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APPLICATION OF LANL BACKGROUND DATA TO ER PROJECT DECISION-MAKING PART II: RADIONUCLIDES IN SOILS, SEDIMENTS, AND TUFF

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Los Alamos National Laboratory
Environmental Restoration Decision Support Council
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REVIEW DRAFT

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INTRODUCTION

The purpose of this policy is to provide guidance to Los Alamos National Laboratory (LANL, the Laboratory) Environmental Restoration (ER) Project personnel on the ER Project's approach to conducting background comparisons for radionuclides. This document is a supplement to EM/ER:96-PCT-010, *Application of LANL Background to ER Project Decision-Making, Part I: Inorganics in Soils, Sediments and Tuff* (Ryti et al. 1996, 1298), which presents the background comparison approach for inorganic chemicals.

This document discusses the three ways in which the consideration of radionuclide background is unique from that of inorganics. First, the spatial distribution and concentration of radionuclides is derived from natural background sources (primordial and cosmogenic), globally elevated concentrations from atomic weapons test fallout, regionally elevated radioactivity from past Laboratory operations, and PRS-specific releases of radionuclides. Both natural radionuclide sources and fallout-related activity are considered to represent background radioactivity. Locally elevated values from Laboratory operations represent baseline radioactivity. Ideally, a PRS-specific release would be identified by comparing PRS radionuclide concentrations to baseline concentrations. Second, the standard practice for reporting radiological analyses is to report all results in the analysis library regardless of the analytical detection limit. Third, the Department of Energy (DOE) regulatory guidance (DOE 1990, 0080; DOE 1993, 1315; EPA 1994, 1316) for establishing cleanup levels for radionuclides is based on a dose above background.

This policy provides the decision logic for selecting background data sets for radionuclides. Making comparisons to radionuclide background provides field units with a mechanism for determining if a release has occurred, for deriving cleanup levels, and for evaluating the attainment of cleanup goals. Existing radionuclide background data include the Laboratory's annual Environmental Surveillance Reports (Fresquez et al. 1996, 1360), isotopic activities estimated from total element abundance measured in rock samples (Longmire et al. 1995, 1266), and background samples of canyons sediments (Ryti et al. 1997, draft).

This policy will be updated as issues discussed in the Issues and Caveats section (below) are resolved. Background data sets for comparisons to radiological screening data and to water data are under development.

Caveat: Comparability of Analytical Methods

It is important to identify appropriate analytical methods before data collection. This issue is an even greater pitfall for radionuclide analyses than for inorganic analyses, because radiochemical

analyses are not as standardized as inorganic analyses. For example, americium-241 may be analyzed by either gamma spectroscopy or alpha spectrometry with multiple options for sample preparation, counting geometries, and counting times possible for each analytical method. Analytical methods for radium, thorium and uranium isotopes also warrant careful review to ensure comparability with the relevant set of background data.

Caveat: Use of Radioactivity Estimated from Total Abundances

There are several data preparation issues relating to estimation of naturally-occurring radionuclide activity from total abundances. These issues include: 1) constants used in the calculations (isotopic abundance, specific activity), 2) secular equilibrium - or how far down the decay chain can radioactivity be estimated (disruption of secular equilibrium at radon daughters or the special case of depleted uranium), 3) guidance for assembling radionuclide data for screening or other uses (converting totals to isotopic, mixed data sets with some totals and some isotopic). This issue will be considered in other documents, which are currently under development by the Decision Support Council.

Programmatic Issue: Collection of Additional Samples for the Radionuclide Background/Baseline Data Set

This issue needs the attention of the ER Project Office for resolution based on review of existing background data. This document will be updated once this issue is resolved.

The Decision Support and Earth Science Councils recommend that several background soil samples be collected from selected ER Project background sampling locations and analyzed for selected radionuclides. These additional data are needed to supplement and validate the current radionuclide background and baseline data, and to provide radionuclides analyses for several of the natural-occurring series in soil and Bandelier tuff. The background soil sites sampled for radionuclides should include both poorly-developed soils (e.g., Tsankowi site and upper Los Alamos Canyon site) and well-developed soils (e.g., TA-39 site and TA-69 site). Soil samples should be collected from the major soil horizons at each site. These samples should be analyzed for the following radionuclides: americium-241, cesium-137, cobalt-60, plutonium-238, plutonium-239/240, radium-226, strontium-90, thorium-227, thorium-232, thorium-234, tritium, uranium-234, uranium-235, and uranium-238.

USING RADIONUCLIDE BACKGROUND FOR DECISION-MAKING

The most common use of a background data set is to determine whether a release from a specific PRS has occurred. Generally, ER Project personnel perform an upper tolerance limit [UTL]

comparison, a hot measurement comparison, or apply statistical tests to determine if the PRS-specific distribution of a radionuclide is significantly elevated relative to the background distribution. Readers interested in more information on choice and application of methods for statistical testing are referred to EM/ER:96-PCT-010, *Application of LANL Background to ER Project Decision-Making, Part I: Inorganic in Soils, Sediments and Tuff* (Ryti et al. 1996, 1298).

A second use of background data is to determine the portion of dose attributable to background. This evaluation may involve a correction (such as subtracting the mean radioactivity of the site data from the mean radioactivity of the background data) or a comparison (such as comparing the dose resulting from site radioactivity with dose resulting from background radioactivity). Each of these methods can lead to different recommendations for future actions at a site. Regulatory guidance for determining radionuclide dose limits are provided in DOE and EPA documents (DOE 1993, 1315; EPA 1994, 1316). Readers interested in more details are also referred to EM/ER:96-PCT-010, *Guidance for the Derivation and Use of Mesa-top Radionuclides Soil Cleanup Guidelines* (Perona 1997, draft).

Another use of radionuclide background data is for making certain types of waste classification decisions. In particular, low-level radioactive waste is defined as material containing added radioactivity from DOE operations. Added radioactivity would take into consideration natural background levels of radionuclides. Readers interested in more information on radioactive waste classification decisions are referred to Radioactive Waste Management Procedure for ER Project Field Operations SOP 1.11 (LANL, 0875).

SELECTION OF RADIONUCLIDE BACKGROUND DATA

General considerations

Certain radionuclides in the PRS-specific data set are extraneous and should be eliminated before an appropriate background data set can be selected for comparison. These radionuclides include analytical laboratory quality control indicators and radionuclides for which all reported activities are less than minimum detectable activity.

Radiological data packages returned by analytical laboratories typically contain indicators designated to ensure the quality control and quality assurance of the data package. These indicators do not represent suspected site contaminants, and thus warrant no further assessment relative to background.

The measured activity of the radionuclide must be greater than the minimum detectable activity for the sample. Radionuclides for which all reported activities are less than the minimum detectable

activity do not warrant further evaluation as potential site contaminants. Frequently, the counting time and the presence of other radionuclides can cause an estimated result for a radionuclide to be less than the sample and radionuclide-specific minimum detectable activity. Thus, the counting time should be reviewed to make sure that the detectable quantity was not arbitrarily inflated due to inadequate counting time. In addition, the spectral quantification windows should be reviewed for possible radionuclide interference.

Selection of an appropriate background data set is based on the type of media from which a sample was collected and whether the sample was collected from the surface or the subsurface. Naturally-occurring radionuclides are distributed at different concentrations in various soil and rock matrices. Thus, as for inorganic chemicals, the type of media from which a sample is collected (tuff versus all other solid media) is an important factor in determining an appropriate background data set. Background concentrations of radionuclides resulting from fallout are typically associated at surface or near-surface depths. Thus, background data for radionuclides resulting from fallout apply to samples collected from the surface (or the near-surface) only.

Site-Specific Background Data

When PRS-specific radionuclide background data are available, the approach described in the Decision Process section below may not be applicable. To date, the ER Project has collected sets of site-specific radionuclide background data. Certain of these are summarized in Table 1. These site-specific background data have been collected to support characterization of geologic sediments in Laboratory canyons, local background conditions in the central or "process" area of TA-21, environmental surveillance data used to calculate dose from laboratory operations to members of the public (see Campbell 1997, draft), and site-specific background data collected at several technical areas (TA-15, TA-18, TA-33, TA-49, and TA-50 [for MCA C]). To determine the adequacy of site-specific background for decision-making, the analytical suite used should be examined to insure that all potential radionuclide contaminants have been included. In addition, the number of samples taken should be evaluated to insure that site-specific background conditions have been adequately represented. The environmental surveillance off-Laboratory and canyons sediment data sets (see Table 1) will be used to represent Laboratory baseline or background conditions.

TABLE 1
Summary of Site-Specific Radionuclide Background Data

Description	Locations sampled	Radionuclide suite	Reference	Comments
Sediment background	Four canyons 28 locations Indio: 2 Ancho: 2 Guaje: 7 Los Alamos: 7 Pueblo: 10	Isotopic uranium, Isotopic plutonium, Gamma spectroscopy, tritium	Ryti et al. (1997, draft)	Used as a surrogate data set for soils
TA-21 Baseline	43 location in the TA-21 "process" area	Isotopic uranium, Total uranium, Isotopic plutonium, Isotopic thorium, tritium, americium-241, strontium-90	Ryti (1996 draft)	Used for evaluations of PRSs in the TA-21 process area.
TA-15 RFI	2 locations	Gamma spectroscopy	Personnel communication from K. Black	Not used
TA-18 RFI	3 location, 4 depths in 5 ft intervals	Isotopic plutonium, Isotopic thorium	TA-18 RFI Report (Environmental Restoration Project 1995, 1283)	Not used
TA-33 RFI	48 locations	Isotopic plutonium, cesium-137, tritium	TA-33 RFI Report (LANL 1995, 1212)	Used as site-specific background in RFI reports
TA-49 RFI	9 locations	Isotopic uranium, Isotopic plutonium, americium-241, cesium-137, tritium, potassium-40	Personnel communication from K. Lockhart	Not used
MDA C	5 locations	Isotopic uranium, Isotopic plutonium, americium-241, cobalt-60, potassium-40, radium-226, thorium-232	TA 50 RFI Report (Environmental Restoration Project, 1993, 1284)	Not used
Environmental Surveillance Laboratory locations	9 locations	Isotopic plutonium, americium-241, cesium-137, strontium-90, tritium	Campbell (1997, draft)	Used by the Environmental Surveillance Group for annual reports of dose to the public
Environmental Surveillance Off-Laboratory locations	14 locations	Isotopic plutonium, americium-241, cesium-137, strontium-90, tritium	Campbell (1997, draft)	Used for laboratory background conditions

Decision process

The process for eliminating extraneous PRS data and for selecting the most appropriate set of radionuclide background data is summarized in Figure 1. Before using the decision process shown in Figure 1 (and discussed below), it is essential that the sample collection, preparation and analytical methods used for PRS samples are comparable to those used for the background samples.

Decision 1. Is the radionuclide a radiological indicator?

"Yes" Decision. Radiological indicators are used for quality control and quality assurance evaluation of analytical laboratory data packages. Thus it is not appropriate to compare radiological indicators to background, dose- or risk-based health protection standards. Radiological indicators used for QC evaluations by the ER Project include: annihilation radiation, cadmium-109, cerium-

139, mercury-203, potassium-40, protactinium-231, protactinium-234m, tin-113, strontium-85, and yttrium-85. It should be noted that if potassium-40 is not included as a radionuclide indicator, it should be treated in the same manner as naturally-occurring radionuclides (i.e., it must be compared to background concentrations).

"No" Decision. The data for the remaining radionuclides warrant further assessment. Proceed to decision 2.

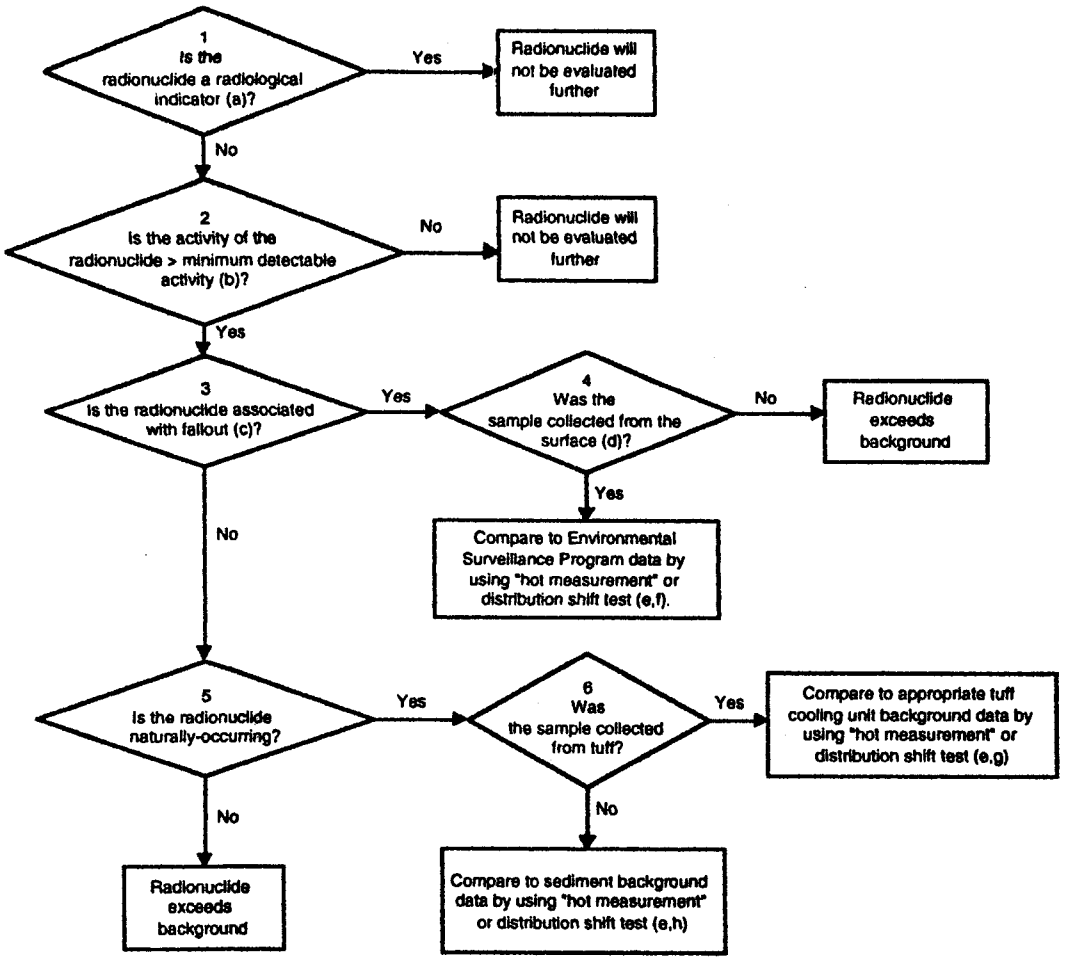
Decision 2. Is the activity of the radionuclide greater than the minimum detectable activity?

"Yes" Decision. The radionuclide requires further evaluation based on the activity reported by the analytical laboratory. Proceed to decision 3.

"No" Decision. None of the results reported for the radionuclide are greater than the minimum detectable activity value. In most cases, this indicates that an insignificant quantity of the radionuclide is present in the PRS-specific data and no further assessment of the radionuclide is necessary. However, before eliminating the radionuclide, the analytical data report should be reviewed to ensure that inadequate counting time and interferences did not lead to erroneous elimination of the radionuclide.

In cases for which the analytical laboratory does not report a minimum detectable activity for the sample, but does report an analytical uncertainty estimate[1], a value of three-times the analytical

[1] The analytical uncertainty is also known as the standard deviation (SD) of the analytical result. In some cases, the value reported in the FIMAD analytical uncertainty field is actually 2•SD or 3•SD. Thus, multiplying the result in the analytical uncertainty field by three to estimate a sample-specific minimum detectable activity could result in an inflated minimum detectable activity value.



(a) Radiological indicators include annihilation radiation, potassium-40, cadmium-109, cerium-139, mercury-203, tin-113 and strontium-85.
(b) The minimum detectable activity value should be reviewed to ensure that adequate counting time and interferences did not cause inappropriate elimination of radionuclides.
(c) Fallout radionuclides include: tritium, cesium-137, americium-241, plutonium-238, plutonium-239/240, and strontium-90.
(d) Surface samples are defined as intervals starting at 0 inch depth and extending no deeper than 6 inches.
(e) Choice and application of methods for statistical testing is presented in "Application of Background to ER Project Decision-Making, Part I".
(f) Use background screening values in Table 2 for hot measurement test.
(g) Use background screening values in Table 3 for hot measurement test.
(h) Use background screening values in Table 4 for hot measurement test.

Figure 1. Flow chart for selecting radionuclide background data sets.

uncertainty may be used as an estimate of the sample-specific minimum detectable activity. Using the minimum detectable activity as a data screening tool is valid only when the data package from the analytical laboratory meets the general guidelines available in the Statement of Work for radiochemical analyses (LANL 1995, 1278). Using different criteria for determining whether a radionuclide has detection status may compromise the comparability of the data sets. In some cases, discrepancies in calculating the certainty range of an analytical value for site and background data sets may compromise the utility of the background data set.

Decision 3. Is the radionuclide associated with fallout?

"Yes" Decision. Radionuclides resulting from fallout are expected to be associated with surface or near surface samples only. At the Laboratory, radionuclides resulting from fallout include: tritium, cesium-137, americium-241, plutonium-238, plutonium-239/240, and strontium-90. Proceed to decision 4.

"No" Decision. Proceed to decision 5 for evaluation of other classes of radionuclides.

Decision 4. Was the sample collected from the surface?

"Yes" Decision. Compare PRS data to data associated with Laboratory operations and global fallout. These data are derived from radionuclide samples collected by the ESH Division Environmental Surveillance Program (ESP) (Fresquez et al. 1996, 1360).

Because radionuclides resulting from fallout are associated with atmospheric deposition, the background activity of this class of radionuclide is limited to surface samples at relatively undisturbed sites. In this context, surface samples are defined as intervals starting at a 0 inch depth and extending no deeper than 6 inches.

Selecting appropriate statistical testing methods is described in Application of Background to ER Project Decision-Making, Part I. When the hot measurement comparison is appropriate (i.e., the background comparison is being used to support a screening assessment), compare PRS data to the fallout background values summarized in Table 2[2]. For other statistical tests, the background data values presented in Campbell (1997, draft) (Local Background Data Sets for Fallout Radionuclides Based on Historical Environmental Surveillance Data) should be used[3]. (Readers

[2] Note that tritium is reported in units representing activity in soil moisture, which should be compared to PRS data in equivalent units or PRS data converted to pCi/ml.

[3] Readers may contact K. Campbell (667-2799) to receive an electronic copy of ESP data to be used for statistical distribution shift tests.

interested in more information on sample locations, analytical methods, and statistical analysis of ESP data are referred to Campbell (1997, draft))

TABLE 2
Summary of ESP Data Screening Values for Radionuclides Resulting from Fallout

Radionuclide	Sample Preparation and Analytical Method	ESP Screening Value
H-3	Distillation and liquid scintillation counting	0.76 pCi/ml
Sr-90	Sample extraction and Beta counting by gas-flow proportional counter	1.31 pCi/g
Cs-137[3]	Sampled milled and gamma spectroscopy	1.65 pCi/g
Pu-238	Ion exchange and alpha spectroscopy	0.023 pCi/g
Pu-239/240	Ion exchange and alpha spectroscopy	0.054 pCi/g
Am-241	Ion exchange and alpha spectroscopy	0.013 pCi/g

"No" Decision. Once this point is reached, it is likely that the radionuclide exceeds background and the radionuclide should be carried forward to a dose- or risk-based assessment.

Decision 5. Is the radionuclide naturally-occurring?

"Yes" Decision. In the context of selecting radionuclide background sets, naturally-occurring radionuclides are limited to uranium, uranium decay-chain daughters, thorium, and thorium decay-chain daughters. Proceed to decision 6 for further evaluation of naturally-occurring radionuclides.

"No" Decision. Once this point is reached, it is likely that the radionuclide exceeds background, and the radionuclide should be carried forward to a dose- or risk-based assessment.

Decision 6. Was the sample collected from tuff?

"Yes" Decision. Compare PRS data to radionuclide background data associated with geologic unit. The source of these data is total abundance measured in rock samples (Longmire et al. 1995, 1266). The total abundance is converted to isotopic activity through isotopic abundances of uranium isotopes and the specific activity of these isotopes. Thus, the geologic unit

radionuclide background data represent a surrogate data set for radioactivity of naturally-occurring isotopes.

When PRS data are compared to background data by geologic unit, special attention should be paid to the analytical methods used for both the PRS data and the estimated background data. For example, the estimated background radioactivity of uranium-235 is less than the typical minimum detectable activity for this isotope when it has been analyzed by gamma spectroscopy.

Although there are potential problems in applying these estimated background values to naturally-occurring radionuclides, the geologic unit background data are based on the natural variation of uranium and thorium present in various tuff cooling units. Thus, these data will assist in correctly interpreting results from boreholes that intersect multiple tuff cooling units.

Selecting appropriate statistical testing methods is described in *Application of Background to ER Project Decision-Making, Part I.* When the hot measurement comparison is appropriate (i.e., the background comparison is being used to support a screening assessment), the background screening values summarized in Table 3 should be used[4]. For other statistical tests, background data values for tuff are available[5]. Readers interested in more information on sample locations and analytical methods are referred to *Natural Background Geochemistry and Statistical Analysis of Selected Soil Profiles, Sediments, and Bandelier Tuff* (Longmire et al. 1995, 1266).

TABLE 3
Summary of Measured and Calculated Radionuclide Background Screening Values by Geological Unit

Analyte	Qbt 4	Qbt 3	Qbt 2	Qbt 1v	Qbt 1g	Qbo
Radium-226[1] (pCi/g)	6.23	2.6	4.82	6.61	6.78	7.15
Total[2] Thorium (mg/kg)	16.5	16.3	25.9	30.1	37.1	19.9
Thorium-232 (pCi/g)	1.80	1.78	2.83	3.29	4.05	2.17
Total[2] Uranium (mg/kg)	4.55	4.37	7.12	7.59	10.1	5.61
Uranium-234 (pCi/g)	1.61	1.55	2.53	2.70	3.59	1.99
Uranium-235 (pCi/g)	0.070	0.067	0.109	0.116	0.155	0.086
Uranium-238 (pCi/g)	1.52	1.46	2.37	2.53	3.37	1.87

1. Directly measured by gamma spectroscopy
2. Total refers to the whole rock result, measured by Instrumental Neutron Activation Analysis, and this value was used to estimate the abundance of primary isotopes thorium and uranium.

[4] Table 3 includes measured activity for radium-226 in addition to estimated activity for thorium and uranium isotopes.

[5] Data are available from FIMAD or the readers may contact Randall Ryti (662-0707, ext. 12) to receive an electronic copy of the tuff data to be used for statistical distribution shift tests.

"No" Decision. For solid media other than tuff (soil, fill and sediments), compare PRS data to background data associated with sediments. Because abundances of naturally-occurring uranium, thorium and their daughters are expected to be similar in sediment, soil, and fill material, the background data collected for naturally-occurring radionuclides in canyon sediment (Ryti et al. 1997, draft) are viewed to represent a conservative background data set for comparing all these media. The use of the canyons sediment data as a surrogate for soil and fill is considered preferable to using estimated isotopic activities from total uranium and thorium analyses for mesa top soil background sampling locations.

Selecting appropriate statistical testing methods is described in *Application of Background to ER Project Decision-Making, Part I.* When the hot measurement comparison is appropriate (i.e., the background comparison is being used to support a screening assessment), the background screening values summarized in Table 4 should be used. For other statistical tests, the background data values for tuff are available[6]. Readers interested in more information on sample locations and analytical methods are referred to *Natural Background Geochemistry of Sediments, Los Alamos National Laboratory* (Ryti et al. 1997, draft)

TABLE 4
Summary of Background Screening Values for Naturally-Occurring Radionuclides Detected in Sediments

Radionuclide	Analytical Method	Background Screening Value (pCi/g)
AC-228	Gamma Spectroscopy	3.30
BI-214	Gamma Spectroscopy	1.61
PB-212 (1)	Gamma Spectroscopy	2.36
PB-214 (1)	Gamma Spectroscopy	1.97
RA-226 (1)	Gamma Spectroscopy	2.03
TH-228 (1)	Alpha Spectroscopy	2.28
TH-230 (1)	Alpha Spectroscopy	2.29
TH-232 (1)	Alpha Spectroscopy	2.33
TL-208 (1)	Gamma Spectroscopy	0.906
U-234 (2)	ICPMS	2.39
U-235 (2)	ICPMS	0.16
U-238 (2)	ICPMS	2.29

ICPMS = Inductively Coupled Plasma Mass Spectrometry

[6] Data are available from FIMAD or the readers may contact Randall Ryti (662-0707, ext. 12) to receive an electronic copy of the sediment data to be used for statistical distribution shift tests.

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