093019

DOI: 10.1007/s00267-004-0098-6

Investigations of Contaminated Fluvial Sediment Deposits: Merging of Statistical and Geomorphic Approaches

RANDALL T. RYTI*
Neptune and Company, Inc.
1505 15th Street, Suite B
Los Alamos, New Mexico, 87544, USA

STEVEN L. RENEAU
DANNY KATZMAN
Earth and Environmental Sciences Division
Los Alamos National Laboratory
Los Alamos, New Mexico, 87545, USA

ABSTRACT / Concentrations of contaminants in sediment deposits can have large spatial variability resulting from geomorphic processes acting over long time periods. Thus, systematic (e.g., regularly spaced sample locations) or random sampling approaches might be inefficient and/or lead to highly biased results. We demonstrate the bias associated with systematic sampling and compare these results to those achieved by methods that merge a geomorphic approach to evaluating the physical system and stratified random sampling concepts. By combining these approaches, we achieve a more efficient and less biased characterization of sediment contamination in fluvial systems. These methods are applied using a phased sampling approach to characterize radiological contamination in sediment deposits in two semiarid canyons that have received historical releases from the Los Alamos National Laboratory. Uncertainty in contaminant inventory was used as a metric to evaluate the adequacy of sampling during these phased investigations. Simple, one-dimensional Monte Carlo simulations were used to estimate uncertainty in contaminant inventory. We also show how one can use stratified random sampling theory to help estimate uncertainty in mean contaminant concentrations.

Contaminants can enter fluvial systems in a number of ways that include erosion or leaching of mine tailings, effluent discharges from point sources, surface runoff from nonpoint sources, redistribution of airborne particulates, or discharges of contaminated groundwater. Some of these contaminants originate as solid particles and others start in solution and eventually bind to particles. Once in streams or rivers, particle-bound contaminants are subsequently redistributed by fluvial processes. Particles carried by floods can be transported long distances from the source and redeposited in the channel or on adjacent floodplains (Lewin and Macklin 1986; Marron 1992; Graf 1994, 1996; Rowan and others 1995; Miller and others 1998; Marcus and others 2001; Malmon and others 2002; Reneau and others 2004). During this process, the contaminated particles are mixed with sediment from other sources and are sorted by particle size, resulting in spatial variations in contaminant concentrations on the landscape. Subsequent floods can partially erode and remobilize contaminated sediment deposits, mixing the particles with upstream sediment, or can bury them with younger sediment. The result of floods acting over many years, combined with temporally varying releases of contaminants from their sources, is a complex, spatially heterogeneous pattern of contamination in fluvial deposits that pose special problems for characterization. Contaminant heterogeneity exists over many spatial scales, including scales of centimeters to meters in a vertical profile and meters to kilometers longitudinally along a fluvial system. To be most effective at characterizing contaminants in fluvial systems, sampling strategies need to explicitly incorporate an awareness of this inherent geomorphic variability (Miller and others 1996; Ladd and others 1998; Rhoads and Cahill 1999).

One approach for investigating complex and heterogeneous sediment deposits has been developed and applied in semiarid fluvial systems within and near the Los Alamos National Laboratory (LANL) in northern New Mexico. This approach combines detailed geomorphically based field investigations with statistically

KEY WORDS: Geomorphology; Contaminant transport; Radionuclides; Stratified random sampling; Plutonium; Cesium-137

Published online ■

*Author to whom correspondence should be addressed; email: rryti@neptuneinc.org

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based sampling and data analysis to efficiently address a series of objectives. These objectives include developing and refining conceptual models of contaminant distribution and transport, evaluating the potential for adverse effects on human health and the environment, and evaluating potential remediation alternatives. Of particular importance is the need to obtain representative concentrations of contaminants from a heterogeneous system that are of sufficient quality for use in risk assessments and decision-making.

This article focuses on the investigation of sediment deposits along 19 km of Los Alamos and Pueblo canyons upstream and downstream from LANL contaminant sources. It includes discussion of both the geomorphic approach to sediment characterization adopted here and the statistical approach used to guide sample collection and evaluate contaminant sample results. Key aspects of the geomorphic approach are to select investigation areas or reaches spaced along the fluvial system and to map and characterize sediment deposits or geomorphic units within these reaches. This article illustrates how measures of uncertainty in indicator contaminant concentrations are tracked through sampling phases and how this information is used to guide additional sampling in a multiphase sampling campaign. Geomorphic information was used to quantitatively define sampling needs using simple statistical formulas, and information was collected to test key assumptions and to help reduce uncertainties in contaminant concentrations and inventory. In this article, we also compare results using the geomorphic approach to results from other methods previously used to characterize contamination in these canyons.

It is important to note we use the term "uncertainty" to include both "variability" (uncertainty in average concentration resulting from inherent heterogeneity within a geomorphic unit) and "incertitude" (uncertainty resulting from incomplete knowledge). Incertitude can be reduced through more complete characterization, but variability cannot be reduced. For the purposes of this article, we are evaluating total uncertainty and not distinguishing between these two categories.

Study Area

The study area includes 19 km of canyon within the 152-km^2 Los Alamos Canyon watershed, including the major tributary of Pueblo Canyon and smaller tributaries to Los Alamos and Pueblo canyons (Figure 1). Los Alamos Canyon heads in the Jemez Mountains and drains east across the Pajarito Plateau to the Rio Grande, a major regional river. These canyons have received contaminants from LANL technical areas and the Los Alamos town site and also from higher elevation areas to the west. Significant contaminant releases began with initiation of the Manhattan Project in 1943, during World War II. The largest amounts of contaminants were released from laboratory facilities in the 1940s to the 1960s, with declining amounts in subsequent years.

Methods

The technical approach for sediment characterization used here was modified from the "representative reach" concept of Graf (1994), using a geomorphic examination of a series of discrete reaches or segments of the canyon bottom. These reaches extend from the contaminant sources downstream to the Rio Grande and are used to document general spatial trends in contaminant concentrations and inventory. Based on these criteria and using aerial photographs, previous sediment characterization data, and field surveys, 34 sampling reaches were established to characterize the Los Alamos and Pueblo watershed (Figure 1and Table 1). Table 1 summarizes the estimated volume of post-1942 sediment (sediment postdating initiation of the Manhattan Project) in each of the reaches, their location relative to the Rio Grande, and the primary contaminants in each reach. Sediment volume is estimated from geomorphic mapping and characterization that delineates the spatial extent and thickness of post-1942 sediment deposits (Reneau and others 1998a, 1998b, 1998c, 2004; Katzman and others 1999; LANL 2004). A number of methods are used to delineate the areas of these deposits, including examination of historical aerial photographs, observations of topographic and vegetation breaks, flood deposits, and flood debris, and analytical measurements of contaminants. Thickness of sediment deposits is typically measured at channel bank exposures and in soil pits.

The contaminant data from these reaches provide estimates of contaminant inventory (amount) and concentration over multiple spatial scales. The broadest scale represents inventory or concentration for affected media in the entire watershed, and one can also estimate inventory or concentration for a subwatershed. Another useful spatial scale is the inventory or concentration for each reach. Evaluations are also possible at smaller scales (geomorphic unit, sediment facies, and flood layer). Because contaminants in this study are measured for sediments within reaches, assumptions about the unsampled parts of the watershed or subwatershed are needed to estimate

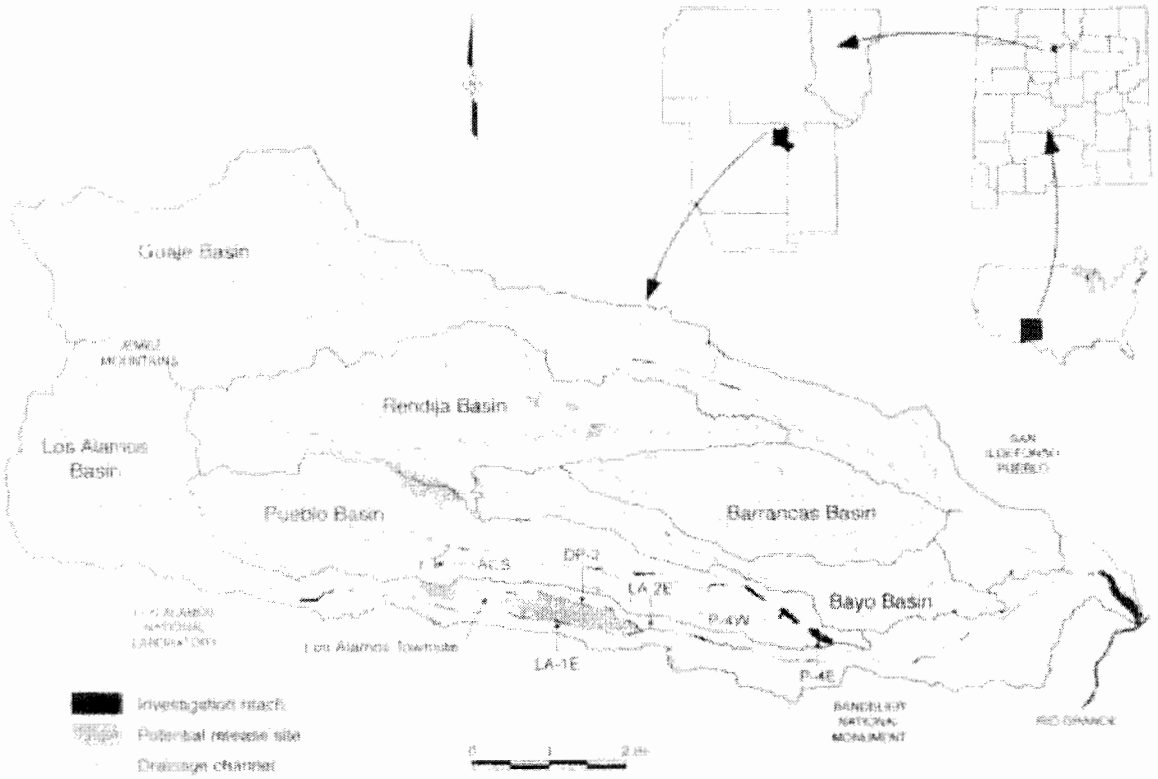

Figure 1. Map of Los Alamos/Pueblo watershed showing the sediment investigation reaches. Reaches discussed in text are labeled.

inventory or average concentrations for these broader spatial scales.

The formula used to calculate contaminant inventory for radionuclides is

$$I_z = \sum_{j=1}^{n} c_{zj} \cdot a_j \cdot t_j \cdot \delta_j \cdot (1 - f_{gj}), \qquad (1)$$

where

I_z = inventory of contaminant z (pCi/g);

c_{zj} = average concentration of contaminant z for geomorphic unit j (pCi/g);

a_j = area of geomorphic unit j (m^2);

t_j = average thickness of geomorphic unit j (m);

δ_j = bulk density of geomorphic unit j (kg/m^3);

f_{gj} = gravel fraction of geomorphic unit j (unitless), and

n = number of geomorphic units being totaled for inventory.

This formula can be applied at any of the above-discussed spatial scales. All that is required is that the number of geomorphic units comprising the total sediment volume be specified along with estimates of the inputs to this equation presented above. This inventory calculation assumes that the gravel fraction (>2 mm) for the geomorphic unit contains no contaminants, based on the preferential adsorption of contaminants to fine-grained particles and analyses of particle size fractions that show decreasing contaminant concentrations with increasing particle size.

Contaminant inventory at the reach scale was used as a quantitative measure of the uncertainty in sediment characterization. According to statistical theory, uncertainty in the estimated inventory should decrease as the number of samples collected to characterize the population of interest increases (Gilbert 1987). Thus, uncertainty in contaminant inventory should decrease through phases of sediment investigation. In each investigation phase, the planned number of samples is based on initial estimates of area, thickness, bulk density, and contaminant concentrations of the mapped geomorphic units in each reach. Our approach merges

Table 1. Summary of investigation reaches in Los Alamos and Pueblo canyons

Subwatershed	Reach	Distance from Rio Grande[a] (km)	Reach length (km)	Estimated post-1942 sediment volume (m^3)	Primary contaminants[b]
Acid Canyon	AC-1	19.3	0.04	827	PAHs[c]
	AC-2	19.2	0.09	779	PAHs
	ACS	19	0.29	686[d]	Pu-239,240
	AC-3	18	0.42	1320	Pu-239,240, PCBs[e]
DP Canyon	DP-1 West	16.4	0.07	210	PAHs
	DP-1 Central	16.3	0.10	124	PAHs
	DP-1 East	15.5	0.10	323	PAHs
	DP-2	14.4	0.32	2400[d]	Cs-137, Sr-90
	DP-3	13.8	0.21	446	Cs-137, Sr-90
	DP-4	13.1	0.45	1220	Cs-137, Sr-90
Los Alamos Canyon	LA-0	19	0.12	n.c.[f]	None
	LA-1 Far West	18.0	0.21	n.c.	None
	LA-1 West Plus	17.5	0.14	633	None
	LA-1 West	16	0.37	2730	None
	LA-1 Central	15.7	0.39	1890	None
	LA-1 East	14.8	0.43	2720	None
	LA-2 West	13	0.21	1300	None
	LA-2 East	12.6	0.68	4720[d]	Cs-137, Sr-90
	LA-2 Far East	11.5	1.00	12200	Cs-137, Sr-90
	LA-3 West	10	0.49	3350	Cs-137, Sr-90
	LA-3 East	9.32	0.44	2160	Cs-137, Sr-90
	LA-4 West	6.04	0.52	5160	None
	LA-4 East	5.05	0.29	2620	None
	LA-5	0.88	1.41	119000	None
Pueblo Canyon	P-1 Far West	18.1	0.10	n.c.	None
	P-1 West	17.9	0.32	n.c.	None
	P-1 East	17.7	0.51	3540	Pu-239,240, PAHs
	Walnut Canyon	17.5	0.06	n.c.	None
	P-2 West	14.6	0.51	9060	Pu-239,240
	P-2 East	13.3	0.46	21800	None
	P-3 West	11.4	0.51	17300	Pu-239,240
	P-3 East	10.4	0.54	37500	None
	P-4 West	9.49	0.52	51400	Pu-239,240, PAHs
	P-4 East	8.6	0.60	42200	None

[a]Approximate distance to midpoint of reach.
[b]Cancer risks >1E-5 or radiation dose levels >15 mrem/year for residential land use using maximum sample results, and the analytes listed contribute >90% of the estimated cancer risk or dose.
[c]Polycyclic aromatic hydrocarbons.
[d]Before sediments were removed in remediation activities.
[e]Polychlorinated biphenyls
[f]n.c. = not calculated (reaches with no significant contamination).

the concepts behind statistical sampling for stratified populations with a detailed understanding of contaminant distribution based on geomorphic processes.

We followed these steps during the phased reach investigations:

a. *Conduct initial geomorphic mapping.* In each reach, a detailed map was made showing the spatial distribution of post-1942 sediment deposits, with the reach subdivided into geomorphic units with varying physical characteristics and/or varying contaminant concentrations. The three basic geomorphic subdivisions used in this investigation are active channels, abandoned channels, and floodplains, and the relations of these units in one reach are illustrated in Figure 2. Active channels ("c1") refer to areas inundated during frequent flows under current conditions, which are typically not vegetated. Sediment here is dominated by coarse to very coarse sand and gravel (coarse facies sediment). Abandoned channels ("c2," "c3," etc.) refer to areas occupied by the stream channel sometime since initial contaminant releases (post-1942), but abandoned following channel migration and/or channel incision. These areas are usually vegetated, and sediment here typically includes

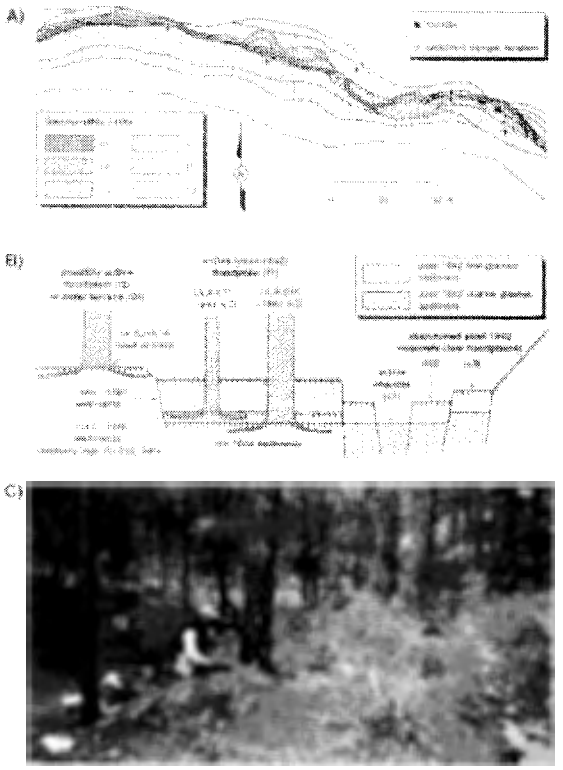

Figure 2. Example of geomorphic units in reach LA-1 East. (A) Geomorphic map of part of reach, showing sample locations. (B) Simplified cross-section showing generalized topography, relation of coarse and fine facies sediment, and partial burial of trees on floodplain. (C) Photograph of area shown in (B). Tree ULA-010, a ponderosa pine dated at approximately 1844 AD, is being cored for dendrochronological analysis, and tree ULA-011, a ponderosa pine dated at approximately 1889 AD, is to the left. The base of ULA-010 is buried by 48 cm of sediment, and the base of ULA-011 is buried by 85 cm of sediment.

coarse to very coarse sand and gravel (coarse facies sediment) overlain by finer-grained sediment deposited by overbank floodwaters (fine facies sediment, dominated by fine to very fine sand, silt, and clay). Floodplains ("FL," "F2," etc.) refer to older surfaces that have also been inundated since initial contaminant releases, but where post-1942 sediment is dominantly fine grained. These basic geomorphic subdivisions are often further divided based on age and/or contaminant levels. For example, in one reach, eight separate abandoned channel units of different age were distinguished based on field mapping and examination of sequential aerial photographs. Age differences are important because contaminant concentrations typically decrease over time following cessation of releases (e.g., Renéau and others 2004). From 3 to 13 different post-1942 geomorphic units have been mapped in various reaches, with the number depending on the degree of geomorphic complexity and the variability in contaminant concentrations. Digitizing the maps allowed areas of each geomorphic unit to be calculated, and examination of bank exposures and pits dug during mapping allowed unit thickness to be estimated. Relative variations in some contaminant concentrations (e.g., cesium-137) could be initially identified using field radiological measurements in many reaches. The procedures used to identify and subdivide these units are discussed in more detail elsewhere (Renéau and others 1998a, 1998b, 1998c, 2004; Kaufman and others 1999).

b. *Implement stratified sampling calculations.* Concentrations and the variability in concentrations of an indicator contaminant were estimated for each unit. This information was entered into calculations to allocate the number of samples based on stratified random sampling theory (Gilbert 1987). The optimal allocation for stratified random sampling designs is based on equation 5.10 of Gilbert (1987). The equation used for either area or volume weighted calculations is

$$f_h = \frac{(v_h/v_{total}) \cdot \sigma_h}{\sum_{h=1}^{L} (v_h/v_{total}) \cdot \sigma_h} \qquad (2)$$

where: f_h = fraction of samples in the *h*th stratum or sampling unit;

v_h = area (or volume) of the *hth* stratum or sampling unit;

v_{total} = total area (or volume);

σ_h = standard deviation of concentration in the *hth* stratum or sampling unit; and

L = the total number of stratum or sampling units considered.

Equations 2 can be applied to determine the fraction of samples to allocate to different units at any phase of sample collection. If applied before contaminant data are obtained, then one could use historical information or professional judgment, based on experience at other sites, to estimate the expected variability in contaminant concentrations for each stratum. Equation 2 could also be applied at a variety of spatial scales. One can use Equation 2 to allocate samples between or within investigation reaches. One can also calculate the

mean and standard error of the stratified mean concentration based on Equations 5.3 and 5.5 in Gilbert (1987) and presented here as Equations 3 and 4

$$\bar{x}_{st} = (v_h/v_{total})\bar{x}_h, \quad (3)$$

where:
$\bar{x}_{st}$ = stratified mean;
$\bar{x}_h$ = mean of the hth stratum or sampling unit;
v_h = volume of the hth stratum or sampling unit;
v_{total} = total volume; and
σ_h = mean concentration in the hth stratum or sampling unit.

$$s(\bar{x}_{st}) = \sqrt{\sum_{h=1}^{L} \frac{(v_h/v_{total})^2 \sigma_h^2}{n_h}} \quad (4)$$

$s(\bar{x}_{st})$=standard error of the stratified mean;
v_h = area (or volume) of the hth stratum or sampling unit;
v_{total} = total area (or volume);
σ_h = standard deviation of concentration in the hth stratum or sampling unit;
n_m = number of samples in the hth stratum or sampling unit; and
L = the total number of stratum or sampling units considered.

Equations 3 and 4 can be used to calculate the standard error for the additional data being planned and to calculate the standard error based on the combination of existing data and additional data. The only modification for using these formulas with additional data plus existing data is that the number of samples per stratum will depend on the previous sample allocations to the geomorphic units.

c. *Determine number of samples needed to reduce uncertainty.* To determine the number of samples to collect, uncertainties in the weighted mean concentrations and in the standard error of the mean were explored using one-dimensional Monte Carlo simulations with the Crystal Ball™ add-in to Microsoft Excel™. Uncertain variables in these simulations included the standard deviations and the thickness of contaminated sediment. Distributions of the uncertain variables were either based on existing data or professional judgment. The distributions used in these simulations might include normal (N), lognormal (LN), exponential (EXP), uniform (U), discrete (D), or extreme value (EXTREME). It is beyond the scope of this article to provide a specific rationale for the distributions and parameters selected for each uncertain variable, but selected uncertain variables will be discussed in some detail to illustrate the process for developing these distributions. For example, the concentrations of contaminants within a geomorphic unit were analyzed to determine mean concentrations and the standard error of the mean. These data were also evaluated using goodness-of-fit testing protocols available in Crystal Ball. We also used theoretical arguments and knowledge about environmental contamination to select certain underlying distributions (e.g., the lognormal is a typical distribution for environmental contaminant concentrations). Sensitivity analyses are recommended to determine the impact of various statistical distributions on the results. Where there was little information on which to base the statistical distribution, a uniform distribution was selected. Geomorphic unit areas are treated as fixed values because the areas are relatively well defined, and, in particular, the total area of post-1942 sediment deposits are typically well constrained by topography. Simulation result variables or "forecasts" (the Crystal Ball terminology for simulation results) were exported and the simulation results were evaluated using other software, including Microsoft Excel and JMP™ statistical software. The fraction of samples per stratum was calculated using Equation 2 and the number of samples per stratum (n_h) was calculated by multiplying the number of samples (n) times the fraction per stratum. The stratified standard error was calculated based on Equation 4 for the additional samples (n_h) and for the additional samples plus the existing samples ($n_h + n_{old}$). We also calculated the stratified mean based on Equation 3.

d. *Collect additional samples.* Samples are collected based on the statistical calculations for the indicator contaminants, and additional samples are also collected to test aspects of the geomorphic conceptual model of historical contaminant transport and to provide data on the nonindicator contaminants. Samples were generally collected from a subset of locations previously examined as part of geomorphic characterization. In some cases, additional test pits were dug or stream bank locations were described to provide a larger set of potential sample sites. The exact sample locations were selected judgmentally instead of randomly to provide good spatial distribution and to include representative sediment layers in each unit.

e. *Determine uncertainty in inventory and initial risk estimates using Monte Carlo simulations.* Contaminant results from the previous sampling events are analyzed for several purposes. Sample results are assessed to determine if geomorphic deposit area or thickness should be revised. Average contaminant concentrations are estimated for each geomorphic unit. The inventory of indicator contaminants is calculated using available data on area, thickness, bulk density, and average concentration. Uncertainty in contaminant inventory is calculated using Monte Carlo simulation methods similar to those presented in step c, with the addition of bulk density that is needed for the inventory calculations. Bulk density is assumed to be a fixed parameter, and it differs between coarse-grained and fine-grained sediment deposits. Distributions of the uncertain variables were selected based on existing data or professional judgment as stated in step c. Simulation result variables were exported and the simulation results were evaluated using other software, as discussed in step c.

f. *Determine if additional samples are needed to reduce uncertainty.* Uncertainty in contaminant inventory, initial risk estimates, and the conceptual model are evaluated along with data quality to determine if additional data are needed. Specific limits on uncertainty in inventory or risk were not developed for this project, although various methods are available from the simple use of thresholds to more complex decision analysis tools. If additional data are determined to be needed based on a review of contaminant inventory and risk estimates by the project team, then the process repeats at step b with the revised estimates of geomorphic unit area, thickness, and concentrations.

These steps are illustrated with examples from selected investigation reaches. Reaches LA-2 East and P-4 West were selected because they contain the two primary radiological contaminants in this watershed and are also two of the more intensively investigated areas.

The results from the geomorphic approach were also compared to results obtained from other investigations in these canyons. As an example, we compared sample results from investigations of plutonium-239,240[1] concentrations in reach ACS (south fork of Acid Canyon) that spanned more than two decades. The objective of all of these investigations was to determine the concentrations of contaminants in locations downstream from the contaminant source. The first investigations were conducted under the Formerly Utilized Sites Remedial Action Program (FUSRAP). Samples were collected systematically in 1977 along transects intersecting the ephemeral stream channel, and transect locations were positioned in a geometrically increasing distance (20 m, 40 m, 80 m, etc.) from the contaminant source (Stoker and others 1981). Later sampling in reach ACS was conducted as part of a Resource Conservation and Recovery Act (RCRA) Facility Investigation (RFI). The RFI samples were collected systematically in 1992 and 1993 from transects at several locations down the stream channel (LANL 1996). Samples were subsequently collected in reach ACS by the New Mexico Environment Department (NMED) in 1999 using a biased sampling approach that included aspects of the geomorphic approach (Yanicak and others 1999). NMED sample locations were selected after a visual survey of the sediment deposits in reach ACS and samples were collected from fine-grained deposits that generally have higher contaminant concentrations. Finally, samples were collected in 1999 following the geomorphic approach outlined in this article (Reneau and others 2000). All of these studies used the same basic laboratory techniques, specifically alpha spectroscopy following standard acid digestions. In addition, all standard quality control and quality assurance protocols were followed in these studies, including analysis of spikes and blanks. Sample collections methods were all basically the same and used stainless-steel scoops to collect the sample, which was homogenized and split into aliquots for laboratory analysis. Thus, data collected in reach ACS provide a comparison of systematic sampling approaches to the geomorphic approach.

We also used simulations to estimate how uncertainty in contaminant inventory would change based on the sampling approach. One set of simulations was used to sample the data represented by various geomorphic units in proportion to their relative volumes. Another set of simulations was used to sample geomorphic units in proportion to their relative areas. Thus, these simulations provide a comparison to random sampling within the area of contaminated sediments to the complete geomorphic assessment and characterization. These sampling simulations consisted of the following steps:

1. Assemble information for the geomorphic units in a reach: area, thickness, and contaminant concen-

[1]Isotopes plutonium-239 and plutonium-240 are unresolved by alpha spectroscopy, although thermal ionization mass spectrometry analyses indicate that 98.5% of the plutonium-239,240 in Pueblo Canyon consists of plutonium-239 (Gallaher and Efurd 2002).

tration. For the purposes of these simulations, we assumed that contaminant sample results for each geomorphic unit are derived from a lognormal statistical distribution, which is a common distribution for environmental contaminant data. The parameters of the lognormal distribution were determined by the available data for each geomorphic unit.

2. Use a loop of $j = 1200$ simulations to collect a set of n samples of contaminant data. This loop constitutes the "outer" loop in the programming sense.
3. Use a loop of $i = 1, n$ samples ($n = 10, 20, 40$, or 80) for each of the j simulations (step 2) collected at random from the geomorphic units. This loop constitutes the "inner" loop in the programming sense. Geomorphic units are selected at random with probability of selection equal to the relative area or volume of the geomorphic units. The associated value obtained from a selected unit is a simulation result from the probability distribution for that unit. The average of the different inventory estimates and other summary statistics were calculated from each set of samples.

The simulations were implemented using the bootstrap tool of Crystal Ball, which is a two-dimensional Monte Carlo procedure using probability distribution models for the uncertain variables. Simulation result were saved and were evaluated with by calculating standard summary statistics and creating plots using other software, including Microsoft Excel and JMP statistical software.

Note on units: The nonmetric unit curie (Ci) is used in this articler to describe concentrations and amounts of radionuclides instead of the metric unit becquerel (Bq), because analytical laboratories in the United States report concentrations of radionuclides in units of picocuries per gram (pCi/g) (1 pCi/g = 0.0367 Bq/g). This convention also simplifies comparisons to other published studies (e.g., Graf 1994, 1996).

Results

Sample Allocation

An example application of the statistical approach to sample allocation is provided for reaches P-4 West and P-4 East (Table 2 and Figure 3). This example uses one-dimensional Monte Carlo simulations to evaluate uncertainty in the expected standard error of the mean versus the number of additional samples collected. These simulations were performed using Crystal Ball, and the structure of the model and input values are provided in Table 2. Standard deviation and thickness of each geomorphic unit were assumptions or uncertain variables in the Crystal Ball model. Geomorphic units appear twice in Table 2 where there is a fine and coarse sediment facies in that unit. Thus, geomorphic units represent the x and y dimensions on a map and the facies represents the z or depth dimension. In some cases, a geomorphic unit might include more than one occurrence of a fine or coarse sediment deposit (e.g., a fine deposit with coarse deposits above and below). We also made the number of samples an assumption in the model, and this variable was also included as a result variable. The weighted mean concentration and the standard error of the mean were also identified as forecast variables. The result variables were exported from Crystal Ball into JMP for further statistical analysis and plotting (Figure 3).

In the sample allocation example, the variables considered to be uncertain included the thickness of the geomorphic units and the standard deviation of the contaminant concentration in that deposit. Thickness distributions are primarily based on measured thickness of post-1942 sediment deposits. If the standard error of the mean is found to be sensitive to uncertainty in the thickness of some units, it is relatively simple and inexpensive to reduce uncertainty in the thickness with additional field measurements. The primary purpose of these simulations is to determine how the estimated uncertainty in mean contaminant concentrations will be reduced by additional collection and chemical analysis of sediment samples. Given that there is typically relatively little uncertainty in sediment deposit thickness, most of the uncertainty in the standard error of the mean will be related to uncertainty in the standard deviation of the contaminant concentration. In this example, the mean of the distributions selected for the standard deviation was based on point estimates of existing data—the standard deviations of the existing contaminant data. The variability of the standard deviation was selected by professional judgment as either 40% or 50% of mean value.

To evaluate how many more samples would be needed to characterize contaminant concentrations in the reach, plots showing the expected decrease in contaminant uncertainty with increasing numbers of samples were reviewed (Figure 3). All of these plots show a decrease in the estimated standard error with increasing sample numbers. The estimate standard error of the mean was calculated based on the projected allocation of additional samples (Figure 3b) or a combination of the existing samples and additional samples (Figure 3a). Because the standard error is most relevant as a relative proportion of the mean va-

Table 2. Volume-variance weighted stratified random sample allocation for Reaches P-4 West and East, using Gilbert (1987, Eq. 5.10).

Reach[a]	Binned Geomorphic Unit: Name	Facies	Area (m^2)	Thickness[b] (m)	Standard deviation of contaminant concentrations in unit (SD) (σ_h)[c]	Proportion of samples allocated to unit (f_h) using Equation 2	Number of samples allocated to unit (n_h)	Unit standard error of contaminant concentrations $s(\bar{x}_{st})$ for n_h only using Equation 4	Previous samples collected in unit (n_{old})	Unit standard error of contaminant concentration $s(\bar{x}_{st})$ for n_h and n_{old} using Equation 4	Stratum mean ($\bar{x}_h$) contaminant concentrations using Equation 3	Stratified mean calculation of contaminant concentration s
P4W	c1,2,3	Coarse	9,900	0.5	1	4.4%	0.9	0.0080	37	0.0002	1.56	0.130
	c4a	Coarse	8,060	1	1	7.1%	1.4	0.0130	0	0.0130	1.56	0.212
	c4b,5	Coarse	8,090	1	4	28.5%	5.7	0.0522	4	0.0307	8.43	1.15
	f1	Coarse	18,600	0.01	4	0.7%	0.1	0.0012	0	0.0012	8.43	0.027
	c6	Coarse	4,900	0.8	3	10.4%	2.1	0.0190	2	0.0097	1.54	0.102
	c1,2,3,4a	Fine	18,000	0.05	4	3.2%	0.6	0.0058	4	0.0008	5.69	0.086
	c4b,5,6,f1,f1a	Fine	31,600	0.1	4	11.1%	2.2	0.0204	0	0.0204	14.0	0.746
P-4E	c1,2,3	Coarse	28,500	0.5	1	12.6%	2.5	0.0230	0	0.0230	1.56	0.375
	f1,f1a	Coarse	33,800	0.05	1	1.5%	0.3	0.0027	1	0.0006	1.56	0.044
	c3	Coarse	13,000	0.6	1	6.8%	1.4	0.0125	0	0.0125	1.56	0.205
	c1,2,3	Fine	28,500	0.05	1	1.3%	0.3	0.0023	0	0.0023	5.69	0.137
	f1,f1a	Fine	33,800	0.1	4	11.9%	2.4	0.0218	4	0.0081	5.69	0.324
	f2	Fine	15,000	0.1	0.5	0.7%	0.1	0.0012	6	<0.0001	0.61	0.015
	Sums						20	0.43	58	0.35		3.55

Note: Cells highlighted in black are the assumptions used in the Crystal Ball simulations, and the value in the cell is the expected or mean value of the distribution. The gray cells are the Crystal Ball result variables.

[a]P4W = P-4 West, P4E = P-4 East.

[b]Mean values for the distributions for these assumptions were set to the value in this table. Specific distributions differed for various mean values: mean=0.01, distribution=EXP(0.01); mean=0.05, distribution=LN(0.05,0.03); mean=0.1, distribution=LN(0.1,0.05); mean=0.5, distribution=LN(0.1,0.05); mean=0.6, distribution=N(0.6,0.24); mean=0.8, distribution=N(0.8,0.32); mean=1, distribution=N(1,0.4).

[c]The distribution used for these assumptions were lognormal with the mean set at the value in this table and the standard deviation was 50% of the mean.

[d]Mean values for the distributions for these assumptions were set to the value in this table. Specific distributions differed for various mean values: mean=0.61, distribution=EXTREME(0.43,0.31); mean=1.54, distribution=LN(1.54,2.74); mean=1.56 distribution=LN(1.56,0.92); mean=5.69, distribution=LN(5.69,4.24); mean=8.43, distribution=LN(8.43,4.07); mean=14.0, distribution=LN(14.0,17.0).

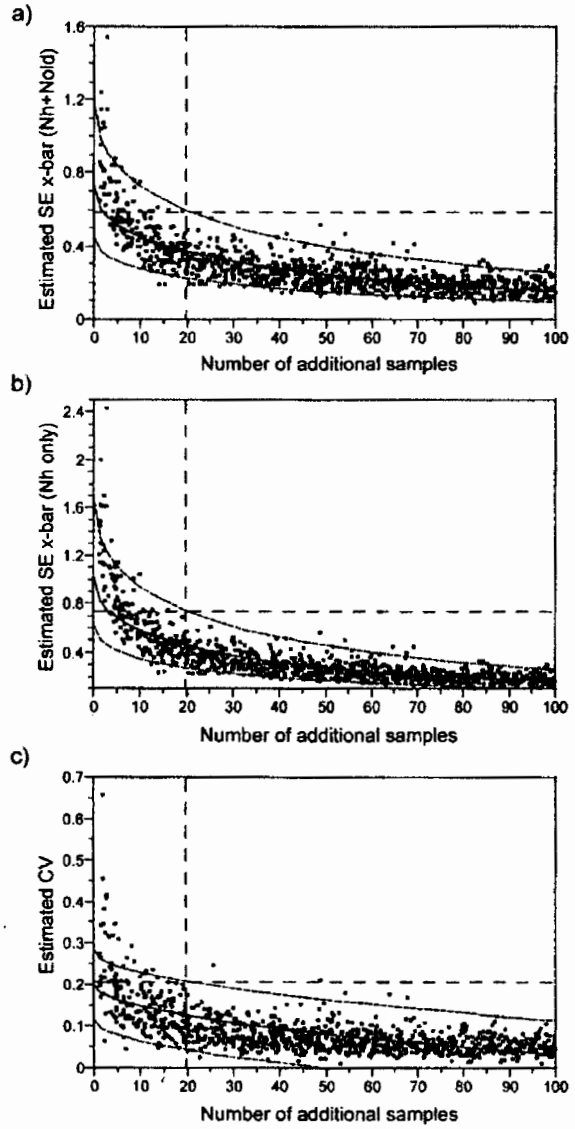

Figure 3. Plots showing the expected change in the standard error of the mean and of the CV (standard error divided by the mean) from 1000 Monte Carlo simulations. SE x-bar = standard error of the mean; Nh = number of additional samples ; Nold = number of existing samples. Curves indicate a transformed fit of the natural logarithm of the standard error versus the square root of the number of additional samples (solid line), and the 95% confidence limits for this curve (dashes). The intersection of the long, dashed lines indicate the estimated upper limit of the SE or CV at 20 additional samples.

lue, we also calculated the coefficient of variation (CV) as the ratio of the standard error divided by the mean (Figure 3c). For the example, a cursor line at 20 additional samples shows the expected upper range in the standard error and the CV. If the CV upper bound of 20% were acceptable, then 20 additional samples would be allocated based on the optimal sample allocations listed in Table 2. Other samples would also be planned to address uncertainties in the conceptual model or to evaluate the concentrations of other contaminants.

Estimating Contaminant Inventory

An example inventory calculation is provided for reach P-4 West in Table 3. This example uses one-dimensional Monte Carlo in Crystal Ball to evaluate uncertainty in plutonium239,240 inventory. Plutonium239,240 inventory and the volume of contaminated sediments were identified as result variables.

In the contaminant inventory calculation example, the variables considered to be uncertain included the thickness of the geomorphic units and the standard deviation of the contaminant concentration in that unit. Thickness distributions were discussed earlier relative to the sample allocation example. Considerations for the thickness assumptions for inventory calculations are basically the same as for sample allocation. The distributions selected for the average plutonium239,240 concentrations were based on available sample results for the geomorphic units. Sample results over several geomorphic units can be binned based on similarity of concentrations. These concentration bins typically reflect a similar time of deposition and contain similar ratios between indicator contaminants. An uncertainty multiplier was also used to account for the adequacy of the characterization (i.e., number of samples) for each binned geomorphic unit. The uncertainty multiplier was constrained to have an average value of 1 in all cases, but two types of distribution were used as uncertainty multipliers to reflect professional judgment in the characterization of those deposits. The specific distributions selected for the uncertainty multiplier had no theoretical basis, rather they were selected to represent uncertainty in characterization for well-sampled and poorly characterized geomorphic units. One uncertainty multiplier was a uniform distribution between 0.75 and 1.25, and this uncertainty multiplier was used for well-characterized bins. Another multiplier was used for more poorly characterized bins, and it was a discrete distribution that took on three values (0.5 at frequency 0.8, 1 at frequency 0.1, and 5 at frequency 0.1). The multiplier for the poorly characterized bins was constructed to have a skewed distribution, thus a small probability (10%) of a large multiplier (5). A wider range uniform

Table 3. Example inventory calculation for reach P-4 West illustrating the parameters assumed to be uncertain in the Monte Carlo simulations.

Geomorphic unit name	Geomorphic unit facies	Geomorphic unit area (m^2)	Estimated average thickness (m)	Thickness distribution	Estimated volume (m^3)	Estimated gravel fraction (%)	Estimated average density (g/cm^3)	Bin	Estimated average Pu-239,240 (pCi/g)	Estimated Pu-239,240 inventory (uCi)	Percent of total
c1	Coarse	1,850	0.45	N(0.45,0.2)	833	15	1.25	1	1.56	1,380	0.84%
c1b	Coarse	1,650	0.45	N(0.45,0.2)	744	5	1.25	1	1.56	1,380	0.84%
c2a	Coarse	274	0.45	N(0.45,0.2)	123	15	1.25	1	1.56	204	0.12%
c2b	Coarse	2,670	0.45	N(0.45,0.2)	1,200	15	1.25	1	1.56	1,990	1.21%
c2c	Coarse	3,180	0.45	N(0.45,0.2)	1,430	15	1.25	1	1.56	2,370	1.44%
c3	Coarse	276	0.45	N(0.45,0.2)	124	15	1.25	1	1.56	206	0.12%
c4a	Coarse	8,060	0.2	LN(0.2,0.1)	1,610	5	1.25	1	1.56	2,990	1.81%
			Bin 1 units c1,c2,c3 uncertainty multiplier						D(.5,.8;1,.1;5,.1)[a]		
c4a	Coarse	8,060	0.8	N(0.8,0.4)	6,450	5	1.25	1	1.56	12,000	7.25%
				Bin 1 unit c4a uncertainty multiplier1						D(.5,.8;1,.1;5,.1)[a]	
c4b	Coarse	3,790	0.2	LN(0.2,0.1)	758	5	1.25	2	8.4	7,590	4.60%
c5	Coarse	4,300	0.2	LN(0.2,0.1)	859	5	1.25	2	8.4	8,600	5.22%
f1	Coarse	12,900	0.01	EXP(0.01)	129	3	1.25	2	8.4	1,320	0.80%
f1a	Coarse	5740	0.01	EXP(0.01)	57	3	1.25	2	8.4	587	0.36%
				Bin 2 units c4b,c5, f1 uncertainty multiplier				U(0.75,1.25)			
c4b	Coarse	3,790	0.8	N(0.8,0.4)	3030	5	1.25	2	8.4	30,360	18.41%
c5	Coarse	4,300	0.8	N(0.8,0.4)	3440	5	1.25	2	8.4	34,400	20.86%
				Bin 2 units c4b,Cc uncertainty multiplier				1	D(.5,.8;1,.1;5,.1)[a]		
c6	Coarse	4,900	0.8	N(0.8,0.4)	3920	5	1.25	3	1.5	6,980	4.23%
				Bin 3 unit c6 uncertainty multiplier				U(0.75,1.25)			
c1	Fine	1,850	0.05	LN(0.05,0.025)	93	0	1.04	4	5.7	549	0.33%
c1b	Fine	1650	0.05	LN(0.05,0.025)	83	0	1.04	4	5.7	490	0.30%
c2a	Fine	274	0.05	LN(0.05,0.025)	14	0	1.04	4	5.7	81	0.05%
c2b	Fine	2670	0.05	LN(0.05,0.025)	134	0	1.04	4	5.7	792	0.48%
c2c	Fine	3180	0.05	LN(0.05,0.025)	159	0	1.04	4	5.7	941	0.57%
c3	Fine	276	0.05	LN(0.05,0.025)	14	0	1.04	4	5.7	82	0.05%
c4a	Fine	8060	0.05	LN(0.05,0.025)	403	0	1.04	4	5.7	2390	1.45%
				Bin 4 uncertainty multiplier				U(0.75,1.25)			
c4b	Fine	3,790	0.1	LN(0.1,0.05)	379	0	1.04	5	14	5,520	3.35%
c5	Fine	4300	0.1	LN(0.1,0.05)	430	0	1.04	5	14	6,255	3.79%
c6	Fine	4,900	0.2	LN(0.2,0.1)	979	0	1.04	5	14	14,260	8.65%
f1	Fine	12,900	0.05	LN(0.05,0.025)	643	3	1.04	5	14	9,083	5.51%
f1a	Fine	5740	0.15	N(0.15,0.05)	862	3	1.04	5	14	12,169	7.38%
				Bin 5 uncertainty multiplier				D(0.5,0.8;1,0.1;5,0.1)[a]			
				Total Volume	35000			Total Inventory in Reach P-4 West	165000	100%	

Pu-239,240 concentration bins: bin 1 is LN(1.56, 0.92); bin 2 is LN(8.43, 4.03); bin 3 is LN(1.5, 2.7); bin 4 is LN(5.7, 4.2); bin 5 is LN(14, 17).

[a]Discrete distributions are provided as a list: value, and the probability of that value; and so forth.

Note: Cells highlighted in black are the assumptions used in the Crystal Ball simulations, and the value in the cell is the expected or mean value of the distribution. The gray cells are the Crystal Ball result variables. The f2 geomorphic unit was excluded due to its small contribution to contaminant inventory.

distribution could also have been selected for the poorly characterized bins; a uniform distribution between 0 and 2 or U[0,2] is required to maintain an expected value of 1.

Reach P-4 West was one of the more intensively investigated reaches and provides a good example of how estimates of contaminant inventory changed after sampling events (Figure 4). As measured by the range of 20th and 80th percentile estimates of plutonium239,240 contaminant inventory, uncertainty in the plutonium239,240 inventory decreased as more samples were collected (Figure 4). Simple statistical sampling models also predict this decrease in uncertainty, but it is worth noting that decreases in uncertainty are not a necessary outcome of our methods. One scenario where uncertainty might increase is where initial field reconnaissance underestimates key inputs in contaminant inventory (like thickness of geomorphic deposits with higher concentrations). In this case, estimates of inventory and uncertainty in inventory might be initially low and increase after characterization better defines the fluvial system. However, once the system is better defined, additional characterization should better bound concentration, thickness, and area and ultimately reduce uncertainty in contaminant inventory after each investigation phase.

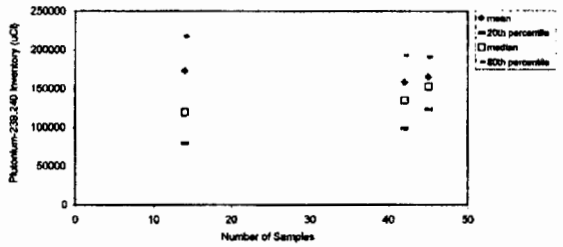

Figure 4. Estimates of plutonium-239,240 inventory in reach P-4 West based on samples collected in that reach versus the number of samples collected.

Comparison of Geomorphic Approach to Other Methods

Because some of these reaches have been sampled previously, it is possible to make empirical comparisons of the geomorphic approach to other methods. One example is reach ACS, adjacent to a primary contaminant source, that was characterized using either systematic sampling methods (FUSRAP or RFI) or biased sampling (NMED) or complete applications of the geomorphic sampling approach (this study). The primary goal of these studies was to obtain representative data on contaminant concentrations for use in risk assessments. Data from these investigations also span about two decades, beginning 13 years after effluent releases stopped, and the inventory and average concentrations of plutonium239,240 should have decreased during this period due to dilution associated with continued sediment transport (Reneau and others 2004). However, the opposite result is evident in average plutonium239,240 concentrations derived from different sampling approaches, and the earlier FUSRAP and RFI concentrations are one to three orders of magnitude lower than later NMED concentrations or concentrations from this study (Figure 5). This indicates that systematic sampling of sediments in fluvial systems (regularly spaced and/or predetermined locations) can lead to underestimates of contaminant concentrations. This bias is likely due to sampling an inappropriate spatial domain, such as only collecting samples from active channel deposits (RFI) or missing relatively small sediment deposits with the highest contaminant concentrations when sampling at predetermined locations (as was the case in FUSRAP sampling). Ladd and others (1998) reached a similar conclusion that random or systematic sampling of fluvial systems are inefficient or inappropriate when significant between-unit differences exist in a reach.

Comparisons of sampling based on the area encompassed by contaminated sediment deposits to sampling based on contaminated sediment volume are also instructive. These comparisons are assessed using the difference in average concentrations obtained from area and volume stratified sampling. These sampling scenarios were explored using two-dimensional Monte Carlo simulations to evaluate uncertainty in average contaminant concentrations. Data for cesium-137 in reach LA-2 East and plutonium-239,240 in reach P-4 are used as two examples. These simulations were performed using Crystal Ball, and the structure of the model and input values are provided in Tables 4 and 5. The estimated average concentrations of cesium-137 or plutonium-239,240 were assumptions or uncertain variables in the model. Area-weighted or volume-weighted concentrations were identified as result variables.

The assumptions used in these simulations were based on the sample results available for lumped geomorphic units (combining different subunits and different facies). The difference in cesium-137 concentrations between geomorphic units is provided in Figure 6, and the concentrations of cesium-137 in background samples are provided for comparison. Cesium-137 concentrations appear to be derived from a lognormal statistical distribution, which justifies using a lognormal distribution for cesium-137 concentrations. Plutonium-239,240 concentrations are provided in

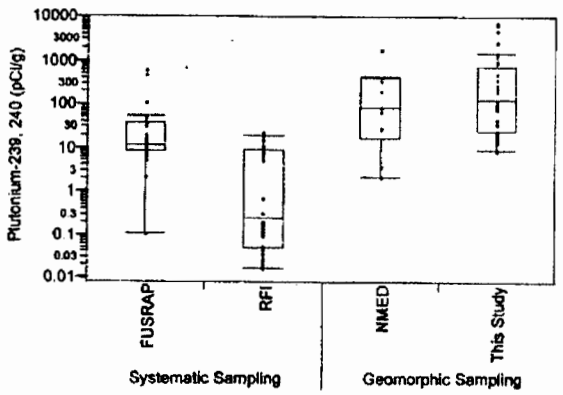

Figure 5. Plutonium-239,240 concentrations in reach ACS as measured in various investigations [systematic sampling under FUSRAP in 1977 and RFI in 1992–1993; ad hoc geomorphic approach by NMED in 1999; and geomorphic approach in 1999 (this study)]. Boxes indicate the interquartile range (25th to 75th percentile) and the lines above and below the boxes represent the 10th and 90th percentile.

Figure 7, and these concentrations also appear to be derived from a lognormal statistical distribution.

The area-weighted and volume-weighted estimates of the mean cesium-137 concentrations for reach LA-2 East differed by about 10% (Table 4). This result is not surprising as the point estimate (values calculated without uncertainty in the assumptions) for the area-weighted and volume-weighted mean concentrations differed by about 10% (Table 4). In contrast, the area-weightd and volume-weighted estimates for the mean plutonium-239,240 concentration in reach P-4 are quite similar, consistent with the smaller difference (about 3%) between the area-weighted and volume-weighted point estimates (Table 5). Area-weighted and volume-weighted calculations might come to similar results for different reasons. One reason is that sediment deposits in various geomorphic units in the study area have roughly the same thickness. Another reason is that although there are differences in the area and volume weightings, these differences cancel each other out in calculating the overall weighted mean. These comparisons suggest that accurate determination of the areal extent of contaminated flood deposits are important to obtaining representative average concentrations of contaminants. Although thickness of the various deposits is a factor, it is not as important a factor as area in these calculations. However, because determining the thickness of deposits is relatively inexpensive and is required for inventory calculations, we consider that collection of these data is worthwhile.

Table 4. Data used for Monte Carlo sampling simulations of cesium-137 concentrations in reach LA-2 East that were used to compare uncertainty in area-weighted and volume-weighted average concentrations

Lumped[a] geomorphic unit	Area (m^2)	Fraction area	Cumulative fraction area	Estimated average thickness (m)	Volume (m3)	Fraction volume	Cumulative fraction volume	Average concentration (pCi/g)	Concentration distribution	Area-weighted average (pCi/g)	Volume-weighted average (pCi/g)
c1	1330	0.20	0.20	0.5	664	0.14	0.14	2.35	LN(2.35, 0.84)	0.47	0.33
c2	3510	0.53	0.73	1	3510	0.73	0.87	15.0	LN(15.0, 10.6)	7.91	11.0
c3	1528	0.23	0.96	0.2	306	0.06	0.94	82.8	LN(8.28, 77.6)	3.71	5.18
f1	299	0.04	1.00	1	299	0.06	1.00	16.8	LN(16.8, 22.8)	3.84	1.07
Sums	6660	1.00			4780	1.00				15.9	17.6

[a]Geomorphic units with similar distributions of contaminant concentrations were lumped (binned) for the purpose of these simulations.

Note: Cells highlighted in black are the assumptions used in the Crystal Ball simulations and the gray cells are the Crystal Ball result variables.

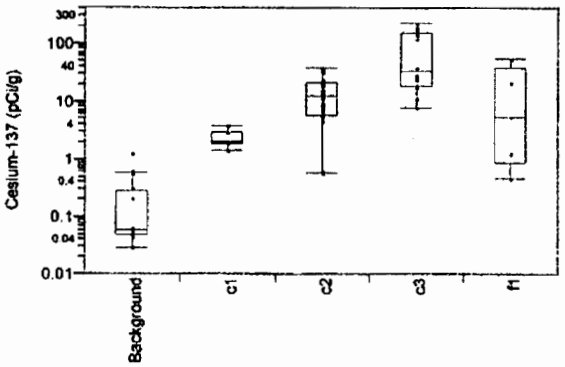

Figure 6. Box plot illustrating differences in cesium-137 concentrations from background samples (n = 24) and from samples collected in channel (c1, c2, c3) and floodplain (f1) sediment deposits in reach LA-2 East (n = 55). Boxes indicate the interquartile range (25th to 75th percentile) and the lines above and below the boxes represent the 10th and 90th percentile.

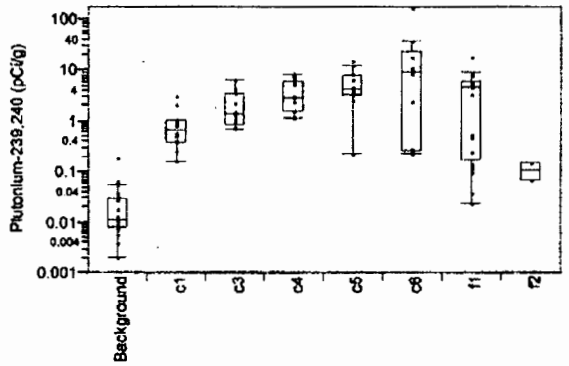

Figure 7. Box plot illustrating differences in plutonium-239,240 concentrations from background samples (n = 24) and from samples collected in channel (c1, c3, c4, c5, c6) and floodplain (f1, f2) sediment deposits in reach P-4 (n = 80). Boxes indicate the interquartile range (25th to 75th percentile) and the lines above and below the boxes represent the 10th and 90th percentile.

Figures 8 and 9 illustrate how uncertainty and bias in the estimate of the mean differs as more samples are collected to characterize a reach. In both cases, uncertainty in the mean (e.g., the range between 5th and 95th percentiles) decreases as more samples are collected in the reach. For reach LA-2 East, the area and volume-weighted sample allocations lead to decreased uncertainty and the general shapes of the distributions of the means are similar (Figure 8). Furthermore, relatively little bias occurs over a range of 10 to 80 samples, as the median estimate does not differ greatly from the point estimate values (Figure 8). Bias is more evident in the reach P-4 example, where the median estimate increases from 10 to 80 samples to a value approaching the point estimate (Figure 9). In the reach P-4 example, uncertainty is also less with the volume-weighted estimate than the area-weighted estimate, particularly for small sample numbers.

It is also instructive to compare statistics generated from individual samples with equal weights to more representative statistics based on area-weighted or volume-weighted calculations. For reach LA-2 East, the average of the 55 cesium-137 results is 30.8 pCi/g and the maximum is 230 pCi/g. In comparison, the point estimate for area weighting is 15.9 pCi/g, and for volume weighting, it is 17.6 pCi/g. For reach P-4, the average of the 80 plutonium-239,240 concentrations is 6.37 pCi/g and the maximum is 170 pCi/g, whereas the point estimate for area weighting is 4.49 pCi/g, and for volume weighting, it is 4.58 pCi/g. These comparisons illustrate that overestimates can occur if weighting associated with stratified sampling is ignored during subsequent compilation of summary statistics. Under some circumstances, underestimates could also occur. Therefore, it is important that biases introduced during sample collection be removed during data analysis.

Discussion

We have shown how geomorphic methods can be combined with simple statistical methods to produce more rigorous and representative characterization of contaminated sediment deposits in fluvial systems. Our example applications are for radionuclide releases in semiarid systems, but these methods can be applied in any environment or for any sediment-bound contaminants. If sampling ignores important components of the spatial domain or if more systematic approaches are taken to characterize such systems, we have shown that the bias can be significant, resulting in orders of magnitude underestimates in concentration.

The tools we have used, stratified random sampling formulas and simple Monte Carlo simulations, are simple to implement and provide a quantitative basis for adequate and appropriate sampling in fluvial systems. In a regulatory environment, it is important to demonstrate a thorough and objective approach for characterizing complex systems. The empirical foundation of the approach adopted in this study facilitates explanation of the methods to regulators and other interested parties.

Stratified random sampling methods easily complement a geomorphic approach when various sediment deposits or geomorphic units can be identified.

Table 5. Data used for Monte Carlo sampling simulations of plutonium-239,240 concentrations in reach P-4 that were used to compare uncertainty in area-weighted and volume weighted average concentrations.

Lumped[a] Geomorphic Unit	Area (m^2)	Fraction area	Cumulative fraction area	Estimated average thickness (m)	Volume (m^3)	Fraction volume	Cumulative fraction volume	Average concentration (pCi/g)	Concentration distribution	Area-weighted average (pCi/g)	Volume-weighted average (pCi/g)
c1	17,700	0.16	0.16	0.5	8,850	0.09	0.09	0.96	LN(0.96, 0.87)	0.152	0.091
c2	7,490	0.07	0.23	0.5	3,740	0.04	0.13	2.31	LN(2.31, 1.95)	0.155	0.092
c3	13,200	0.12	0.34	2.2	29,100	0.31	0.45	2.31	LN(2.31, 1.95)	0.273	0.718
c4	11,900	0.11	0.45	2.5	29,600	0.32	0.76	3.88	LN(3.88, 2.52)	0.411	1.23
c5	4,300	0.04	0.49	2.5	10,700	0.11	0.88	5.99	LN(5.99, 4.51)	0.230	0.688
c6	4,300	0.04	0.53	1	4,900	0.05	0.93	27.7	LN(27.7, 54.9)	1.21	1.45
f1	52,400	0.47	1.00	0.125	6,550	0.07	1.00	4.39	LN(4.39, 4.63)	2.06	0.308
Sums	112,000	1			93,500	1				4.49	4.58

Note: Cells highlighted in black are the assumptions used in the Crystal Ball simulations and the gray cells are the Crystal Ball results variables.
[a]Geomorphic units with similar distributions of contaminant concentrations were lumped (binned) for the purpose of these simulations.

The geomorphic methods work best when multiple indicators can be used to identify various sediment deposits. For example, simple field measures like particle size or floodplain height (often reflecting relative deposit age) can be used to initially subdivide an area into geomorphic units with potentially varying contaminant concentrations. These initial assignments can be revised through more detailed characterization of contaminant signatures with field screening (e.g., field radiation measurements) or using analytical data from initial phases of sampling. Following reconnaissance investigations with more detailed geomorphically based sampling has also been advocated by Ladd and others (1998) and Rhoads and Cahill (1999) in different settings. Field screening was an effective tool for sediment deposits in Los Alamos Canyon downstream of the source of cesium-137 contamination (Reneau and others 1998b; Katzman and others 1999; LANL 2004) and of plutonium-239,240 close to the source in Acid Canyon (Reneau and others 2000; LANL 2004), but field screening was not an effective tool in most of Pueblo Canyon due to the low concentrations of plutonium-239,240 relative to the sensitivity of gross alpha radiation field measurements. However, in Pueblo Canyon, the identification of channel changes through examination of historical aerial photographs were useful to define geomorphic units of varying age and guide sampling (Reneau and others 1998a, 2004). In a nearby canyon, X-ray fluorescence (XRF) analyses of sediment samples in a field trailer have been successful at identifying variations in concentration of a metal (barium) in sediment deposits, also allowing reliable subdivision of geomorphic units prior to designing a sampling strategy (LANL 2003). In summary, this general approach has been successfully applied in a series of field sites with varying types and levels of contaminant, indicating that it has potential for broader application as well.

The quantitative methods can be modified to use two-dimensional Monte Carlo simulations to model variability and incertitude as separate components of contaminant inventory uncertainty. Bayesian methods can also be used as a more formal method to develop distributions for uncertain variables based on measurements and professional judgment. Even without these enhancements to the methods, the approach has an intuitive appeal to regulators because it is a data-centered approach. The emphasis on the data also lends toward establishing and testing various assumptions. As applied in the phased characterization approach of contaminated sediments in Los Alamos and Pueblo canyons, sampling objectives were identified after each characterization phase. These objectives and

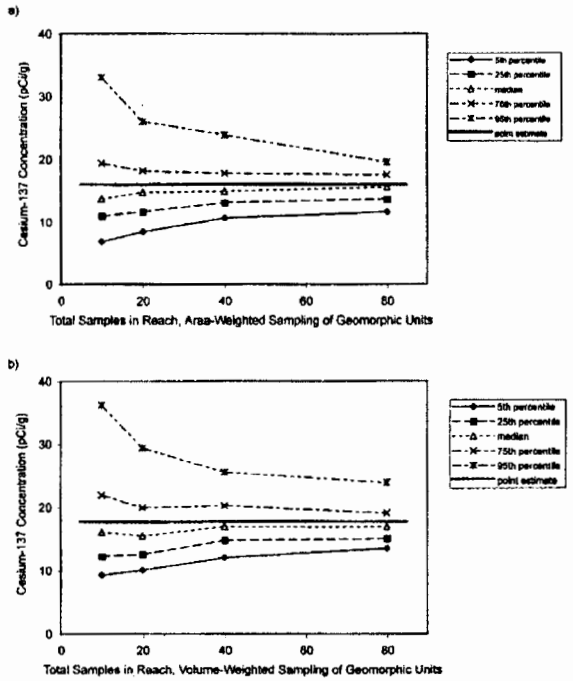

Figure 8. Estimates of the average cesium-137 concentrations in reach LA-2 East based on simulations of samples allocated by area-weighted (a) and volume- weighted (b) sampling. The point estimate is the result calculated without uncertainty and the percentiles depict the results of the Monte Carlo simulations.

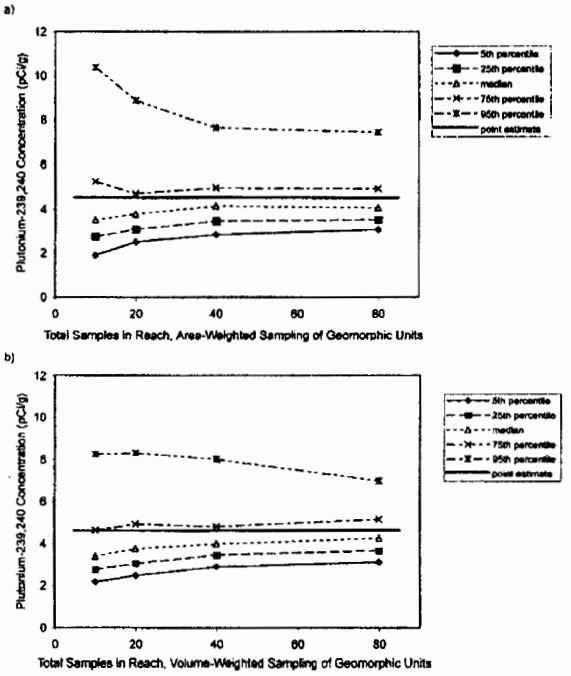

Figure 9. Estimates of the average plutonium-239,240 concentrations in reach P-4 based on simulations of samples allocated by area-weighted (a) and volume-weighted (b) sampling. The point estimate is the result calculated without uncertainty and the percentiles depict the results of the Monte Carlo simulations.

the intermediate results can be communicated with regulators and other interested parties leading to greater acceptance and understanding of the ultimate characterization results.

Acknowledgments

Funding was provided by the US Department of Energy through the Los Alamos National Laboratory Environmental Restoration Project and the Risk Reduction and Environmental Stewardship Remediation Services Project. We thank David Broxton, Julie Canepa, Jorg Jansen, Dave McInroy, Allyn Pratt, and Kent Rich for technical, financial, and administrative support during various phases of this project, and David Broxton, Andrew Marcus, Sara Rathburn, Wendy Swanson, and an anonymous reviewer for their comments on this manuscript. The lead author was funded under subcontract S80002 to Los Alamos Technical Associates (Prime Contract N0. G66160008-8M). This article is report LA-UR-04-1370 of the Los Alamos National Laboratory.

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