocol for analyzing only a fine-grained fraction (sand, silt, and clay) and then relating the measured COC activities and concentrations in this fraction to those in the predominantly coarse-grained (gravel) whole soils in the 100 Area. Selectively sampling only the finer fractions of a coarse-grained soil would introduce a systematic bias that results in consistently overestimating the actual activities and concentrations of COC in whole soil. Such sampling bias, if uncorrected, would result in over-remediation (remediating below the remedial action goal [RAG] levels) of these soils. Therefore, we conducted a study to assess the scope of this problem by analyzing available particle size-COC activity and concentration data for 100 and 300 Area soils, and determined the effects of sampling bias. Our recommendations include sampling protocols and methods to determine the COC activities and concentrations of the whole soil based on the measurements conducted on a finer size fraction of the whole soil.

The important findings of our investigations are that

1. In gravelly soils (100 and 300 Areas), the activity and concentration of any COC per unit mass in any finer fractional soil material was consistently biased higher than those of the same COC in the whole soil (all grain sizes sampled). The degree of sampling bias for each COC could be quantified as a sampling bias factor (SBF) that relates the activity and concentration in any finer size fraction of a soil to that in the whole soil.

2 The SBF for any COC in a soil increases with decreasing particle size sampled. SBFs for different COC in a soil can vary significantly (up to an order of magnitude) depending on their chemical properties and affinity for specific mineral surfaces.

3. SBF for a single COC in soil samples obtained from different locations from the same waste site can be significantly different (two to three times higher than the lowest value), reflecting spatial variability engendered by the source, the type, and the distribution of contamination.

4. The SBF for a COC in soils from different waste sites were different, and these values were affected by the soil texture, the degree, the source, and the distribution of contamination.

5. Waste site soils would be over-remediated (remediated below remedial action goal values) without SBF corrections. SBF corrections could be made by using corrected remedial action levels (CRAL) computed from RAG and SBF (for selected particle-size finer) for each COC.

6. The soils in the vicinity of the 116-C-1 waste trench would be over-remediated (excavation of ~57% to 280% of excess soil volume) without appropriate SBF corrections for contaminant radionuclides ^{60}Co and ^{152}Eu.

Based on our analyses and conclusions, we recommend that

1. A sampling grid should be established (using a geostatistical basis) for each waste site, and representative whole soil sample should be drawn from selected depth intervals as specified in ASTM C-702. The quantities of whole soil selected for analysis should be based on the standard practice as specified in ASTM D-2487-98 (Appendix A).

2. The particle size and COC distribution in soil samples should be determined by wet sieving (ASTM D 422), and each soil fraction of each sample should be chemically and radiologically characterized.

3. The SBF for each COC for a specified particle size finer and CRAL values for each COC in each sampling grid should be computed.

4. During remediation, representative soil samples for the same specified particle size finer should be drawn for monitoring the activity/concentration of each COC and sample grid, and relevant COC-specific CRAL values should be used to establish the limits of remediation.

Acknowledgments

The authors wish to thank R. Jeff Serne of the Pacific Northwest National Laboratory and Dr. Scott Petersen of Bechtel Hanford, Inc. for providing technical reviews of this document. We also thank Sheila Q. Bennett for editorial assistance and Lila Andor and Kathy Neiderhiser of the Pacific Northwest National Laboratory for text processing.

Acronyms

CERCLA	*Comprehensive Environmental Response, Compensation, and Liability Act*
COC	contaminants of concern
cpm	counts per minute
CRAL	corrected remedial action level
DOE	U.S. Department of Energy
DOT	Department of Transportation
EPA	U.S. Environmental Protection Agency
NPL	National Priorities List
OU	Operable Unit
PQL	practical quantitation limit
RAG	remedial action goal
RASAP	Remedial Action Sampling and Analysis Plan
RAWP	Remedial Action Work Plan
RDR	Remedial Design Report
ROD	Record of Decision
SBF	sampling bias factor
USCS	Unified Soil Classification System
WHC	Westinghouse Hanford Company

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1.0 Introduction

The Hanford Site is a federal facility that occupies an area of 560 square miles. It is situated within the semiarid Pasco Basin in southeastern Washington State (Figure 1.1). During the years between 1943 and 1990, these facilities were used to produce fissionable nuclear materials. Following the cessation of these activities in 1990, the primary goal at the Hanford Site was shifted towards remediation of contaminated facilities, soil, and groundwater. The extensive contamination at the Hanford Site resulted in many large areas of the site, the 100, 200, 300, and 1100 Areas, being included on the National Priorities List (NPL) compiled under the *Comprehensive Environmental Response, Compensation, and Liability Act* (CERCLA) by the U.S. Environmental Protection Agency (EPA).

The 100 Area of the Hanford Site contains nine inactive nuclear reactors that were operated for the production of fissionable material. All these water-cooled reactors (B, C, D, DR, F, H, KE, KW, and N) situated along the southern bank of the Columbia River have been shut down and are now being evaluated for decommissioning. Waste streams that were generated during the operation of these reactors were disposed of in the vicinity, resulting in widespread contamination of both soil and water. The bulk of the contamination originated from the disposal of very large volumes of reactor cooling water containing both fission and activation products. As part of the remediation activities in the 100 Area, each of these nine reactor sites has been divided into distinct Operable Units (OU). Records of Decision (ROD) have been concluded for several OU in the 100 Area (EPA 1995, 1997), and the Remedial Design Report/Remedial Action Work Plan (RDR/RAWP) has been completed (DOE 1998a). Also, a Remedial Action Sampling and Analysis Plan (RASAP) for this area has been completed (DOE 1998b).

Currently, the RASAP for the 100 Area soils requires sampling and analyzing the soil material for contaminants of concern (COC) during the remediation process. However, the RASAP does not include a protocol for analyzing only a fine-grained fraction (sand, silt, and clay) and relating the measured COC activities/concentrations in this fraction to the COC activities/concentrations in the predominantly coarse-grained (gravel) whole soils in the 100 Area. Selectively sampling finer fractions of a coarse-grained soil would introduce a systematic bias that results in consistently overestimating the actual activities/ concentrations of COC in whole soil. Such sampling bias, if uncorrected, would result in over-remediation (remediating below the remediation action goal [RAG] levels) of these soils.

The objectives of this study were to 1) examine currently available particle size-COC activity/ concentration data for 100 and 300 Area soils, 2) assess the effects of sampling bias, 3) suggest sampling protocols, and 4) formulate a method to determine the COC activities/concentrations of the whole soil based on the measurements conducted on a finer size fraction of the whole soil.

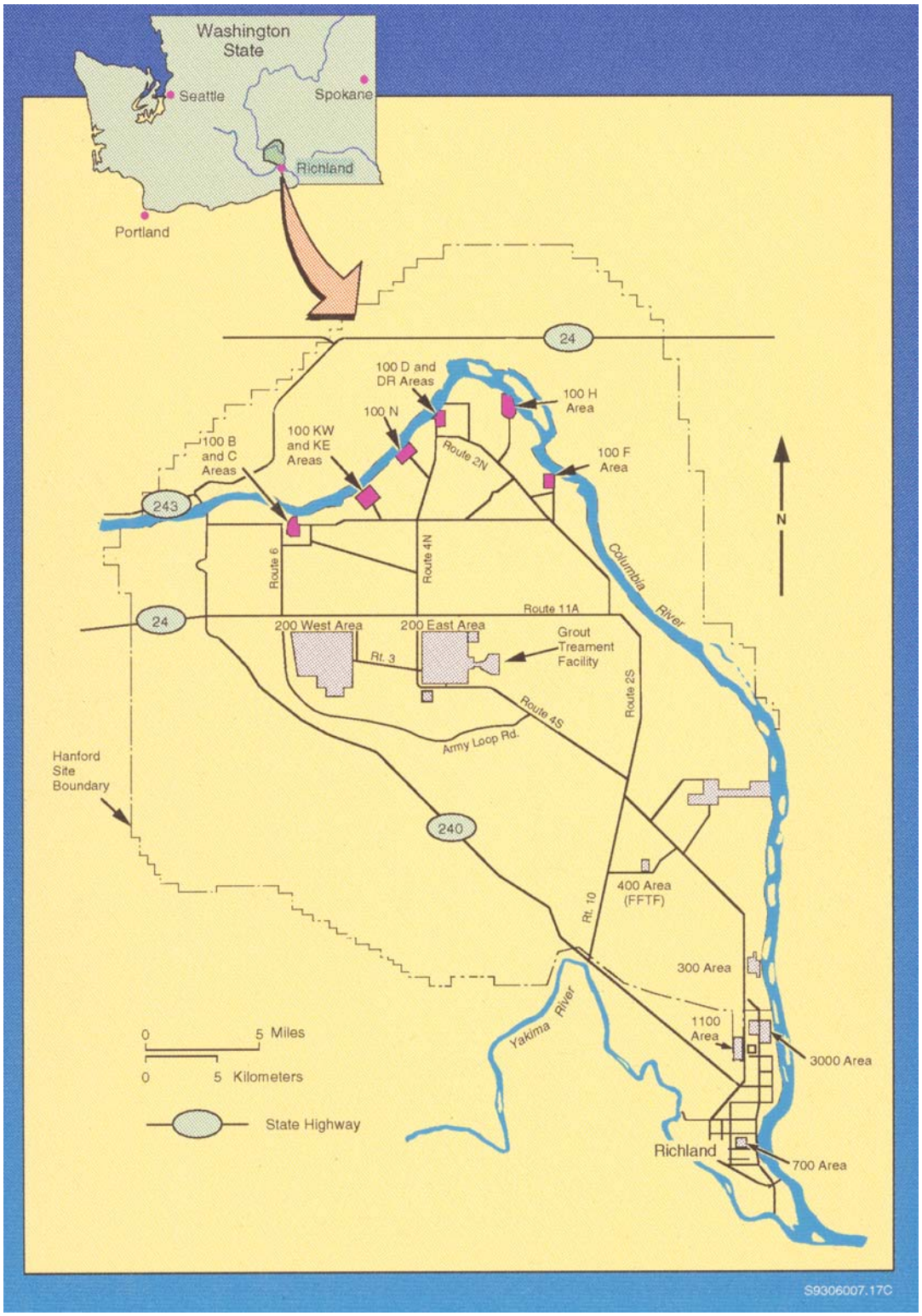

Figure 1.1. Location of the Hanford Site

2.0 Waste Sites and Soil Samples from the 100 and 300 Areas

2.1 Location of Waste Sites

The 100 and 300 Areas of the Hanford Site are located in a structural basin (Pasco Basin) on the Columbia Plateau (see Figure 1.1). A generalized geologic cross-section through the 100-B and C Areas (Figure 2.1) shows a sequence of unconsolidated surfacial sedimentary deposits (Hanford formation). The topmost layer of this formation consists of a thin layer (1 to 15 ft thick) of light brown, fine, slightly silty, eolian deposits.

The principal sediments of the Hanford formation underlying the surfacial layer range from 20 ft to several hundred feet in thickness and consist of poorly sorted, unconsolidated glaciofluvial material (Pasco gravels). The origin of these sedimentary deposits is believed to be the periodic cataclysmic floods that occurred during the late Pleiostocene epoch (Baker 1981; Mullineaux et al. 1977). The Pasco gravels are variable mixtures of particle sizes that range from boulders to silt. The bulk of Pasco gravels are very coarse-textured and are classified as silty sandy gravels typically consisting of about 50% gravel, 40% sand, and 10% silt and clay (DOE 1988). Mineralogically, these deposits consist of quartz, feldspars, and ferromagnesian minerals (DOE 1992a). All the trenches and cribs that were engineered for waste disposal in the 100 and 300 Areas of the Hanford Site were located in the Pasco gravels.

The Ringold Formation, ranging up to several hundred feet in thickness, underlies the Hanford formation. This formation, late Miocene to late Pleistocene in age, consists of stratified deposits of material ranging in size from gravel to clay. The uppermost unconfined aquifer system in the Hanford area, ranging up to 300 ft in depth, is located within the Hanford and Ringold Formations. The Saddle Mountain Basalt Formation underlies the stratified Ringold deposits.

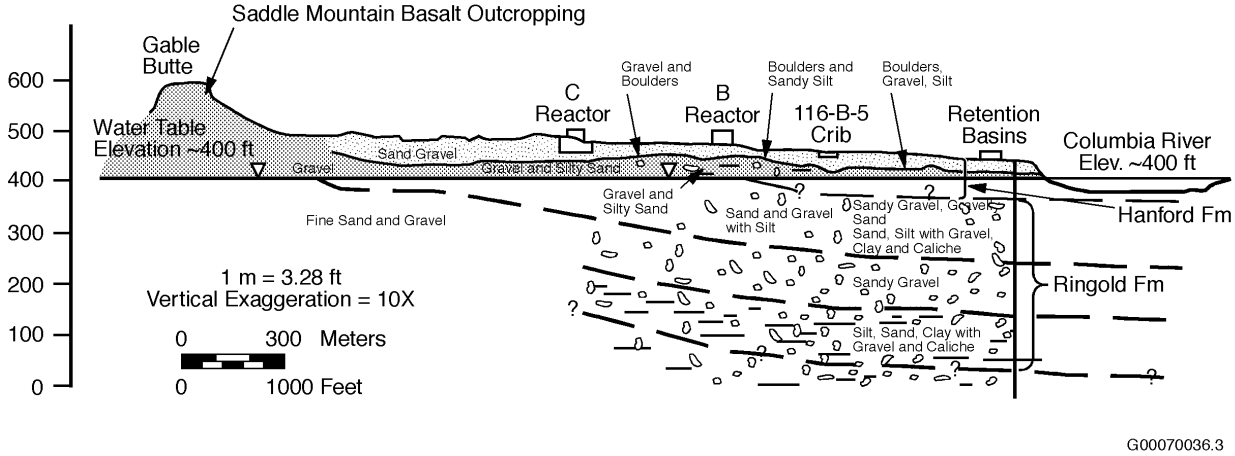

Figure 2.1. Generalized Geologic Cross-Section at 100-B and C Areas at Hanford

2.2 Waste Disposal Sites and Sources of Contamination

A literature survey indicated that only a limited set of particle size-contaminant concentration data is available for soils from the Hanford site. These data are based on a series of bench-scale tests conducted on single soil samples from one waste site each in 100-BC-1, 100-DR-1, and 100-FR-1 OUs (Mattigod et al. 1994 a,b), and three samples from the North Pond site of 300-FF-1 OU (Serne et al. 1992). Therefore, all additional data analyses conducted for this report were based on this limited data set.

The 116-C-1 liquid waste trench is located in the 100-BC-1 OU of the 100 Area (Figures 2.2 and 2.3). This unlined trench is 500 ft long, 50 ft wide, and 16 ft deep. This trench received reactor cooling effluent from the 116-C-5 retention basin from 1952 to 1958. During this period, this trench received a

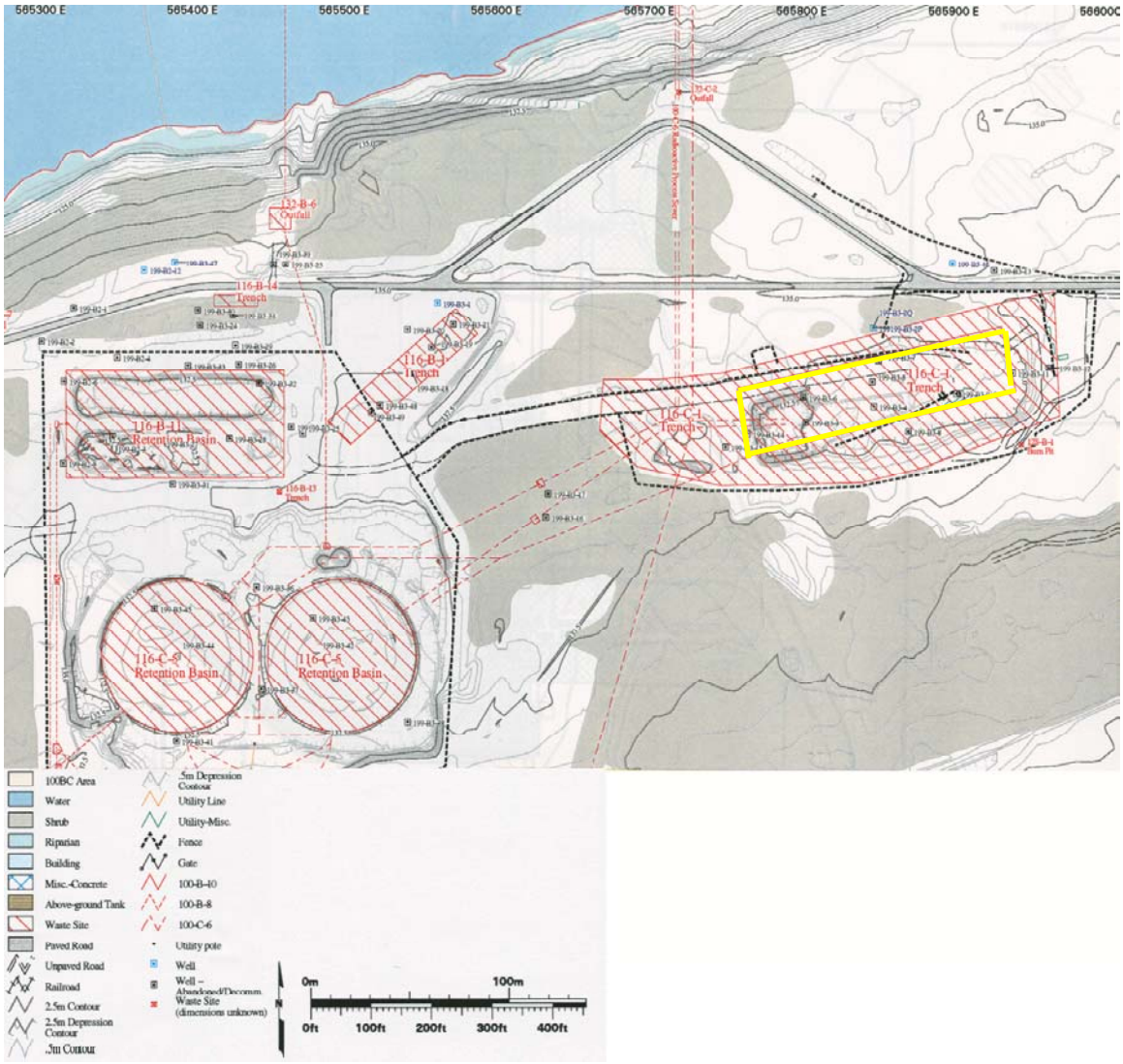

Figure 2.2. Location of 116-C-1 Waste Trench in the 100-BC-1 Operable Unit (from BHI-01119)

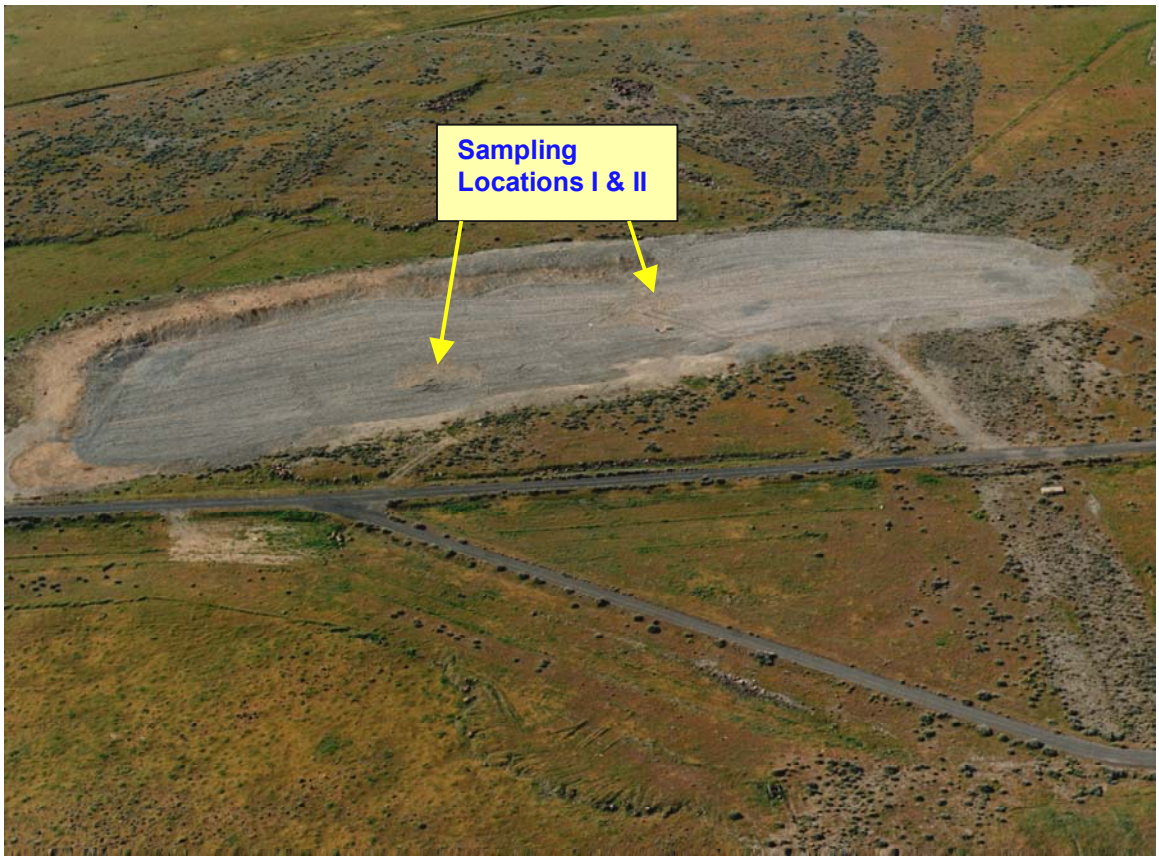

Figure 2.3. An Aerial View of 116-C-1 Waste Trench in 100-BC-1 Operable Unit

total estimated volume of 26 million gallons of effluent that was contaminated with activation and fission products caused by fuel-cladding failures in the 100-C Reactor. The 116-C-1 trench also received chromium-bearing wastes in the form of sodium dichromate (DOE 1992b). This trench was partially filled with rocks to prevent the spread of contamination by wind action.

Based on the soil sampling conducted in 1976, the total volume of contaminated soil in this trench was estimated by Dorian and Richards (1978) to be about 100,000 yd^3(600 by 150 by 30 ft). The average activities of radionuclides in the 116-C-1 trench are listed in Table 2.1. The activity data for soil in this trench indicate that ^{60}Co, ^{137}Cs, ^{152}Eu, ^{154}Eu, and ^{90}Sr are the potential contaminants of concern. In 1995, about 11,690 m^3 of material from this trench was removed as part of a 100-B/C Expedited Response Action Demonstration Project (DOE 1998a).

The 116-D-1B liquid waste trench is located in the 100-DR-1 Operable Unit of the 100 Areas (Figures 2.4 and 2.5). This trench is 100 ft long, 10 ft wide, and 15 ft deep. It was used mainly to dispose of radionuclide-contaminated sludge and water from the 118-D-6 fuel storage basin. This material originated as the result of storing ruptured fuel elements in the 118-D-6 basin. The 116-D-1B trench was used from 1953 through 1967 and probably received a major fraction of sludge (estimated to be 110,000 lb) that originated in the fuel storage basin. Other contaminants disposed of in this trench included about

Table 2.1. Average Activities of Radionuclides in Selected Waste Disposal Trenches and a Crib in the 100-BC-1, 100-DR-1, and 100-FR-1 Operable Units[a]

Radionuclide	116-C-1 Trench (pCi/g)	116-D-1B Trench (pCi/g)	116-F-4 Pluto Crib (pCi/g)	Hanford Background (pCi/g)	Remedial Action Goals[c] Medial Action Goals (pCi/g)
^{60}Co	7.6[b]	0.59[b]	0.02[a]	0.008	1.4
^{137}Cs	21.4[b]	24.1[b]	877.6[a]	1.1	6.2
^{152}Eu	31.5[b]	7.5[b]	2.2[a]	NA	3.3
^{154}Eu	21.7[b]	1.9[b]	12.9[a]	0.033	3.0
^{90}Sr	5.7[b]	7.3[b]	573.9[a]	0.18	4.5
^{235}U	0.2[c]	0.18[c]	1.3[b]	0.11	1.0
^{238}U	(d)	(d)	(d)	1.1	1.1
$^{239/240}$Pu	0.74	0.48	35	0.025	33.9
Contaminated soil volume (cubic yards)	100,000	7,800	670	--	-

(a) Data Sources: Average activities in trenches and crib (Dorian and Richards 1978), decay corrected. Particle size of soils used for radionuclide measurements not known.
(b) Hanford Background: DOE 1996.
(c) RAG values for 100 Area soils: DOE 1998a.
(d) Decay corrected to 2000. Samples from 116-D-1B obtained in 1993 near the trench inlet (south side) indicated average activities of 5.6 pCi/g, for ^{60}Co, 103.5 pCi/g for ^{137}Cs, and 83.7 pCi/g for ^{152}Eu, respectively. The results of the pilot-scale soil washing tests (116-D-1B) showed average activities of 0.34 pCi/g, for ^{60}Co, 11.9 pCi/g for ^{137}Cs, and 4.07 pCi/g for ^{152}Eu, respectively (DOE 1995).

1,540 lb of sodium dichromate, 4,400 lb of either sodium formate or oxalate, and 4,400 lb of sodium sulfamate. After waste discharges were stopped in 1967, the trench was blanketed with a layer of clean soil. As a result of sampling and analyses of soils from this trench, the total volume of contaminated soil was estimated by Dorian and Richards (1978) to be about 7,800 yd^3(150 by 40 by 35 ft). The average activities of radionuclides in this trench (Dorian and Richards 1978) are listed in Table 2.1. These data indicate that the radionuclide COC in this trench are ^{137}Cs, ^{152}Eu, and ^{90}Sr. In 1995, a pilot-scale soil washing test was conducted, and 101.1 tons of material from this trench was processed (DOE 1995). The data collected during this field test indicated that the radionuclide COCs in this trench were mainly ^{137}Cs and ^{152}Eu.

The 116-F-4 Pluto Crib is located in the 100-FR-1 Operable Unit (Figure 2.6). This Pluto Crib consisted of a cobble-filled 55-gal steel container with sealed top and open bottom resting on one end of an 6- by 6- by 6-ft zone of cobble-size aggregate (DOE 1994). This crib was one of the typical liquid disposal structures constructed below the ground surface with open bottoms. A distribution pipe discharged wastewater into the rockfill to generate uniform flow over the crib bottom. The top of the crib was backfilled with soil to provide protection from radiation. This structure received water from process tubes in the 100-F Reactor that had failed fuel claddings. This crib was used only for a short time (1952 to 1954) and was contaminated with activation and fission products. Based on the soil sampling conducted in 1976, the total volume of contaminated soil in this trench was estimated to be about 670 yd^3

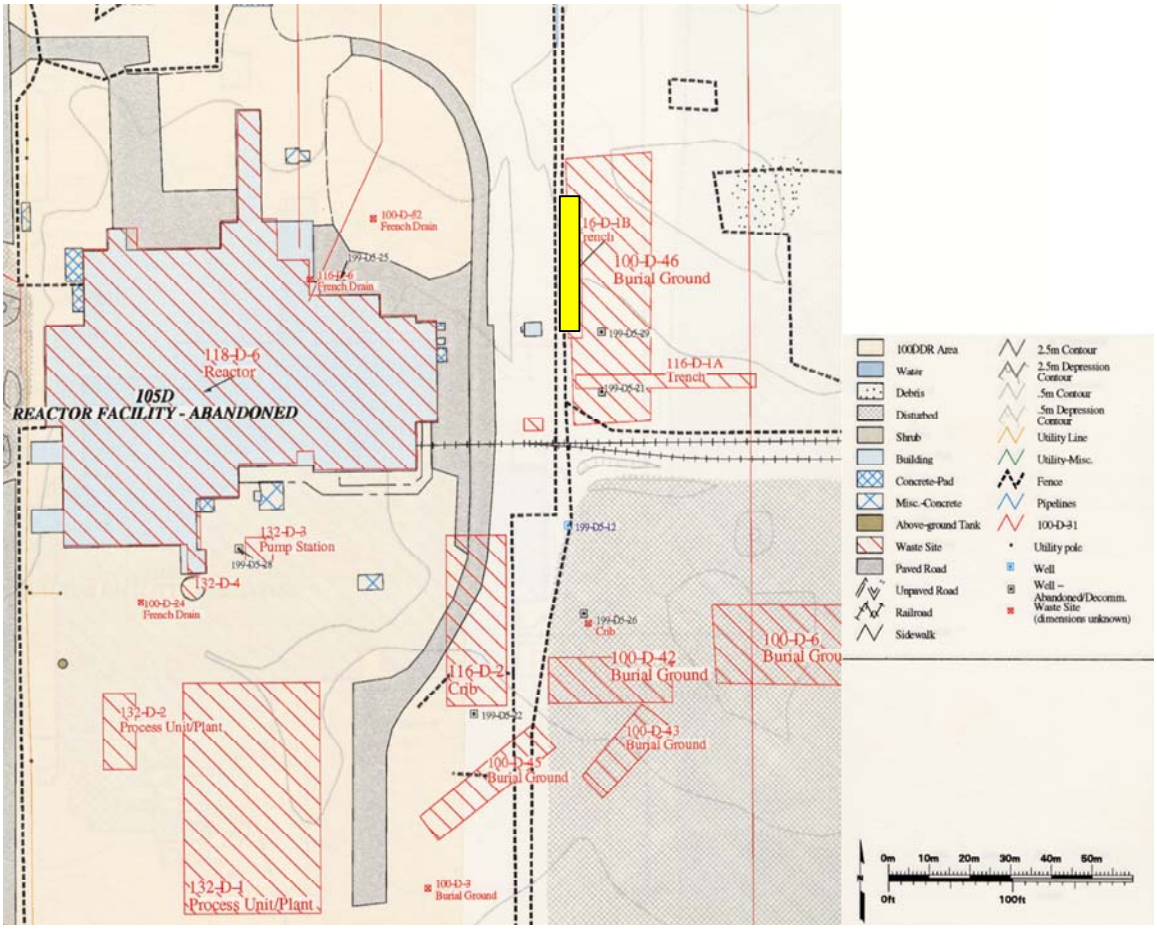

Figure 2.4. Location of the 116-D-1B Liquid and Sludge Waste Trench in the 100-D/DR Area (from BHI-01119)

(30 by 30 by 20 ft) (Dorian and Richards 1978). The average activities of radionuclides for the 116-F-4 Pluto Crib are listed in Table 3.1. In 1994, this crib was excavated to a depth of 5.5 m (~4,500 yd^3 of material) and the contaminated material was disposed of at the Environmental Restoration Disposal Facility (DOE 1994, 1998a).

The North Process Pond (316-2) is in the northern part of 300-FF-1 Operable Unit in the Hanford 300 Area (Figures 2.7 and 2.8). This pond of about 4 hectares (~10 acres) was constructed in 1948 to receive low-level radioactive liquid wastes and sludges from the fuels fabrication process and liquid wastes from other types of operations in the 300 Area. Parts of the North Pond were also used to dispose of fly ash derived from a coal-burning power plant. The pond was dredged periodically to increase the infiltration rate. It was retired in 1974. A detailed description of waste discharge history, pond operations, and organic, inorganic, and radioactive contaminants found in this pond can be obtained from several previously published reports (Dennison et al. 1989; Young et al. 1990; Young and Fruchter 1991). An analysis of the pond sediments conducted in 1992 indicated that activities of ^{235}U and ^{238}U, and concentrations of Cr, Ni, Cu, Zn, Zr, Ag, and Sr were elevated above the background concentrations found in the uncontaminated sediments from the 300 Area (Serne et al. 1992).

Figure 2.5. An Oblique Aerial View of 100-D-1B Waste Trench

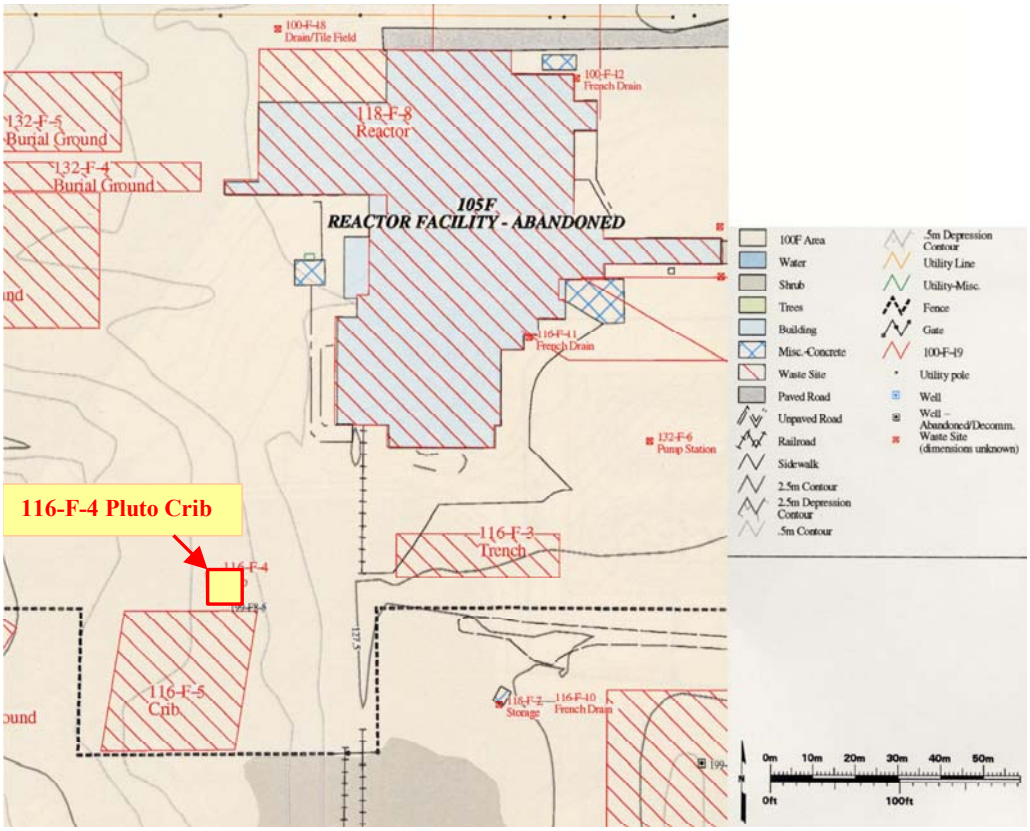

Figure 2.6. Location of the 116-F-4 Pluto Crib in the 100-FR-1 Operable Unit (from BHI-01119)

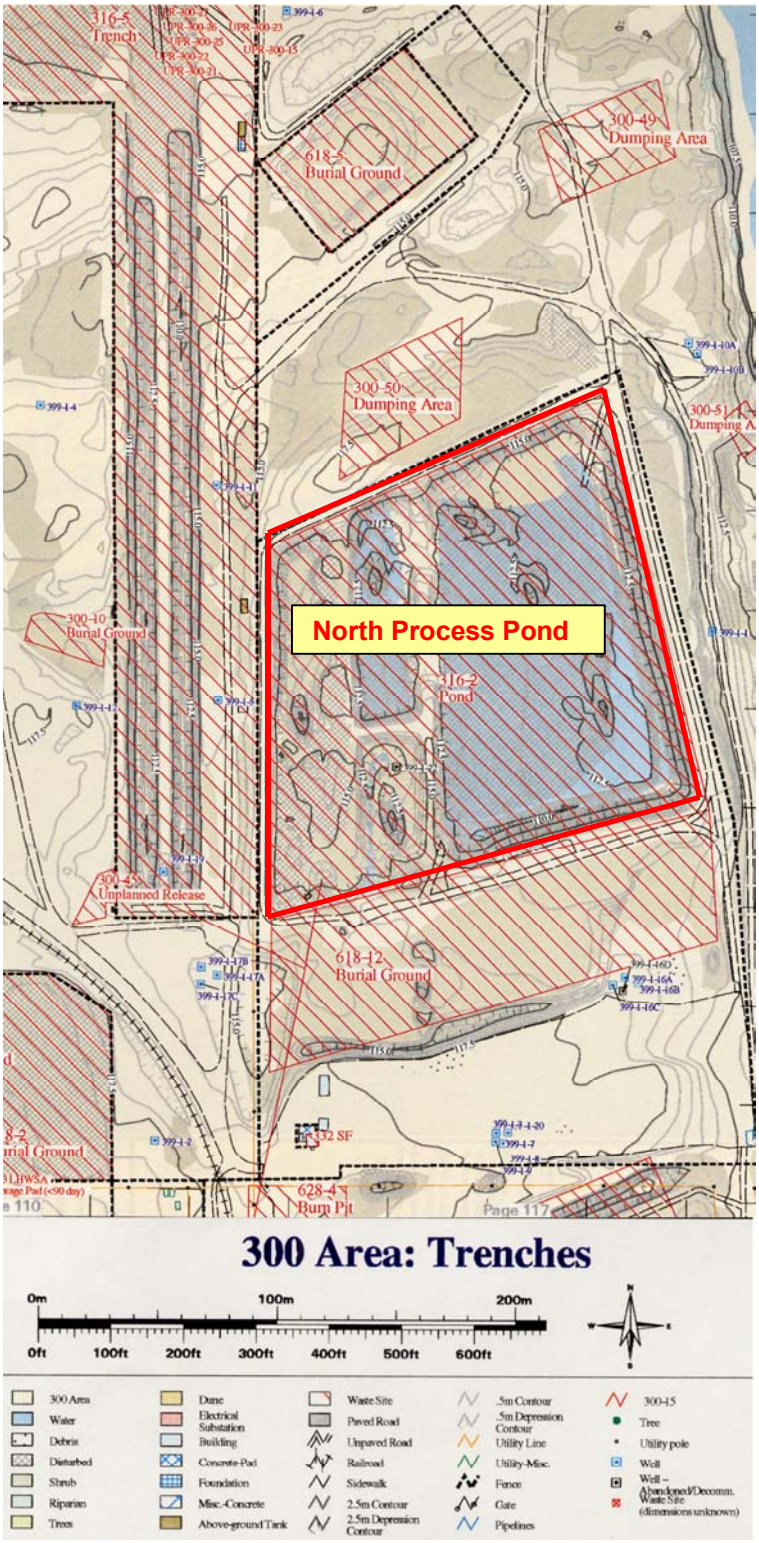

Figure 2.7. Location of North Process Pond in the 300-FF-1 Operable Unit (from BHI-01119)

Figure 2.8. An Oblique View of North and South Process Ponds in the 300-FF-1 Operable Unit

2.3 Soil Sample Collection

All the soil samples were collected by WHC personnel and transported to the laboratory in Department of Transportation (DOT)-specified containers. The soil sampling was conducted according to the protocol described in the excavation and sampling plan (WHC 1993). According to this sampling plan, bucket loads of soils that exceeded 100 counts per minute (cpm) above the background were deemed as acceptable samples.

The first sample was collected at a depth of 20 ft from a pit excavated in the middle of the 116-C-1 trench (see Figure 2.3). This sample was designated as "Batch I." A second set of soil samples (Batch II) was obtained from the vicinity of the inlet pipe (Guzek and Field 1993).

Field monitoring indicated radioactivity increasing from approximately 1,000 cpm at a 10-ft depth to about 20,000 cpm at a depth of 20 ft. At a depth of 22 to 24 ft, the measured activities were lower (1,500 to 2,000 cpm), indicating that the maximum soil contamination in this trench occurs near the inlet at a depth of 20 ft. The soil samples were collected from 10 ft, 15 to 18 ft, and 18 to 20 ft depth intervals. A total volume of 55 gal of each soil was collected and transported to the laboratory.

The soil samples (Batch III) from the 116-D-1B (Figure 2.5) trench were collected at depth intervals of 5 to 10 ft, and 15 to 20 ft. Activities at these sampling depths were about 600 cpm. The activity was

observed to decline to a level of 150 to 200 cpm at a depth of approximately 22 to 24 ft. A total volume of 55 gal of soil was collected and transported to the laboratory in 11 5-gal containers.

About 4,500 yd^3 of soil including the material from the 116-F-4 Pluto Crib was excavated as part of an Excavation Treatability Test (DOE 1994). About 500 yd^3 of this material was classified as contaminated soil and stored in a modular storage unit. During excavation, the soils were treated with dust suppressants such as XDCA (a beet polysaccharide) and lignosulfate (a paper manufacturing byproduct). Samples of contaminated soil were collected from the modular storage unit by Westinghouse Hanford Company (WHC) personnel and transported to the laboratory in DOT-specified containers. A total volume of 40 gal of soil was collected and transported to the laboratory in eight 5-gal containers. Three bulk samples of sediments from North Process Pond were collected by WHC and sent to the laboratory.

2.4 Soil Characterization Data Sources

The physical, chemical, and mineralogical properties of the soil samples obtained from these waste sites were characterized, and the results were published in a series of reports (Mattigod et al. 1994 a, b; Serne et al. 1992). The types of characterization data collected for these soil samples are listed in Table 2.2.

Table 2.2. Characterization Matrix for Soil Samples from Selected Waste Sites from 100 and 300 Areas, Hanford

Characterization	116-C-1 I & II	116-D-1B	116-F-4	NPP 316-2 Samples
Physical Characterization				
Moisture content	●	●	●	●
Particle size distribution	●	●	●	
Specific gravity	●	●	●	
Chemical Characterization				
Total organic carbon	●	●	●	
Soil pH	●	●	●	
Cation exchange capacity	●	●	●	
Total elemental analysis	●	●	●	
Selected radionuclide analysis as a function of particle size	● I	●	●	●
Metal analysis as a function of particle size				●
TCLP	● II	●	●	
Sequential extraction	● II	●	●	
Attrition scrubbing of selected particle fractions		●	●	
Autogenous grinding of gravel fractions	● II		●	
Mineralogical Characterization				
Optical microscopy		●	●	
X-ray diffraction analysis		●	●	
SEM-EDX		●	●	
Auto radiography		●	●	

3.0 Contaminant Activity-Particle Size Data Analysis

3.1 Particle Size and Contaminant Activity Data Sources

Particle size distribution and particle size dependent contaminant activity/concentration data used in this analysis were derived from results of previous studies conducted on soils obtained from 100-C-1 and 100-D-1B waste disposal trenches and 116-F-4 Pluto Crib (Mattigod et al. 1994a, b). Additionally, information obtained from sampling and characterization of sediments from the North Process Pond disposal site (316-2) in the 300-FF-1 Operable Unit (Serne et al. 1992) was also used in our analysis. It is important to note that the following data analyses are based on single soil samples each from 116-D-1B, and 116-F-4 sites, one to two soil samples from 116-C-1, and three samples from 300 North Pond site. Therefore, these analyses of particle size-COC activity relationships are relevant only for the soil samples analyzed and do not represent the spatial variability expected at each of these waste sites.

3.2 Particle Size Distribution

The particle size distribution data for the six soil samples (Figures 3.1, 3.2; Table 3.1) showed that all the soils are coarse-grained with gravel fractions ranging from ~42 to ~97% by mass. According to the Unified Soil Classification System (USCS) system, the soils from the 116-F-4 Pluto crib and the samples from the 300 Area North Pond were classified as well-graded gravels with sand. In contrast, the 116-C-1 (II) Trench soil was classified as a poorly graded gravel, and the soil from the 116-D-1B (III) Trench was categorized as a well-graded sand with silt and gravel. Except for the 116-D-1B Trench soil, all other soil samples were found to be typically coarser grained (~66-97% gravel and ~2–31% sand) than the average Pasco gravel (~50% gravel and ~40% sand) that constitutes the Hanford formation. Such soil textural differences may be attributable to the fact that waste sites were engineered with coarser soil material to increase rates of infiltration of liquid effluents.

Table 3.1. Classification of Selected Soils from the Waste Sites in 100 and 300 Areas at Hanford

Soil Sample	Gravel (wt%)	Sand (wt%)	Silt and Clay (wt%)	Group Name[(a)]	Group Symbol[(a)]
116-C-1 (II) trench	97.2	1.8	1.0	Poorly graded gravel	GP
116-D-1B(III) trench	41.6	51.3	7.1	Well-graded sand with silt and gravel	SW-SM
116-F-4 pluto crib	80.7	17.2	2.1	Well-graded gravel with sand	GW
BO1F87-NPT-1 North Pond	72.4	27.3	0.3	Well-graded gravel with sand	GW
BO1F93-NPT-2 North Pond	70.4	25.8	3.9	Well-graded gravel with sand	GW
BO1F93-NPT-3 North Pond	66.3	30.6	3.1	Well-graded gravel with sand	GW
(a) Classification according to the Unified Soil Classification System (ASTM D2487-98).					

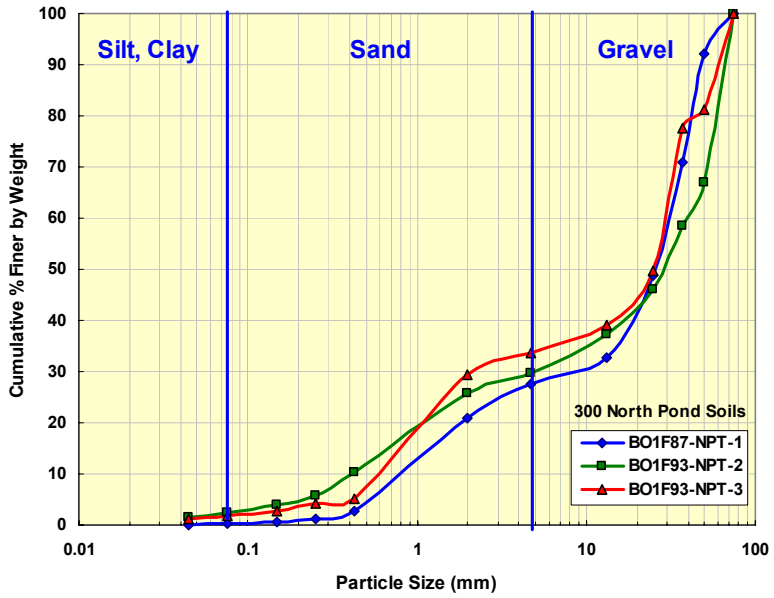

Figure 3.1. Particle Size Distribution for 116-C-1 and 116-D-1B Trenches and 116-F-4 Pluto Crib Soils

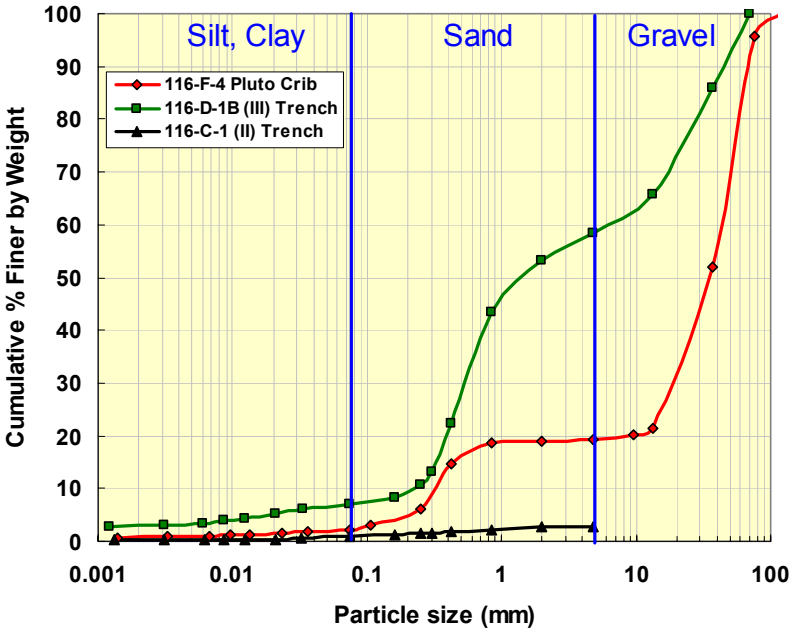

Figure 3.2. Particle Size Distribution for Sediment Samples from North Process Pond in the 300 Area

3.3 Particle Size-Contaminant Activity/Concentration Relationships

The contaminant activity/concentration data for these samples were collected by wet sieving (Samples 116-C-1, 116-D-1B, and 116-F-4) or dry sieving (North Process Pond samples) representative material and counting or measuring the activities/concentrations in each size fraction. These data were used to develop a relationship between the activity/concentration for soil material finer than each selected particle size as follows:

$$A_f = \Sigma(A_i \times m_i)/m_f \quad (3.1)$$

where A_f = activity/concentration per unit mass in soil finer than selected particle size
A_i = activity per unit mass in each particle size fraction
m_i = mass of each particle fraction
m_f = total mass of material finer than the selected particle size.

The resulting particle size-contaminant activity/concentration relationships are depicted as a series of graphs (Figures 3.3 through 3.7). The data showed that, in all cases, the activity/concentration per unit mass in any fractional soil material finer than the whole soil were higher than the activity/concentration per unit mass of whole soil. Typically, the distribution curves for all contaminants in a soil are similar in shape, reflecting similar partitioning of each contaminant between particle size fractions in the soil. These data showed that the degree to which the activity/concentration in subfractional material exceeded the whole soil activity/concentrations depended on the particle size distribution and the distribution of COC in each size fraction.

As an example, the consequence of such particle size-dependent contaminant activity/concentration relationship can be assessed by examining the ^{152}Eu activity in 116-C-1 Trench soil (Figure 3.3). The ^{152}Eu activities in this sample of whole soil (2.2 pCi/g) is well below the RAG of 3.3 pCi/g. However, if soil material finer than 2 mm is selectively sampled for ^{152}Eu and the measured activity of 16.3 pCi/g is presumed to be representative of the whole soil, one could erroneously conclude that the volume of soil represented by this sample needs to be remediated. Similarly if material finer than medium sand (<0.25 mm) from 116-F-4 Pluto Crib is preferentially sampled and if ^{152}Eu activity in this fraction (6.1 pCi/g) is assumed to represent the activity in whole soil sample, one can again mistakenly conclude that the ^{152}Eu activity in this soil sample exceeds the RAG value of 3.3 pCi/g (Figure 3.5).

These data clearly demonstrate that biased sampling of a finer particle size fraction of a soil would lead to the measured contaminant activity/concentration to be biased consistently higher than the actual contaminant activity in a soil.

3.4 Sampling Bias Factor

Based on the data from activity/concentration and particle size relationship, it was possible to quantify the degree of bias if only finer particles of soil are sampled for activity/concentration

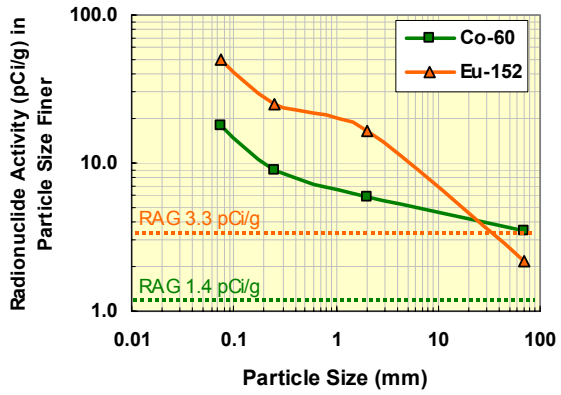

Figure 3.3. Particle Size-Radionuclide Activity Relationship for Soil from 116-C-1 Trench

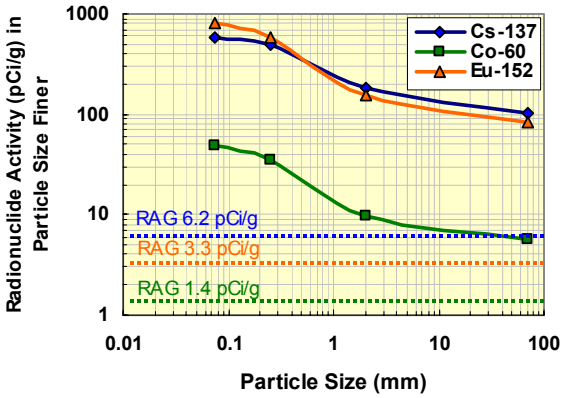

Figure 3.4. Particle Size-Radionuclide Activity Relationship for Soil from 116-D-1B Trench

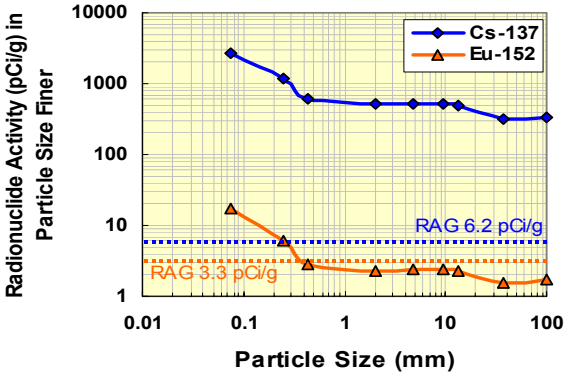

Figure 3.5. Particle Size-Radionuclide Activity Relationship for Soil from 116-F-4 Pluto Crib

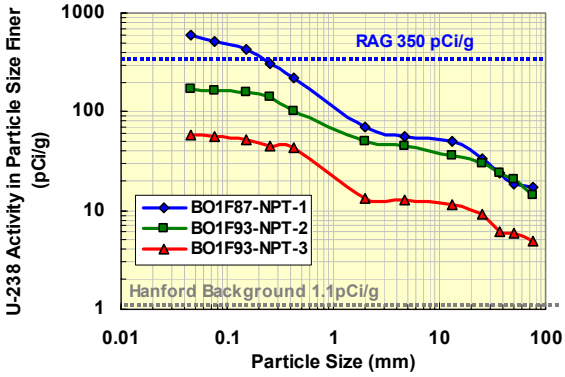

Figure 3.6. Particle Size-Radionuclide Activity Relationship for Soil Samples from North Process Pond (316-2) Located in 300-FF-1 Operable Unit

measurements and these values attributed to the whole soil (includes all particle sizes). For each contaminant in a soil, sampling bias factors (SBF) were calculated as a function of sampling material of less than a specified particle size.

The SBF values were calculated as

$$SBFij = [\Sigma(Ai \times mi)/mf)/Ab]-1 \qquad (3.2)$$

where SBF = sampling bias factor for ith particle size finer for jth contaminant
Ab = activity/concentration per unit mass of whole soil
Ai = activity per unit mass in each particle size fraction
Mi = mass of each particle fraction
mf = total mass of material finer than the selected particle size.

For instance, if a representative sample of a whole soil (all particle sizes included) is counted for the activity of a selected radionuclide, the SBF for the radionuclide would be zero, indicating that there is no sampling bias. If, however, only particle sizes finer than the maximum particle size in a soil are sampled, a positive sample bias would result, indicating the degree to which the measured activities/concentrations exceed the activity/ concentration in a whole soil. The SBFs for each contaminant in each soil sample were plotted as a function of particle size finer (Figures 3.8 through 3.12). Also, values of SBF were tabulated (Table 3.2) for each soil for cases when sampling would be consistently conducted on particles finer than sand (<0.075 mm). These data indicated that, for a selected particle size finer sample,

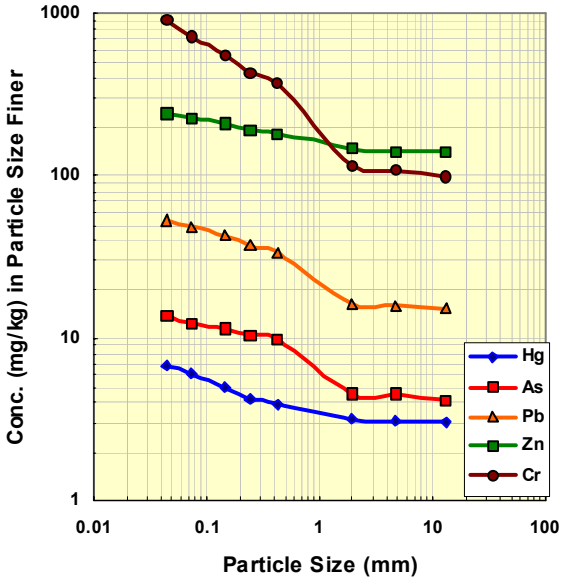

Figure 3.7. Particle Size-Concentration Relationships for Selected Inorganic Constituents in the B01F93-NPT-3 Soil Sample from North Process Pond (316-2)

1) SBF for any COC in a soil increases with decreasing particle size sampled.

2) SBFs for different COC in a soil can vary significantly (up to an order of magnitude) depending on their chemical properties and affinity for specific mineral surfaces.

Table 3.2. Sampling Bias Factors for Analytes for Selected Sampling of Silt and Clay (<0.075 mm) Soil Materials

Analyte	100 Area Soil Samples			300 Area North Pond Samples		
	116-C-1	116-D-1B	116-F-4	NPT-1	NPT-2	NPT-3
Co^{60}	4.2	7.7	--	--	--	--
Cs^{137}	--	4.7	7.0	--	--	--
Eu^{152}	22.0	8.8	8.7	--	--	--
U^{238}	--	--	--	29.6	10.5	10.6
As	--	--	--	1.8	1.8	2.0
Cr	--	--	--	9.0	3.1	6.3
Hg	--	--	--	1.8	1.8	1.0
Pb	--	--	--	5.3	2.3	2.2
Zn	--	--	--	0.8	0.8	0.6

3) SBF for a single COC in soil samples obtained in different locations from the same waste site can be significantly different (two to three times higher than the lowest value) reflecting spatial variability engendered by the source, type and distribution of contamination.

4) SBF for COC in soils from different waste sites were different and these values were affected by the soil texture, the degree, the source, and the distribution of contamination.

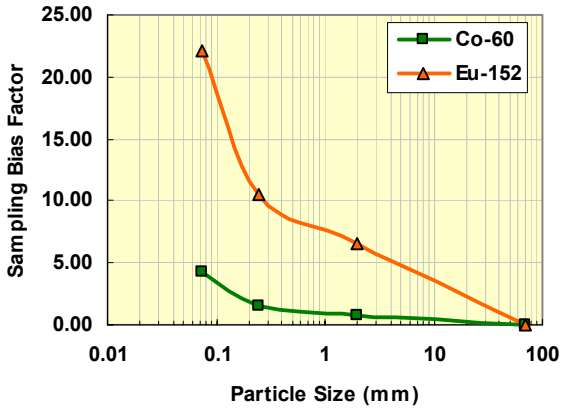

Figure 3.8. Sampling Bias Factors for ^{60}Co and ^{152}Eu in Soil from 116-C-1 Trench

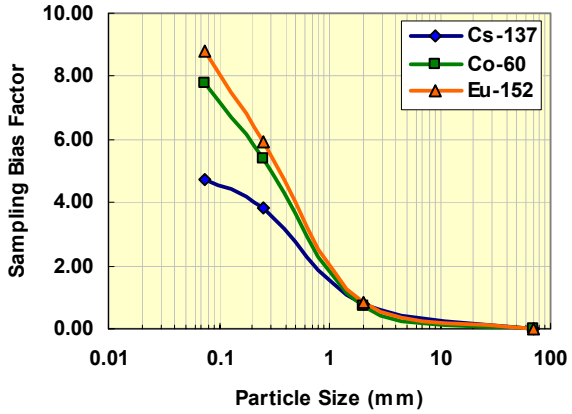

Figure 3.9. Sampling Bias Factors for ^{60}Co, ^{137}Cs, and ^{152}Eu in Soil from 116-D-1B Trench

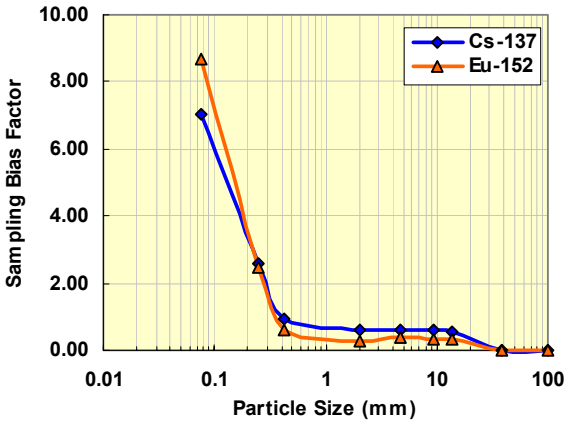

Figure 3.10. Sampling Bias Factors for ^{137}Cs and ^{152}Eu in Soil from 116-F-4 Pluto Crib

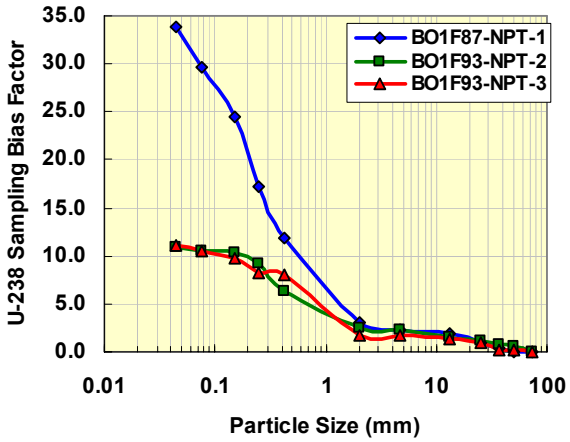

Figure 3.11. Sampling Bias Factor for ^{238}U in North Process Pond (316-2) Soil

3.5 Sampling Bias and Over-Remediation of Whole Soils

As discussed in previous section, bias sampling of a selected particle fraction results in overestimating COC activities/concentrations in whole soil. One of the major consequences of bias sampling is over-remediation of contaminated soils. Over-remediation is defined as the degree to which the activity/concentration of a contaminant in the remediated whole soil is lower than the specified RAG. The degree to which the over-remediation occurs can be calculated from using RAG and SBF values as follows:

$$WS_j = RAG_j / (SBF_{ij} + 1) \qquad 3.4$$

where WSj = activity/concentration of jth COC in whole soil
RAGj = remedial action goal for jth COC
SBFij = sampling bias factor for jth COC in ith particle size finer fraction.

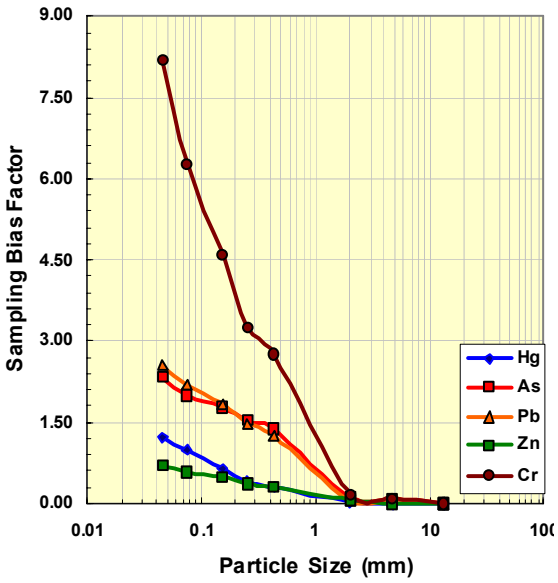

Figure 3.12. Sampling Bias Factors for Selected Trace Metal Contaminants in B01F93-NPT-3 Soil Sample from North Process Pond (316-2)

Calculated whole soil values for each contaminant radionuclide were graphed as a function of sampled particle size finer (Figures 3.13 through 3.16) and listed (Table 3.3) for particle size finer than sand (<0.075 mm). These data showed that, for samples of any particle size finer than the whole soil, the radionuclide activity in remediated whole soil would be consistently less than the specified RAG value.

The degree of over-remediation increases with decreasing particle size sampled (Figure 3.13 through 3.16). As an example, if soil particles finer than sand are sampled preferentially, the activity of radionuclides in remediated whole soil would be up to an order of magnitude lower than the RAG (Table 3.3). In some cases, the radionuclide activities in remediated whole soil would be up to an order of magnitude lower than the Hanford background activity (^{238}U activity in 300 Area North Pond soil) and, in some cases, (^{60}Co in 116-C-1 trench soil) lower than the rapid turnaround practical quantitation limit (PQL).

Practical considerations dictate that whole soil from 100 and 300 Hanford Areas (containing gravel and even cobble-size material) cannot be sampled representatively and quantified rapidly under field conditions. Therefore, field sampling for rapid turnaround quantitation needs to be restricted specifically to soil particles finer than gravel or sand. To meet this requirement, it is essential to establish a relationship between measured activity/concentration in the sampled soil size fraction and the RAG for the whole

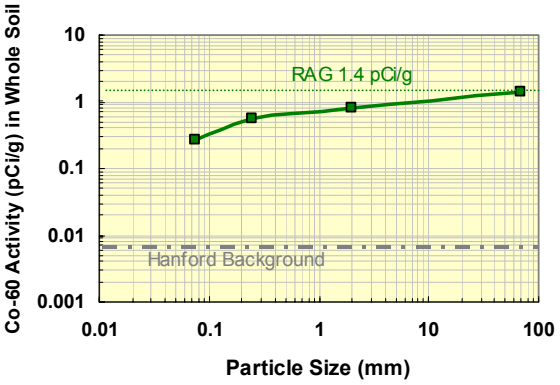

Figure 3.13. Activity of ^{60}Co in Remediated Whole Soil (116-C-1 Waste Trench) Without SBF Corrections

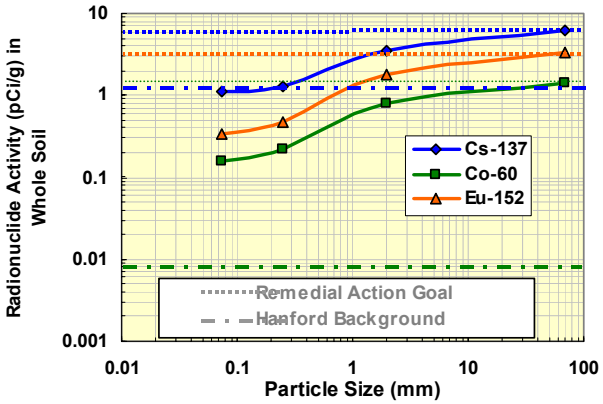

Figure 3.14. Activities of ^{60}Co, ^{137}Cs, and ^{152}Eu in Remediated Whole Soil (116-D-1B Waste Trench) Without SBF Corrections

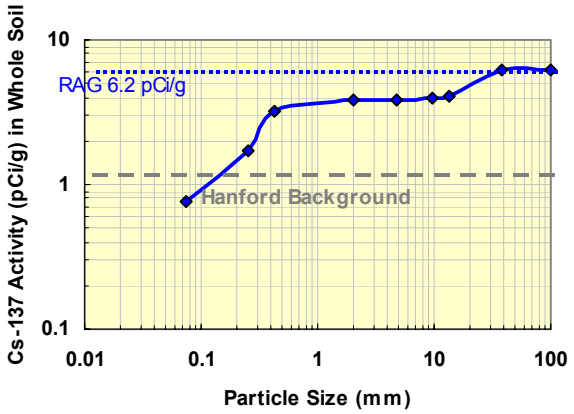

Figure 3.15. Activity of ^{137}Cs in Remediated Whole Soil (116-F-4 Pluto crib) Without SBF Corrections

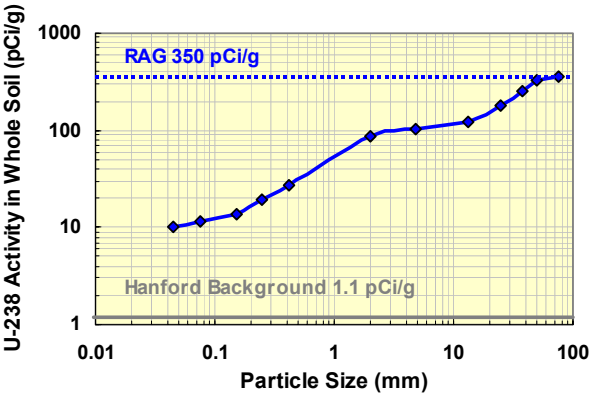

Figure 3.16. Activity of ^{238}U in Remediated Whole Soil (Sample BO1F87-NPT-1 from North Process Pond, 316-2) Without SBF Corrections

soil. These sampling bias corrected remedial action levels (CRAL) for the specified particle size finer (that would meet the whole-soil RAG) can be calculated using the appropriate RAG and SBF values as follows:

$$CRAL_{ij} = RAG_j \times (SBF_{ij} + 1) \qquad (3.5)$$

where CRALij = Corrected remediation action level for jth contaminant in ith particle size finer
RAGj = Remedial action goal for jth contaminant
SBFij = Sampling bias factor for jth contaminant in ith particle size finer.

Table 3.3. Radionuclide Activities in Whole Soil Based on Biased Sampling of Soil Material Finer than Sand (<0.075 mm)

Radio-nuclide	100 Area Soil Samples (pCi/g)			300 Area NP Sample (pCi/g)	RAG (pCi/g)	Hanford Background (pCi/g)	PQL (pCi/g)
	116-C-1	116-D-1B	116-F-4	NPT-1			
Co^{60}	0.3	0.2	--	--	1.4	0.008	0.7
Cs^{137}	--	1.1	0.8	--	6.2	1.1	0.7
Eu^{152}	--	0.3	--	--	3.3	NA[(b)]	0.1[(c)]
U^{238}	--	--	--	11.4	350[(a)]	1.1	1.0[(c)]

(a) RAG values are 350 pCi/g for 300 Area soils (DOE 1997) and 1.1 pCi/g for 100 Area soils (DOE 1998a).
(b) Not Available, not measured during background study.
(c) Practical Quantitation Limit (PQL) not attainable for rapid turnaround measurements, attainable only through longer turnaround "protocol" method.

The CRAL for particle sizes sampled finer than gravel (<4.75 mm) and finer than sand (<0.075 mm) are listed in Table 3.4. The results indicated that, depending on radionuclide, soil type, and the degree of contamination, the CRAL values for biased sampling would be 0 to ~3 times the RAG values for finer-than-gravel material and ~5 to ~30 times the RAG values for finer-than-sand material.

These assessments, based on detailed particle-size and activity distribution data for selected 100 and 300 Area waste site soils, suggested that

1. SBFs should be determined for each waste site prior to excavation.

2. A protocol needs to be established to consistently obtain representative soil samples of selected particle size (finer than gravel or sand).

3. From the predetermined SBF for each contaminant in soil CRAL, values need be established for each waste site.

4. CRAL values need be established for each COC to significantly attenuate the risk of over-remediation of waste sites.

Table 3.4. Corrected Remedial Action Levels for Contaminant Radionuclides for Biased Sampling of Soil Particles Finer than Gravel (<4.75 mm) and Sand (<0.075 mm)

Radio-nuclide	CRAL Values for 100 Area Soil Samples (pCi/g)						CRAL Values for 300 Area North Pond Sample (pCi/g)		RAG (pCi/g)	PQL (pCi/g)
	116-C-1		116-D-1B		116-F-4		NPT-1			
	<4.75	<0.075	<4.75	<0.075	<4.75	<0.075	<4.75	<0.075		
Co^{60}	1.5	7.3	1.5	12.3	--	--	--	--	1.4	0.7
Cs^{137}	--	--	6.5	35.3	10.0	43.5	--	--	6.2	0.7
Eu^{152}	--	--	3.3	32.3	--	--	--	--	3.3	0.1[(b)]
U^{238}	--	--	--	--	--	--	1,155	10,710	350[(a)]	1.0[(b)]

(a) RAG values are 350 pCi/g for 300 Area soils (DOE 1997) and 1.1 pCi/g for 100 Area soils (DOE 1998a).
(b) PQL not attainable for rapid turnaround measurements, attainable only through longer-turnaround "protocol" method.

4.0 Effects of Sampling Bias on Remediation Limits for 116-C-1 Waste Trench—A Preliminary Assessment

Analysis of particle size-contaminant activity/concentration data in Section 3 showed that biased sampling of a finer size fraction (assuming that the measured activity/concentrations of a contaminant in this fraction represents the actual contaminant activity/concentration of a whole soil) results in potential over-remediation of the soil. The analysis also showed that if detailed particle size distribution and activity/concentration data for a contaminated soil are available, it is possible to correct the sampling bias and thus reduce the probability of over-remediating the soil.

We conducted a preliminary assessment of effects of sampling bias using a set of historical borehole data for 116-C-1 waste trench. The location, and the waste disposal history for this trench is described in Section 2. For the assessment, we used ^{60}Co and ^{152}Eu activity data for soil samples selected from various depths along a SW-NE transect of the trench (collected in 1974, Dorian and Richards 1978). Because these data were collected in 1974 (^{60}Co half-life, 5.263 yr and ^{152}Eu half-life, 12.7 yr) we decay-corrected the reported values. The preliminary assessment was conducted using the following assumptions:

1. All reported activities were measured using soil fractions finer than gravel.

2. The particle size distribution and contaminant partitioning data obtained on a single sample from the middle of the trench in 1993, is representative of the soil in the whole waste trench.

3. Contaminant distribution measured along a single longitudinal transect is representative of contaminant distribution along the width of the trench.

To display the distribution of ^{60}Co and ^{152}Eu activities in this waste trench, the corrected whole soil activities for these contaminants were plotted and contoured (Figures 4.1 and 4.2). The distribution data indicates that the highest activities of ^{60}Co and ^{152}Eu are found in two locations within the trench. The first “hot spot” is a location about 100 ft downstream from the effluent inlet and at a depth of about 30 ft. The second “hot spot” is located near the northeast end of the trench at a depth of about 20 ft. It appears that these contaminants have moved deeper into soil profile near the inlet possibly due to higher rate of effluent infiltration at this location. The second location showing the highest activities were encountered near the northeast end of the trench at a depth of about 20 ft where the effluent flow rate was probably the lowest within the trench.

These ^{60}Co and ^{152}Eu activity distribution data were used to estimate soil volumes that require remediation because they exceed RAG values of 1.4 and 3.3 pCi/g for these radionuclides respectively. Calculations were also made to estimate the volumes of soil that would be needlessly remediated if soil sampling were to be conducted selectively to exclude all particle sizes coarser than sand (>4.75 mm) without any corrections for sampling bias.

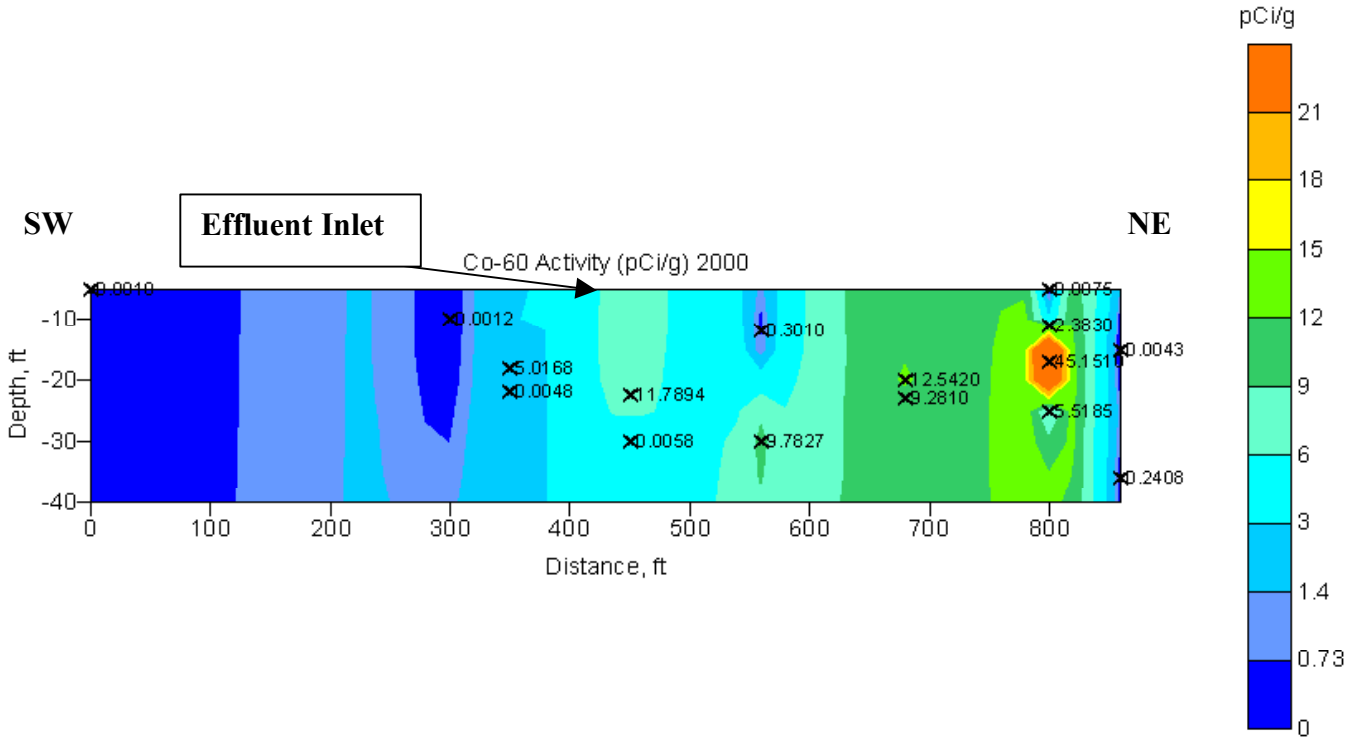

Figure 4.1. Depth Distribution of Corrected ^{60}Co Activities Along a Longitudinal Transect Through 116-C-1 Waste Trench

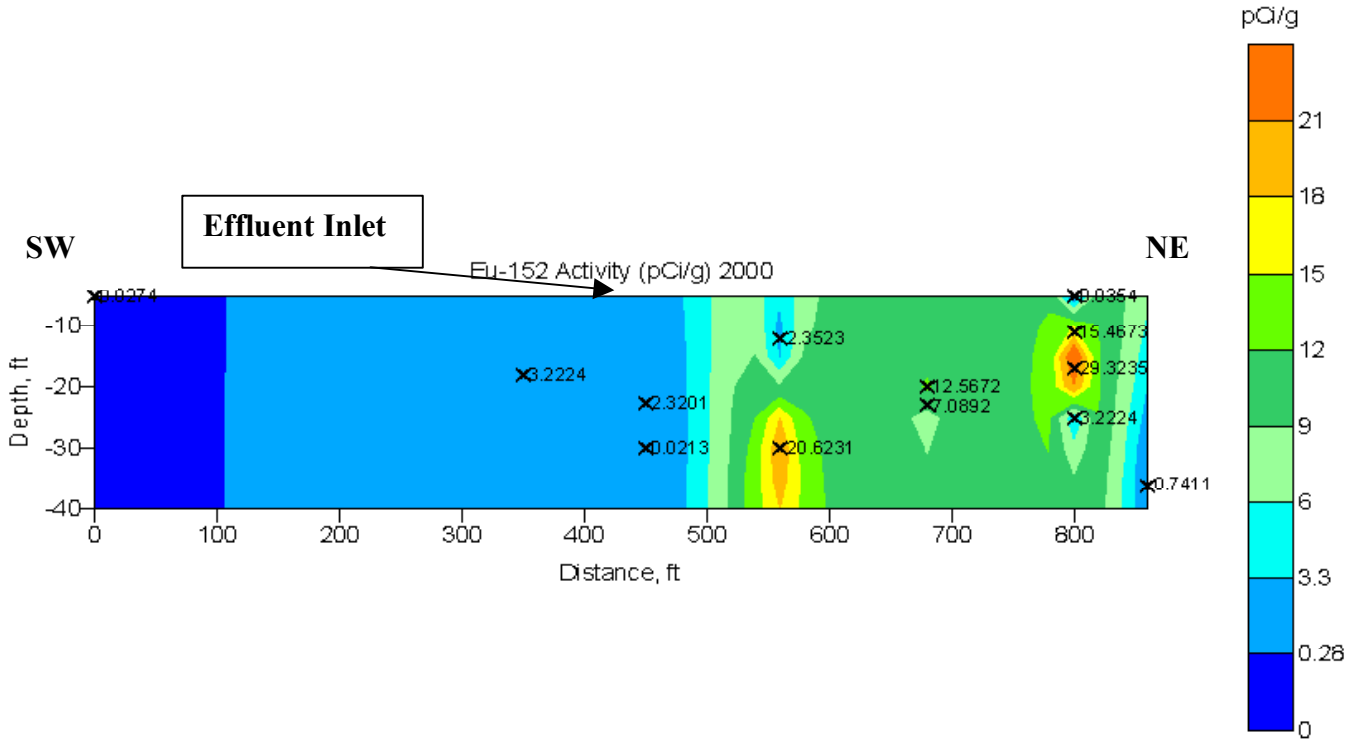

Figure 4.2. Depth Distribution of Corrected ^{152}Eu Activities Along a Longitudinal Transect Through 116-C-1 Waste Trench

The contaminated areas surrounding the 116-C-1 waste trench were estimated according to each of the scenarios (Figure 4.3). First approximation estimates of the volumes of contaminated soil are listed in Table 4.1. The estimates indicate that excess soil volume to be remediated without sampling bias

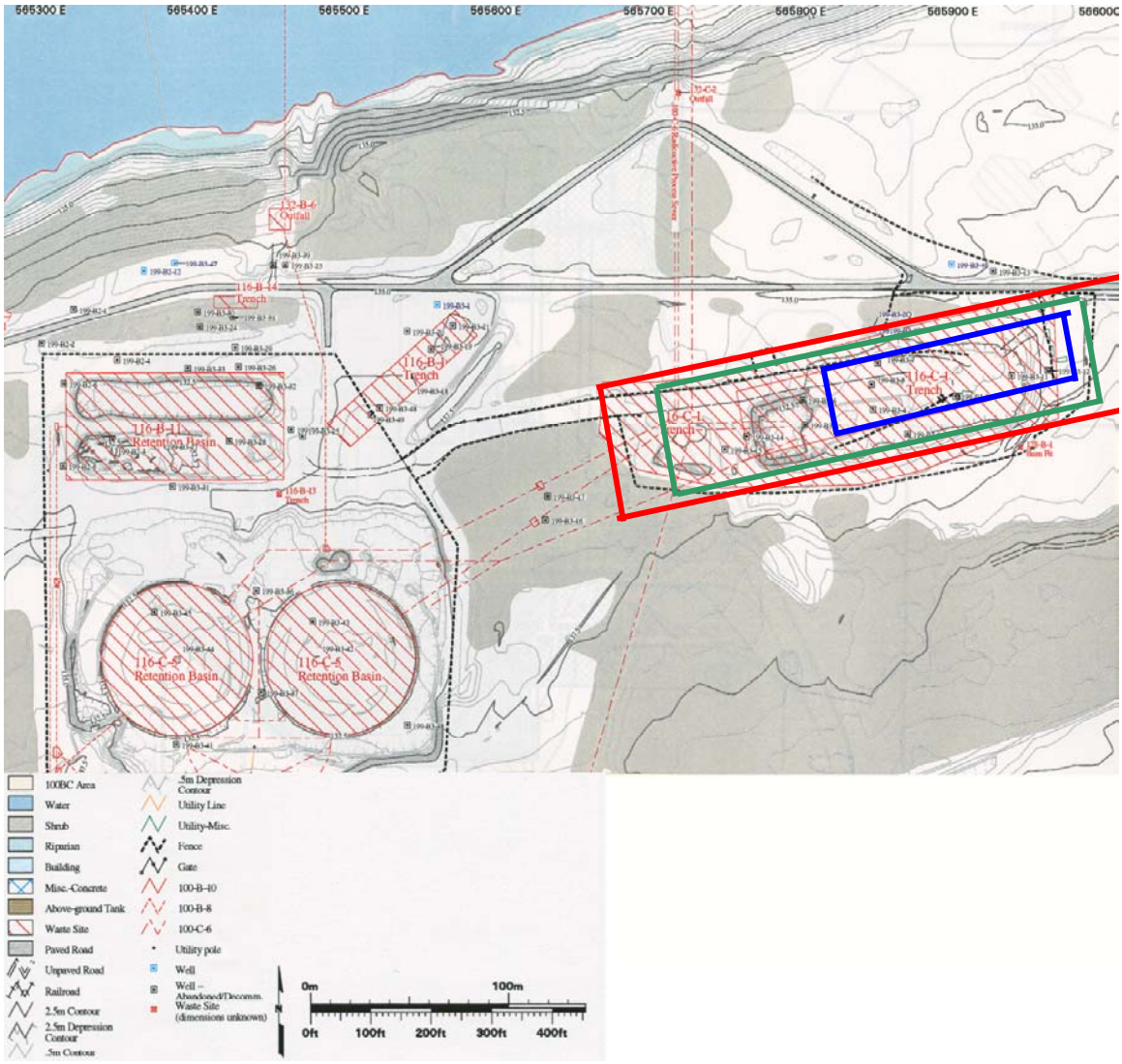

Figure 4.3. Estimated Area of Contamination from Radionuclide Distribution Along a Longitudinal Transect Through 116-C-1 Waste Trench. Blue lines mark the RAG limits for (<3.3 pCi/g) ^{152}Eu activity in whole soil. The green lines mark the RAG limits (<1.4 pCi/g) for ^{60}Co activity in whole soil. The red lines mark the estimated limits of ^{60}Co and ^{152}Eu contamination without SBF corrections.

corrections is about 57% for ^{60}Co and about ~280% for ^{152}Eu. These estimates showed that excess remediation volumes are a function of radionuclide distribution in the soil and their SBF for the contaminated site. This preliminary assessment showed that, higher SBF values leads to estimates of greater contaminated soil volumes and, if uncorrected, would result in over-remediation of a waste site.

Table 4.1. Estimates of Contaminated Soil Volume Surrounding the 116-C-1 Waste Trench

Radionuclide	Vol w/o SBF Corrections (yd^3)	Vol SBF Corrections (yd^3)	Excess Vol Remediated (yd^3)
^{60}Co	~296,600	~189,300	~107,300
^{152}Eu	~189,300	~66,800	~122,500

5.0 Conclusions and Recommendations

A detailed analysis of particle size-radionuclide distribution data for selected coarse-grained soils from 100 and 300 Hanford Area waste sites indicated that

- In these gravelly soils, the activity/concentration of any COC per unit mass in any finer fractional soil material was consistently biased higher than the activity/concentration of the same COC in the whole soil (all grain sizes sampled).

- The degree of sampling bias for each COC could be quantified as an SBF that relates the activity/ concentration in any finer size fraction of a soil to the activity/concentration in the whole soil.

- SBF for any COC in a soil increases with decreasing particle size sampled. SBFs for different COCs in a soil can vary significantly (up to an order of magnitude) depending on their chemical properties and affinity for specific mineral surfaces.

- SBF for a single COC in soil samples obtained from different locations from the same waste site can be significantly different (two to three times higher than the lowest value), reflecting spatial variability engendered by the source, type, and distribution of contamination.

- SBF for a COC in soils from different waste sites were different, and these values were affected by the soil texture, the degree, the source, and the distribution of contamination.

- Waste site soils would be over-remediated (remediated below RAG values) without SBF corrections. SBF corrections could be made by using CRAL computed from RAG and SBF (for selected particle-size finer) for each COC.

- The soils in the vicinity of 116-C-1 waste trench would be over-remediated (excavation of ~57% to 280% of excess soil volume) without appropriate SBF corrections for contaminant radionuclides ^{60}Co and ^{152}Eu.

Based on these analyses and conclusions, we recommend that

- A sampling grid should be established (using a geostatistical basis) for each waste site, and representative whole soil sample should be drawn from selected depth intervals as specified in ASTM C-702. The quantities of whole soil selected for analysis should be based on the standard practice as specified in ASTM D-2487-98 (Appendix A).

- The particle size and COC distribution in soil samples should be determined by wet sieving (ASTM D 422), and each soil fraction of each sample should be chemically and radiologically characterized.

- The SBF for each COC for a specified particle size finer, and CRAL values for each COC in each sampling grid should be computed.

- During remediation, representative soil samples for the same specified particle size finer should be drawn for monitoring the activity/concentration of each COC and sample grid, and relevant COC-specific CRAL values should be used to establish the limits of remediation.

6.0 References

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Appendix A

ASTM Specifications for Minimum Sample Quantity Needed for Accurate Particle Size Analysis of Soils

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ASTM Specifications for Minimum Sample Quantity Needed for Accurate Particle Size Analysis of Soils

According to ASTM D2487-98, the quantity of soil sample to be used for accurate particle size distribution determination depends on the maximum particle size of the soil. The recommended minimum quantities are listed in Table A.1. The quantity of field sample would be reduced to minimum sample quantity needed for analysis by using a sample splitter or by coning and quartering as per ASTM C 702-87.

Table A.1. Minimum Soil Sample Quantity Size Needed for Particle Size Analysis of Soils

Maximum Particle Size (mm)	Sieve Opening (in.)	Minimum Analytical Sample Size – Dry wt (kg)	Field Sample Size – Dry wt (kg)
75	3.0	60	120-240
38.1	1.5	8	16-32
19	3/4	1	2-4
9.5	3/8	0.2	0.4-0.8
4.75	3/16 (#4)	0.1	0.2-0.4

Appendix B

Radionuclide Activity and Mass Relationship

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Radionuclide Activity and Mass Relationship

Mass in grams of a radionuclide with an activity of 1 Ci is calculated as follows.

A unit of radioactivity in Ci is defined as the mass of any radionuclide in which there are 3.70×10^{10} disintegrations per second (-dN/dt).

The mass of a radionuclide is related to the number of atoms N, by the following relationship:

$$M = (N \times \text{atomic wt of the isotope})/N_A$$

where M = mass of the isotope in grams
N = number of atoms of isotope
N_A = Avogadro's number = 6.022×10^{23} moles.

The number of atoms of a radionuclide is a function of decay rate and the decay constant:

$$N = (-dN/dt)/\lambda$$

where -dN/dt = number of disintegrations per second = 3.70 x 1010
λ = decay constant = 0.693/(radionuclide half-life in years, t1/2) x 3.156 x 107

Therefore, $M = ((3.7 \times 10^{10}) \times \text{atomic wt of the isotope} \times t_{1/2})/ (2.1958 \times 10^{-8}) \times (6.022 \times 10^{23})$
$= (2.7981 \times 10^{-6}) \times \text{atomic wt of the isotope} \times t_{1/2}$

1 Ci of activity = $[(2.7981 \times 10^{-6}) \times \text{atomic wt of the isotope} \times t_{1/2}]$ g of radionuclide.