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BASELINE DATA FOR FALLOUT RADIONUCLIDES AT LANL

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1.0 INTRODUCTION

The Environmental Restoration (ER) Project guidance document *Application of LANL Background Data to ER Project Decision Making, Part II: Radionuclides in Soils, Sediments and Tuff* (LANL ER Decision Support Council and Earth Science Council, 1997) includes background upper tolerance limits (UTLs) for six fallout radionuclides. These UTLs are based on data collected by the Environmental Surveillance Program (ESP) of the Los Alamos National Laboratory's (hereafter referred to as the Laboratory or LANL). The purpose of this supporting paper is to provide a detailed discussion of the ESP data and of the criteria by which subsets of these data were selected as appropriate baseline data sets for use by the ER Project. Appendix A includes tables of the selected subsets. The statistical details of the calculation of the UTLs and of other summary statistics bases on these data sets is provided in Appendix B. These data sets can also be used for other statistical comparisons with site data; see Ryti et al. (1996) for a discussion of two-sample statistical tests that may be used to compare site data with background or baseline data.

2.0 ENVIRONMENTAL SURVEILLANCE DATA

2.1 Background vs. Baseline Data

A number of relatively long-lived radionuclides are present at low but measurable levels of activity in soils world-wide as a result of atmospheric testing prior to the 1963 Atmospheric Test Ban Treaty. These fallout isotopes include those listed in Table 1, in addition to uranium. The ESP has sampled extensively for these isotopes in soil, sediment, water and biota on and around the Laboratory. Other radionuclides from atmospheric testing have much shorter half-lives and are no longer present at detectable levels (i.e., above the minimum detectable activity or MDA) in the environment. This anthropogenic background would affect surficial media on the Pajarito Plateau even if the Laboratory were not here.

Uranium is also present on the Pajarito plateau as a constituent of its rocks and soils, i.e., as part of the natural background that would exist even if humans did not. Uranium is not discussed in this

Table 1. Fallout Radionuclides

Isotope	Half-life*	MDA**
Tritium (^{3}H)	12.33 years	0.5 pCi/mL
Strontium-90 (^{90}Sr)	28.78 years	1 pCi/g
Cesium-137 (^{137}Cs)	30.07 years	0.06 pCi/g
Plutonium-238 (^{238}Pu)	87.7 years	0.002 pCi/g
Plutonium-239/240 ($^{239+240}Pu$)	24110 years	0.002 pCi/g
Americium-241 (^{241}Am)	432.2 years	0.002 pCi/g

* Half-lives from the National Nuclear Data Center, http://www.nndc.bnl.gov/nndc/nudat/

** MDAs from LANL Method documentation, see Appendix A for discussion

paper, but the guidance document mentioned above presents background UTLs for both total and isotopic uranium.

Operational sources of the radionuclides listed in Table 1 also exist at LANL. In particular, there are some stacks on Laboratory property that may have released small quantities of radionuclides to the atmosphere. While not associated with the Potential Release Site (PRS) being investigated by the ER Project, these releases may lead to slight increases in activity in surface soils at distances of hundreds or thousands of feet from the sources, including soils at ER PRSs.

Baseline activity levels for the fallout radionuclides in surficial materials include both contributions from natural and anthropogenic background, as defined above, and from Laboratory-wide operational releases. These are the levels to which it is appropriate to compare data from ER sites on or near the Laboratory. It may be difficult or impossible to determine what fraction of this baseline activity is due to general Laboratory operations, but this particular piece of information is not needed in order to develop a defensible baseline data set for use by the ER Project.

2.2 Spatial and temporal variability

The distributions of the fallout isotopes exhibit both spatial and temporal trends. (See Fresquez et al., 1996, and the references therein.) Spatially, activity patterns are controlled by a number of factors, including wind patterns, elevation, and precipitation In the case of the Pajarito Plateau, these factors would tend to lead to higher activities of fallout isotopes on the mesa tops where the Laboratory is located compared to the surrounding lower, dryer areas, even in the absence of any contribution from Laboratory operations. It is factors such as these that make it difficult to separate the contribution of Laboratory operations from the world-wide effects of atmospheric testing and other global anthropogenic sources

In addition, activity of fallout radionuclides decreases rapidly with soil depth in undisturbed soils, including those sampled by the ESP. However, many of the sites being investigated by the ER Project are disturbed or back-filled, which may result in the redistribution of fallout radionuclides to greater depths

Temporally, activity due to fallout from atmospheric testing of the shorter-lived isotopes (those with half-lives of 30 years or less) has decreased measurably since the end of atmospheric testing in the early 1960s. Weathering (wind, water erosion, and leaching) may also lead to decreased measured activity at many sites. Other world-wide anthropogenic sources such as satellite reentry could lead to a slight increase in the environmental activity of some isotopes over time Temporal patterns associated with activities at the Laboratory depend on the level of operations and the sophistication of engineering controls Overall trends in both of these areas would be expected to lead in the direction of decreasing levels of measured activity, especially for the shorter-lived isotopes.

All of these factors must be considered in proposing baseline data sets for comparison with PRS-specific data collected by the ER Project. Specifically

- because of temporal trends (which in fact are observed not only for the shorter-lived isotopes but even for some long-lived isotopes, as discussed in Section 4), only recent data are included in the baseline data sets, with which ER data collected in the 1990s will be compared, and

- because of spatial trends, a baseline data set that is based on ESP stations located near the perimeter of the Laboratory may not adequately reflect conditions prevailing in the center of the Laboratory. This issue is further discussed in Section 3 3 and in the appropriate subsections of Section 4

- because of trends with depth, prior disturbance at a sampling location must be evaluated when comparing site data to baseline data presented in this report. In general, it is not appropriate to compare results for subsurface samples to the baseline data.

2.3 ESP Sampling Locations

The Environmental Surveillance Program of the Laboratory has collected data on the activity of the isotopes listed in Table 1, as well as total uranium and gross alpha, beta and gamma activity, in surface soils within the Laboratory and surrounding areas since 1974. These data are published in the Laboratory's annual Environmental Surveillance reports. The data for soil samples through 1994 have also been summarized by Fresquez et al. (1995) and are presented in full in Appendix A of Fresquez et al. (1996). 1995 data are presented in the Environmental Surveillance Program report for 1995. Discussions of temporal and spatial trends in these data is provided in Section 4, with a view to selecting appropriate subsets of the historical ESP data that are appropriate for use as baseline data for ER Project sites.

Twenty-three soil sampling stations, listed in Table 2, have been sampled by the ESP almost annually since 1978. Seven of these stations are classified by the ESP as Regional stations, i.e., probably too far from the Laboratory to be affected by Laboratory operations. Six of the Regional stations are more than 15 miles from the Laboratory boundary, while the seventh, Otowi, is less than five miles from the boundary. Another six Perimeter stations lie up to 2.5 miles from the Laboratory boundary. Finally, ten On-Site stations are located within the Laboratory boundary. Coordinates for these 23 stations (plus others that have not been sampled since the early 1980s) are supplied on Figure 1 of Fresquez et al. (1995)

In this report, the three ESP categories are modified as shown in the first column of Table 2. The categories used herein are

Remote (6 stations)	• Regional stations that are more than 15 miles from the Laboratory
Marginal (8 stations)	• All of the Perimeter stations except the one at TA-8 • The Otowi Regional station • Two ESP On-Site stations (Test Well DT-9 and TA-33) located in the southeastern part of the Laboratory
Central (9 stations)	• The remaining eight ESP On-Site stations • The TA-8 Perimeter station.

These alternative groupings better reflect the levels of activity found at the ESP stations for radionuclides with significant spatial trends. The proposed baseline data sets are composed of subsets of data collected since 1990 at Marginal and Remote stations.

Some stations are potentially affected by their proximity to operational sources. Most of these are Central stations, such as TA-21 (potentially affected by historical stack emissions from TA-21) and TA-50 (potentially affected by the liquid radioactive waste treatment facility at TA-50). Among the Marginal stations that are candidates for inclusion in baseline data sets, the TA-33 station has been affected by the tritium facility that operated at TA-33 until 1991. All TA-33 data is eliminated from the baseline data set for tritium (see Section 4.1).

Table 2. Environmental Surveillance Soil Sampling Stations

Category(1) and name	ESP category(2) and name	Location
R Bernallilo	R Bernallilo	
R Cochiti	R Cochiti	
R Embudo	R Embudo	
R Jemez	R Jemez	
R Rio Chama	R Rio Chama	
R Santa Cruz	R Santa Cruz	
M North Mesa	P North Mesa	North of Broadview, near the horse stables
M Otowi	R Otowi	West of the intersection of S.R. 501 and S.R. 30
M Rendija Canyon	P Sportsman Club	Near the Sportsman Club in Rendija Canyon
M TA-33	O Near TA-33	Across S.R. 4 from the entrance to TA-33.
M TA-49	P Near TA-49	Along S.R. 4, about 500 feet SE of the entrance to TA-49
M Test Well DT-9	O Near Test Well DT-9	About half way between TA-49 and Ancho Canyon sites
M Tsankawi	P Tsankawi/PM-1	At entrance to Tsankawi on S.R. 4
M White Rock	P White Rock (east)	Near curve of S.R. 4 approaching White Rock from the north
C R-Site Road	O R-Site Road East/ R-Site Road	North side of Potrillo Canyon across from Phermex
C TA-8	P Near TA-8 (GT Site)	Along S.R. 501 directly west of TA-8
C TA-11	O TA-16 (S-Site)/ TA-11	Mesa-top at S-Site, two miles east of S.R. 501 and 1/2 mile north of S.R. 4.
C TA-21	O TA-21 (DP Site)	North of DP Road, about 200 feet NW of the gate into TA-21.
C TA-36	O Potrillo Drive/ TA-36	Mesa-top site along the road into TA 36, about 2000 feet east of gate
C TA-50	O TA-50	North of TA-50.
C TA-53	O West of TA-53	About 350 feet east of East Jemez turnoff along TA-53 access road.
C TA-54	O East of TA-54	At east end of TA-54
C Two Mile Mesa	O Two-Mile Mesa	South of old TA-6 complex

(1) O On-site (within Laboratory boundary)
P Perimeter (less than 3 miles from Laboratory boundary)
R Regional (more than 3 miles from Laboratory boundary)

(2) C Central (not more than 0.1 mile outside boundary, excluding southeastern part of Laboratory)
M Marginal (southeastern part of Laboratory and up to 5 miles from boundary)
R Remote (more than 15 miles from the Laboratory)

3.0 ISSUES AND CAVEATS

3.1 Data Comparability

Whenever two data sets are to be compared, the first issue that arises is always the problem of data comparability, including the comparability of both sampling methods and analytical procedures used in generating the data in the two sets.

The ESP data are intended to represent levels of activity in soils that can be ascribed to atmospheric deposition from sources such as fallout and stack emissions. Soil samples are composites of five 0-2" grab samples collected at the corners and center of a 32 foot square at a given sampling station. (See Chapter 6 of 1995 Environmental Surveillance Program report.) ER surface samples, by contrast, are typically 0-6" grab samples. The fact that the ESP samples are composites will tend to reduce their variability compared to ER grab samples, but such composites do provide better estimates of the average activity at each sampling station. To the extent that what is being measured at either an ESP site or an ER site is atmospheric deposition, the fact that the ESP samples represent only the top two inches of soil may lead to ESP results being biased slightly upward relative what would be expected in a deeper ER 0-6" sample, particularly for the less mobile isotopes. However, neither of these effects is likely to be large compared to the overall variability within the baseline data sets.

The ESP generally uses the most sensitive analytical methods available for its work. For many of the fallout radionuclides, background levels are near the lower limit of detection by these methods. The methods used are documented in by Gautier (1995) and are briefly described in Appendix A. For some analytes, a substantial fraction of the baseline data are below the minimum detectable activities (MDAs) for the analytical method employed, although data are reported as measured even below these levels.

ER samples that are submitted to an offsite analytical laboratory may be handled similarly to ESP samples. However, alternative methods do exist (in particular, gamma spectroscopy for ^{241}Am) which might result in higher detection levels. Even when two samples are analyzed by the same general method, sensitivity is a function of a number of parameters that may vary among laboratories, including standard aliquot size, counting time, and counting geometry. In some cases the MDAs for ER samples may exceed background activity levels and/or the MDAs associated with ESP samples. In addition, differences in method, including sample dissolution method, extraction method, and instrumentation, can lead to relative biases between the results for the same sample.

3.2 Statistical Issues

Once a baseline data set is selected, there remain a number of statistical issues to be addressed before calculating summary statistics. The first and most significant is the use of these data as if they constituted a simple random sample from the baseline population, consisting of sample locations that have not been impacted by a specific LANL release. (This definition of the "baseline population" excludes, for example, locations within a PRS or immediately downwind from a known release point, but locations within the range of influence of some of the more widespread stack releases are included.) The ESP samples from Marginal and Remote stations are not a true random sample of this population. Rather they represent repeated sampling at a handful of stations in a handful years. In theory, observations from one station in different years should not be considered independent observations. In practice, evidence of "station effects" is not found within the proposed baseline data sets, and this evidence of lack of correlation within stations provides at least partial justification for treating the observations as independent.

Evidence of "year effects" is much more common, and is discussed for the affected isotopes in Section 4. These limitations of the the ESP data for the purpose of generating a representative baseline data set should be kept in mind. The ER guidance document mentioned in Section 1 recommends the collection of additional baseline data to address this problem as well as some of the data comparability issues discussed in Section 3.1.

For most of the radionuclides evaluated (all except ^{241}Am), the selected baseline data appear to be approximately lognormally distributed. Some of the data depart from lognormality in the lower tails, but this can be ascribed to the fact that data in these tails is being reported near or below the lower limits of the analytical methods used. For lognormally distributed radionuclides, means, standard deviations, and upper tolerance limits are estimated using lognormal assumptions. These calculations are described in Appendix 2.

The use of logarithms requires substituting positive numbers for zeros and negative values in the data set. (Negative values are sometimes reported because the reduction of the raw data requires subtraction of an instrumental background count.) Some statistical calculations are extremely sensitive to how this substitution is done. A value of 0.5 times the smallest reported positive observation is used for the calculations presented in this report. To further reduce the sensitivity of the estimated parameters describing the baseline distributions to this choice, robust moment estimators are used, as described in Appendix 2.

Normal or lognormal probability plots of the selected baseline data sets are presented for each radionuclide in the following text. Plots of the raw data by station are presented in Appendix 1. In the logarithmic plots, substituted values (i.e., samples for which the results actually reported are zero or negative) are indicated by a square, while measured positive values are indicated by a "+".

A few individual observations appear to be high outliers relative to the remainder of the data. The effect of these observations on the estimates of baseline parameters such as the mean and the UTL would be limited because robust estimators are used, but they are excluded from the baseline data set. In particular, they should not be used when two-sample tests are used to compare site and baseline data. These outliers are explicitly mentioned in the text and tables of Section 4.

One of the statistics calculated is the (.95, 95) UTL for the baseline distribution, which is a 95% upper confidence bound for the true 95th percentile of the baseline population. The calculation of the UTL is based on the robust estimates of mean and standard deviation, as described in Appendix 2. For small data sets (fewer than about 40 observations), the UTL may exceed the unknown 95th percentile by 25% or more, while for larger data sets it should be a better approximation to this percentile. By definition, the 95th percentile of a distribution is exceed by 5% of the observations from that distribution. As a result, some observations in the local background data sets can be expected to exceed the UTLs, but they are nevertheless to be retained (e.g., when these data sets are used in other statistical comparisons) unless they appear to be outliers based on other criteria.

3.3 Spatial Trends

Differences between Central, Marginal and Remote stations are evaluated using nonparametric tests such as the Wilcoxon two-sample tests. These tests suggest statistically significant spatial trends as a function of distance from the central Laboratory for tritium, ^{238}Pu and $^{239+240}Pu$. In particular, for these isotopes the Central sites as a group are significantly elevated relative to the Marginal and Remote sites on which the baseline data sets are based.

Table 3. Statistical Comparison of Individual Central Stations to Baseline Data*

Isotope	^{3}H	^{90}Sr	^{137}Cs	^{238}Pu	$^{239+240}Pu$	^{241}Am
Number of years	4	3	4	4	4	2
R-Site Road	wqu(2)	w		u(1)		
TA-8		w	wqu(1)	u(1)	qu(1)	wqu(1)
TA-11	wq	w	u(1)			
TA-21	wqu(1)		u(1)	wu(1)	wu(1)	
TA-36				u(1)		
TA-50	wu(1)			wqu(1)	wqu(2)	wqu(2)
TA-53	wqu(1)			u(1)	wqu(1)	
TA-54	u(1)		u(1)	wqu(1)	wqu(1)	
Two Mile Mesa	wqu(2)	w	w	w	w	u(1)

* w = Significantly different from baseline (P<0.1) by the Wilcoxon test (nonparametric test for shift of entire station distribution relative to baseline)

q = Significantly different from baseline (P<0 1) by the quantile test (nonparametric test for shift of upper quartile of station distribution relative to baseline)

u(n) = n observations exceed the (.95, 95) UTL calculated from the baseline data This is neither a nonparametric nor a statistical test, but it is a commonly reported statistic.

In addition to these general spatial trends, several of the Central stations individually "fail" one or more statistical tests when compared with the selected baseline data. The results of these tests for the Central stations are summarized in Table 3. (See Ryti et al, 1996, 1298, for a description of the Wilcoxon and quantile tests whose results are tabled in Table 3)

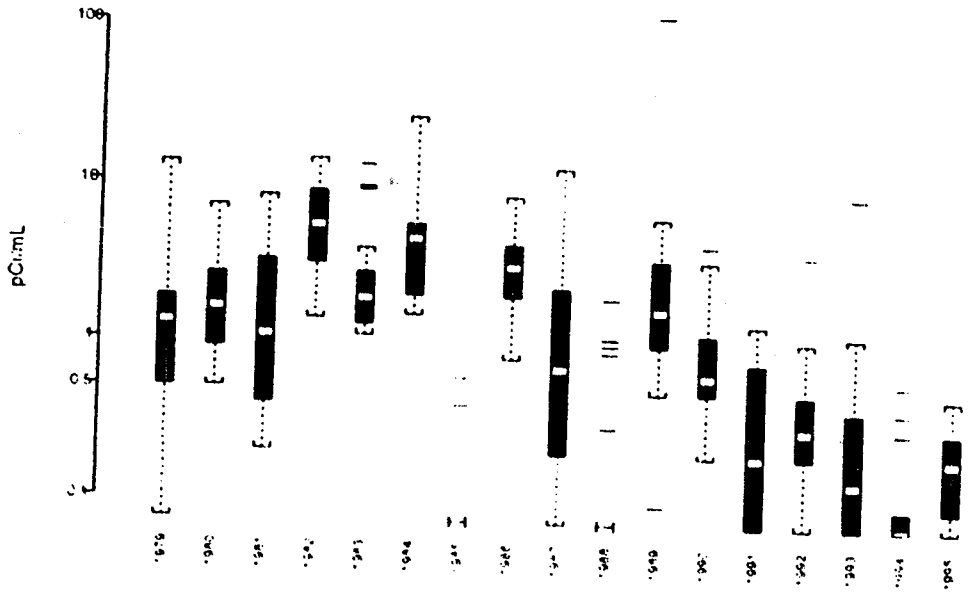
While the Central stations are less likely than Marginal stations to be representative of general Laboratory effects and more likely to be influenced by localized sources, the above calculations do suggest that comparison of centrally located ER sites with the baseline data might result in the conclusion that a release has occurred at this site when in fact it has merely been affected by a non-ER release. Fortunately, the three isotopes with clear spatial trends (^{3}H, ^{238}Pu and $^{239+240}Pu$) have quite high threshholds below which they do not contribute significantly to risk In all three cases, the screening action levels (SALs) used by the ER Project are far above any levels found in the ESP data. Thus the relatively small "false positives" that might result from using a baseline data set that is not entirely appropriate for the central part of the Laboratory should never, by itself, lead to unwarranted corrective action at an ER PRS

4.0 BASELINE DATA SETS

4.1 Tritium

Tritium (^{3}H) in soil moisture has been measured at all 23 stations since the early 1980s. However, the measurements in some years--specifically, 1985, 1988, and 1994--dipped across the entire set of stations, for unexplained reasons (see Figure 1) Excluding these years, tritium activity in soil moisture appears to have been rising in all groups of stations in the late 1970s and early 1980s, and to have decayed since that time more rapidly than would be predicted by its half-life of 12.3 years. In particular, observations have decreased markedly since 1990 The recommended baseline data for tritium are subsets of the data collected in 1991 1992, 1993 and 1995 All of the data for these four years are presented in Appendix A (Table A-1 and Figure A 1)

Figure 1. Temporal and Spatial Trends for ^3H, exluding TA-33



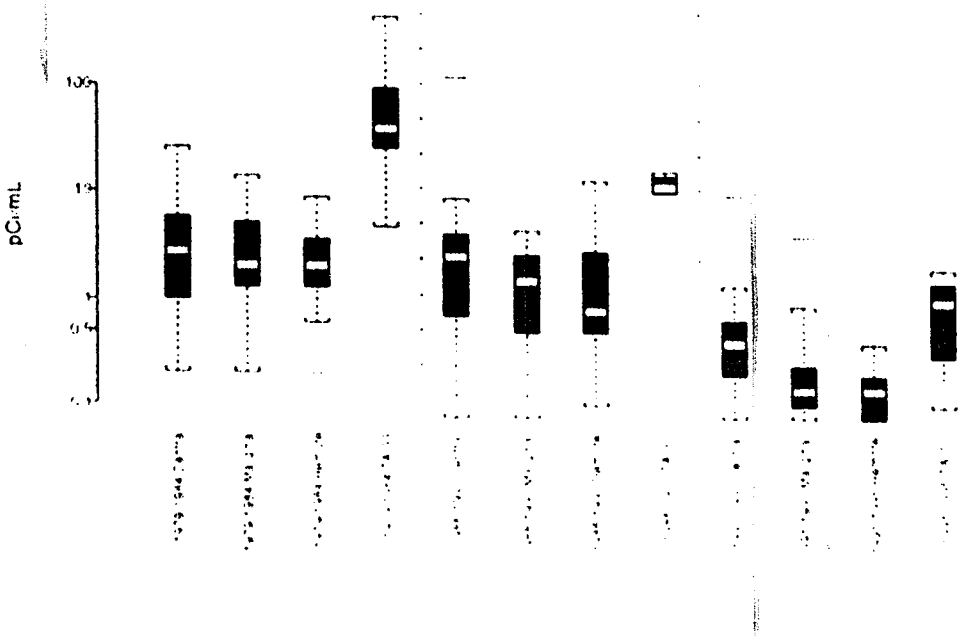
Throughout the years, measurements from the station near TA-33 have been consistently high, a result of the proximity of this station to the tritium facility at TA-33 that operated until 1990 (Figure 2). Since the TA-33 tritium facility has been shut down, measurements at this station have been decreasing, but they are still significantly higher than observations from other Marginal and Remote stations. Therefore data from TA-33 are excluded from the baseline data set and from all statistical tests and calculations reported here.

Figure 2 reveals two observations in the most recent time period that appear as outliers: 9.5 pCi/mL at TA-54 in 1993 (a Central station) and 4 pCi/mL at Test Well DT-9 in 1992 (a Marginal station). The DT-9 observation is exluded from the baseline data set for tritium, both for the purposes of computing estimates of the parameters of the baseline distribution (Table 4) and when this data set is used in other statistical tests. (The TA-54 observation is not a candidate for the baseline data set, as it comes from a Central station.)

In summary, the proposed baseline data set for tritium consists of 51 observations from thirteen Remote and Marginal stations (excluding TA-33 and one outlier from Test Well DT-9), for the years 1991, 1992, 1993 and 1995. Table 4 summarizes the parameters of the baseline distribution as estimated from these data. Figure 3 is a lognormal plot of these data on which the fitted lognormal distribution is represented by the solid straight line and the (.95, 95) UTL value by a horizontal dashed line. One observation (0.9 pCi/L at North Mesa in 1991) exceeds the UTL.

Tritium data are reported to the nearest 0.1 pCi/mL, while reported measurement errors are on the order of 0.2 to 0.4 pCi/mL. The MDA is conservatively estimated as high as 0.5 pCi/mL (Appendix A). Thus most of the reported observations are statistically indisguishable from zero (in fact 30% of them are reported as less than or equal to zero), and only the upper tail of the distribution rises above the level of measurement error. We should be concerned primarily with the fit of the estimated distribution to the upper tail, which appears from Figure 3 to be adequate.

Figure 2. Temporal and Spatial Trends for ^3H, excluding the years 1985, 1988 and 1994

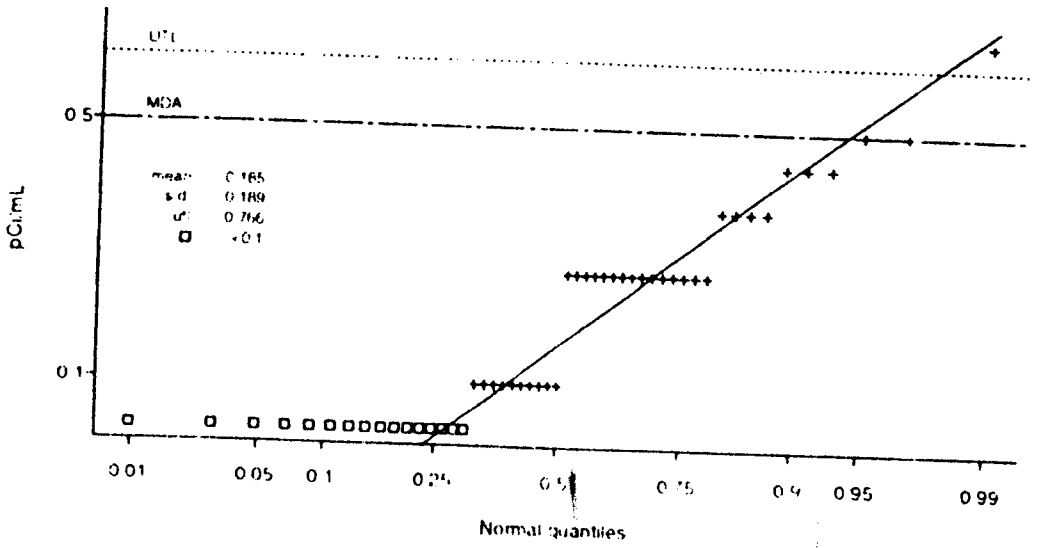


The difference between the Central and Marginal/Remote stations is statistically highly significant (P<.001). Table 3 suggests that this difference is due primarily to observations from the Two Mile Mesa and R-Site Road stations in the west-central part of the Laboratory. Therefore it is possible that the baseline data set, which is based on Marginal and Remote data, may not be completely representative for ER sites the west-central part of LANL, although the effects of this on ER decisions should be negligible, as discussed in Section 3.3.

Table 4. Baseline Statistics for ^3H

Years	1991, 1992, 1993, 1995
Stations	Marginal and Remote excluding TA-33
Excluded outliers	Test Well DT-9 1992
Number of observations used	51
Mean	0.185 pCi/mL
Standard deviation	0.189 pCi/mL
Coefficient of variance	1.02
Smallest positive value	0.1 pCi/mL
Median	0.1 pCi/mL
Maximum	0.9 pCi/mL
(.95,.95) UTL	0.766 pCi/mL
Number of observations above the UTL	1

Figure 3. Lognormal Plot of Baseline Data for ^{3}H



Within the baseline data set, there are no statistically significant differences among stations or between the Marginal and Remote stations, and the difference among years is only marginally significant (P~0 09). As explained in Section 3.2, this provides some justification for treating these data as an independent and identically distributed sample from the baseline distribution for the purposes of the statistical calculations summarized in Table 4, although strictly speaking they do not form such a sample.

4.2 Strontium-90

Strontium-90 (^{90}Sr) was measured several times in the late 1970s and early 1980s, but recently it has been included only in the 1992, 1994 and 1995 surveys, where it was measured at all 23 stations listed in Table 2. Data for these three years are presented in Appendix A in Table A-2 and Figure A 2

Overall, a decrease in activity comparable to that expected based on a half-life of 29.1 years is observed between the earlier measurements and those made in the 1990s (Figure 4).

Differences between Central, Marginal and Remote stations that were highly significant in the 1970s and 1980s are statistically insignificant in the 1990s No individual observations appear to be outliers

The recommended baseline data set for ^{90}Sr consists of the most recent three years of data from the 14 Remote and Marginal stations. Statistics based on this data set are presented in Table 5. Figure 5 is a log-normal probability plot of these 42 observations None exceed the (.95, 95) UTL

Figure 4. Temporal and Spatial Trends for ^{90}Sr

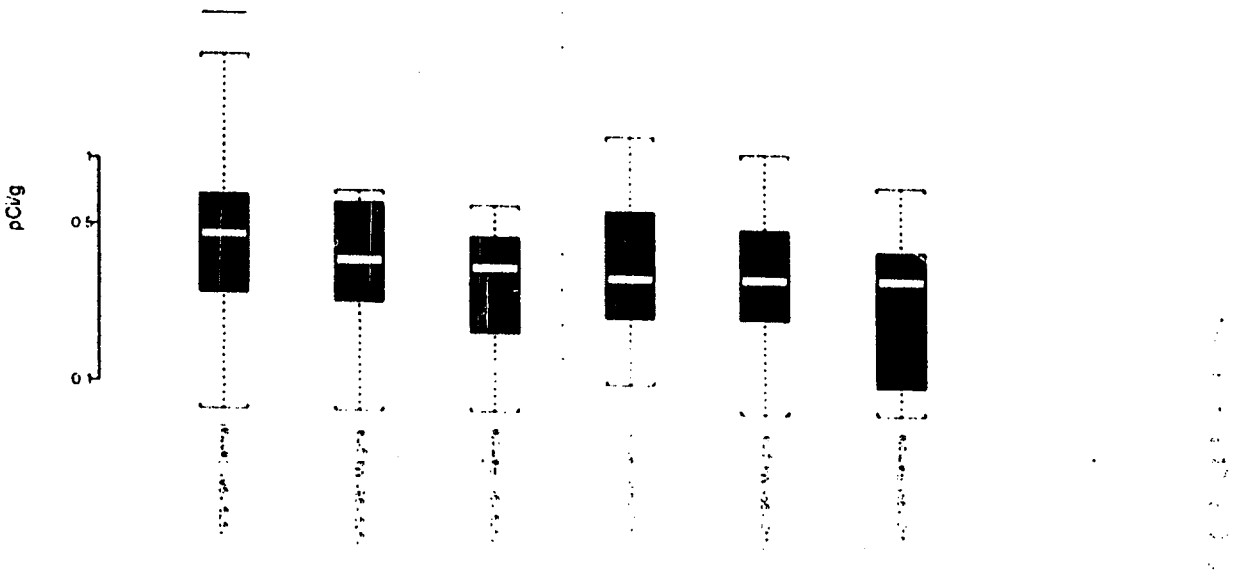


Figure 5. Lognormal Plot of Baseline Data for ^{90}Sr

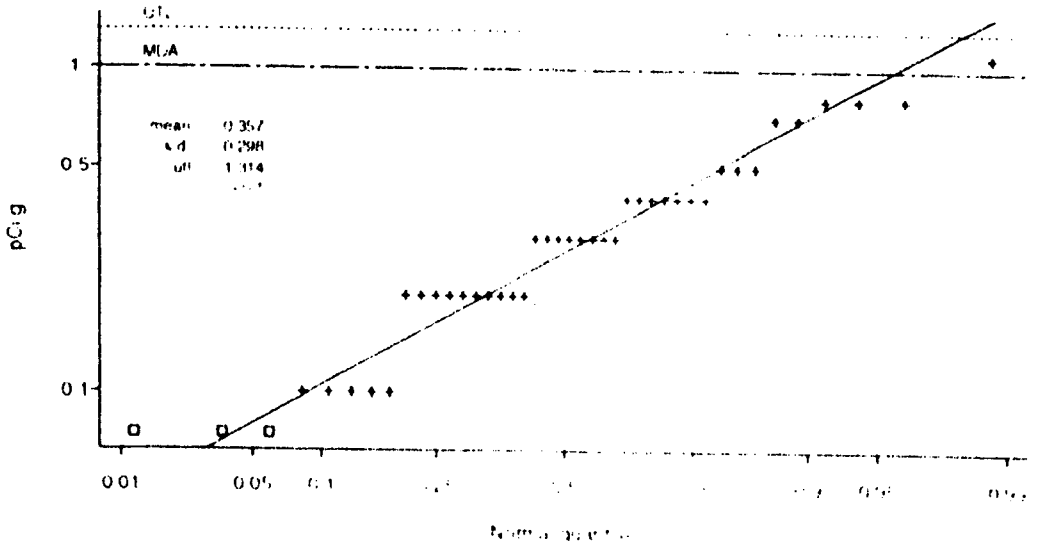


Table 5. Baseline Statistics for ^{90}Sr

Years	1992, 1994, 1995
Stations	**Central/Marginal**
Number of observations	42
Mean	0.36 pCi/g
Standard deviation	0.30 pCi/g
Coefficient of variance	0.83
Smallest positive value	0.1 pCi/g
Median	0.3 pCi/g
Maximum	1.1 pCi/g
(.95,.95) UTL	1.31 pCi/g
Number of observations above the UTL	0

Strontium-90 data are reported to the nearest 0.1 pCi/g and the reported measurement error is typically 0.2 pCi/g. The MDA is conservatively estimated as high as 1 pCi/g (Appendix A). Thus most of the reported observations are statistically indisguishable from zero although only a handful are reported as less than or equal to zero, and only the upper tail of the distribution rises above the level of measurement error. We should be concerned primarily with the fit of the estimated distribution to the upper tail, which appears from Figure 5 to be adequate

Analysis of variance of the baseline data by year and region shows no significant differences between Central, Marginal and Remote stations in the 1990s, but there is a highly significant year effect ($P<0.01$) The interquartile ranges for the three years are (0.1, 0.3) for 1992, (0.3, 0.7) for 1994, and (0.2, 0.4) for 1995, so in fact the data for 1992 and 1994 hardly overlap. The presence of this unexplained effect suggests that measurement of ^{90}Sr may be significantly affected by environmental factors such as humidity during the periods when samples were collected.

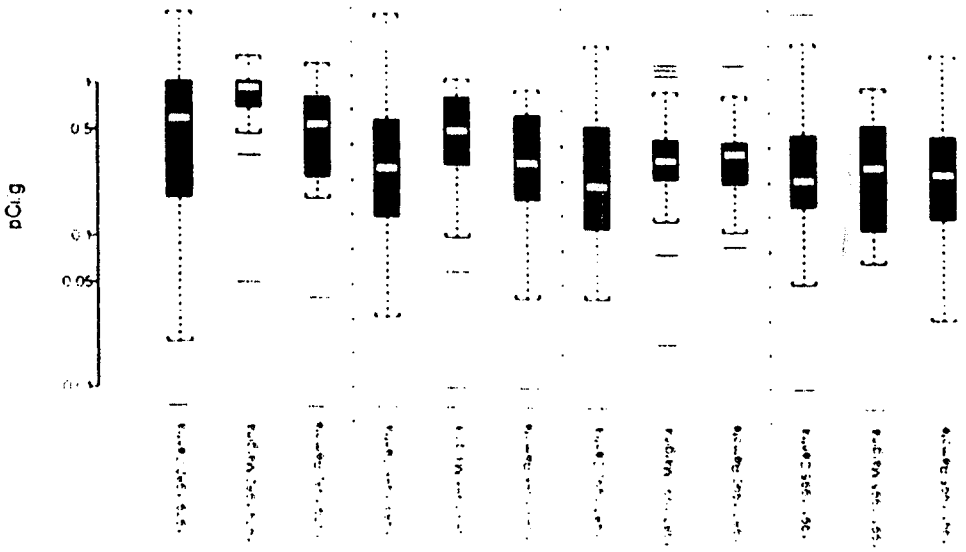
The stations with the highest measured activities in the 1990s are located in the western part of the Laboratory—TA-8, TA-11, R-Site Road and Two Mile Mesa Individually these stations are found to be significantly different from background by the Wilcoxon rank-sum test (see Table 3). Therefore it is possible that the baseline data set, based on Marginal and Remote data, may not be completely representative for ER sites the west-central part of LANL The effects of this on ER decisions should be small, although the SAL for ^{90}Sr (4.4 pCi/g) is closer to the background levels than for the three isotopes discussed in Section 3.3 for which the spatial trends are more widespread.

4.3 Cesium-137

Cesium-137 (^{137}Cs) has been sampled at the 23 locations in Table 2 since the mid 1980s, except for the year 1990 when no ^{137}Cs results were reported at the Perimeter stations The general trend since the early 1980s is consistent with the 30-year half-life, except for an unexplained but significant dip in almost all reported results in 1992

Differences between Central Marginal, and Remote stations that were significant in the early 1980s are no longer statistically significant in the data collected since 1990 The highest measurements are recorded at stations on the western side of the Laboratory In particular, observations from TA-8 are consistently high, although not higher than isolated observations from several other stations and observations from TA-8, as well as those from the anomalous year 1992, are excluded from Figure 6 (The TA-8 measurements are shown Figure A 3 in Appendix A however)

Figure 6. Temporal and Spatial Trends for ^{137}Cs, excluding station TA-8 and year 1992



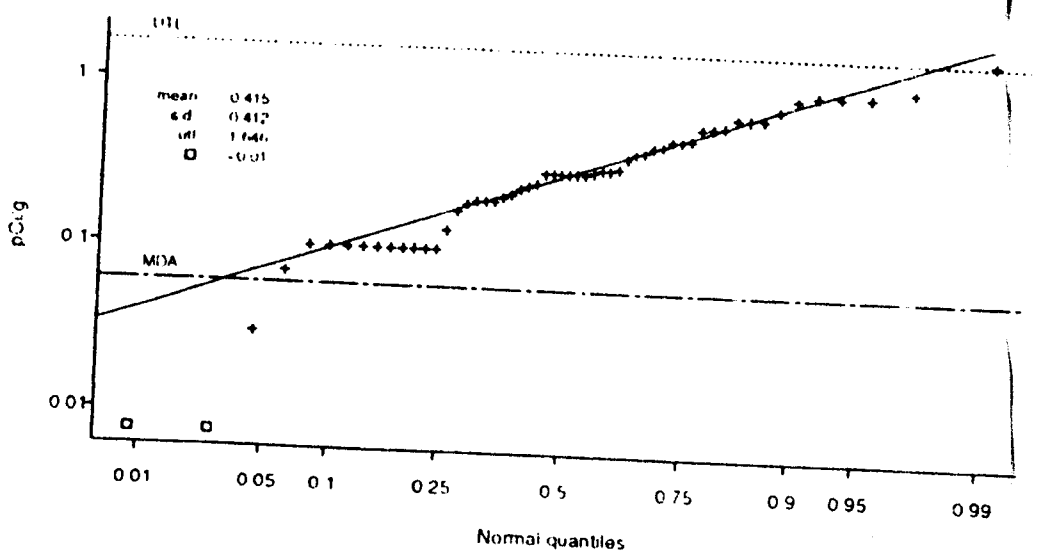
The recommended baseline data set consists of data from all Marginal and Remote stations for the years 1990-1995, excluding 1992, a total of 56 observations. Statistics based on this data set are summarized in Table 5. All of the data for these five years are presented in Appendix A (Table A-3).

One observation in the baseline data set, from the Cochiti Regional station in 1991, exceeds the (.95,.95) UTL, but it is well in line with a lognormal distribution fit to the middle of the data (Figure 7); that is, it does not appear to be an outlier relative to the bulk of the data.

Table 6. Baseline Statistics for ^{137}Cs

Years	**1990-1991, 1993-1995**
Stations	**Marginal and Remote**
Number of observations	56
Mean	0.42 pCi/g
Standard deviation	0.41 pCi/g
Coefficient of variance	0.99
Smallest positive observation	0.03 pCi/g
Median	0.3 pCi/g
Maximum	1.7 pCi/g
(.95, .95) UTL	1.65 pCi/g
Number of observations above the UTL	1

Figure 7. Lognormal Plot of Baseline Data for ^{137}Cs



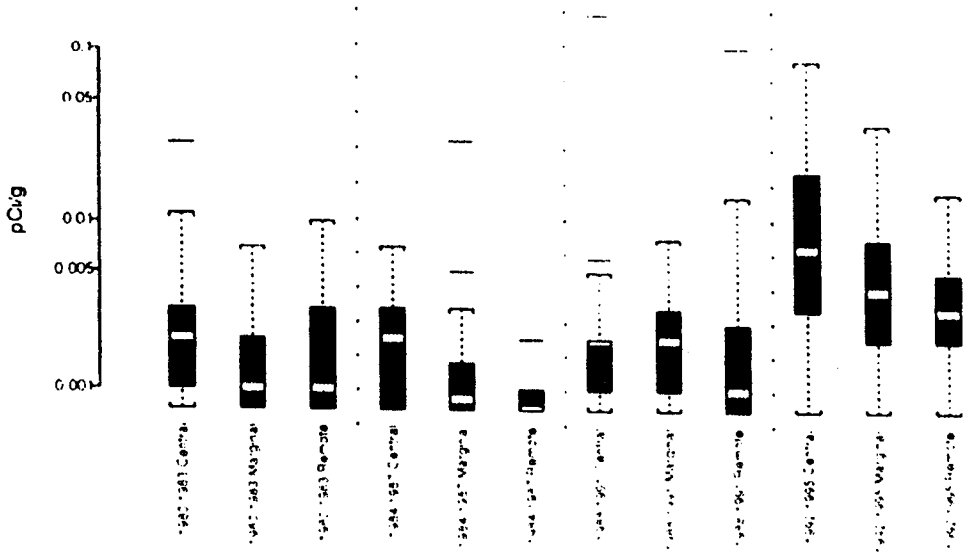
Observations in the lower tail of the data set are not fit by the estimated lognormal distribution, which is represented in Figure 7 by a solid line. Although activities as small as 0.01 pCi/g are reported, the measurement errors are typically on the order of 0.03 to 0.1 pCi/g and the estimated MDA is 0.06 pCi/g (Appendix A), so the data in the lower tail are not reliable. However, these data have been downweighted by the robust estimation procedure which is described in Appendix B, and the resulting estimate appears to be a reasonable representation for the bulk of the data.

Analysis of variance of the baseline data by year and region shows a marginally significant "year effect" ($P<0.1$), which appears to be due to some low observations in 1993. The data for 1991 through 1993 are reported to the nearest tenth of a pCi/g, while for other years (both before and after) they are reported to the nearest hundredth. The anomalous results for 1993 appear to be, at least in part, a result of rounding down to 0.1 pCi/g. There are no significant differences between the Remote, Marginal and Central subsets of data. Individually, observations from the TA-8 and Two Mile Mesa stations are significantly elevated relative to the baseline data set (see Table 3).

4.4 Plutonium-238

Plutonium-238 (^{238}Pu) has been sampled consistently at the 23 locations in Table 2 since the mid 1980s, except for the year 1990 when no ^{238}Pu results were reported at the Perimeter stations. A significant increase with time is observed in all categories of sampling stations starting in the late 1980s. (See Figure 8.) In addition, statistically significant differences among the categories appear in this time frame.

Figure 8. Temporal and Spatial Trends for ^{238}Pu*



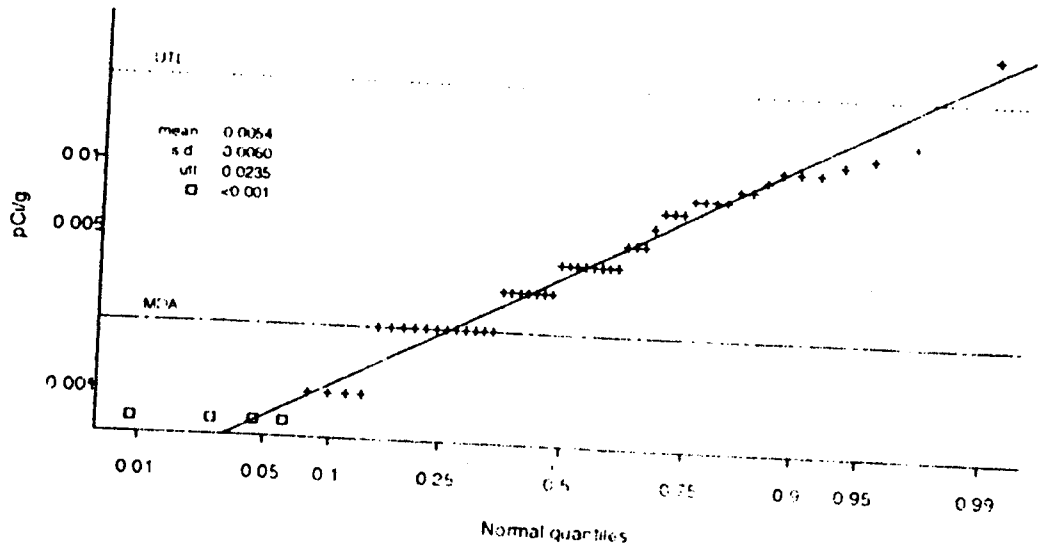
* Two extreme outliers, values greater than 1 pCi/g reported at TA 54 in 1980 and in 1985, have been eliminated from this plot.

The recommended baseline data set for ^{238}Pu consists of the data from the Marginal and Remote stations from the most recent four years of sampling (1992-1995) All of the data for these four years are presented in Appendix A (Table A-4 and Figure A 4). Statistics based on this data set are presented in Table 6. One observation in the baseline data set (0 37 pCi/g at the Rendija Canyon Perimeter station in 1995) exceeds the UTL In Figure 9, this observations appears not far off the fitted distribution (the solid line), and so it is not considered an outlier and is not eliminated from the baseline data set

Table 7. Baseline Statistics for ^{238}Pu

Years	**1992-1995**
Stations	**Marginal/Remote**
Number of observations	56
Mean	0.0054 pCi/g
Standard deviation	0.0060 pCi/g
Coefficient of variance	1.11
Minimum positive value	0 001 pCi/g
Median	0.004 pCi/g
Maximum	0.037 pCi/g
(95, 95) UTL	0.023 pCi/g
Number of observations above the UTL	1

Figure 9. Lognormal Plot of Baseline Data for ^{238}Pu



Plutonium-238 results are reported to the nearest 0.001 pCi/g with reported measurement errors of 0.001 or 0.002 pCi/g and the estimated MDA at 0.002 pCi/g (Appendix A). The majority of the observations in the baseline data sets are thus above the level of measurement error, although quantization effects are apparent in the lower tail of the data in Figure 9.

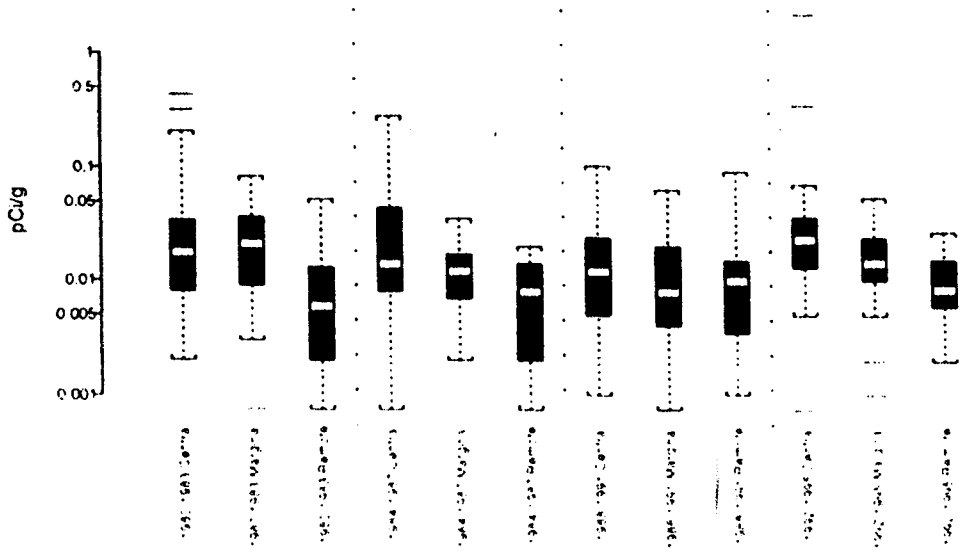
The difference between the Central and Marginal/Remote stations is statistically highly significant ($P < .001$). Table 3 suggests that this difference is quite widespread across the central Laboratory. One observation out of four from seven of the nine Central stations exceeds the UTL, and two stations (at TA-50 and TA-54) are significantly different from the baseline by other statistical tests. Therefore it is possible that the baseline data set based on Marginal and Remote data may not be representative for ER sites the central part of the Laboratory, but the effects of this on ER decisions should be negligible, as discussed in Section 3.3.

Within the baseline data set there are no statistically significant differents among stations or years, or between Marginal and Remote stations.

4.5 Plutonium-239/240

Plutonium-239/240 ($^{239+240}Pu$) has been sampled consistently at the 23 locations in Table 2 since the mid 1980s, except for the year 1990 when no $^{239+240}Pu$ results were reported at the Perimeter stations. In the earlier part of this period higher levels were measured near the Laboratory than at Remote stations. More recently the differences have been less pronounced but they remain statistically significant in the most recent time period (1992-1995), see Figure 10. The changes between this most recent time period and the preceding time period (1988-1991) are also statistically significant for Central and for Marginal stations.

Figure 10. Temporal and Spatial Trends for $^{239+240}Pu$



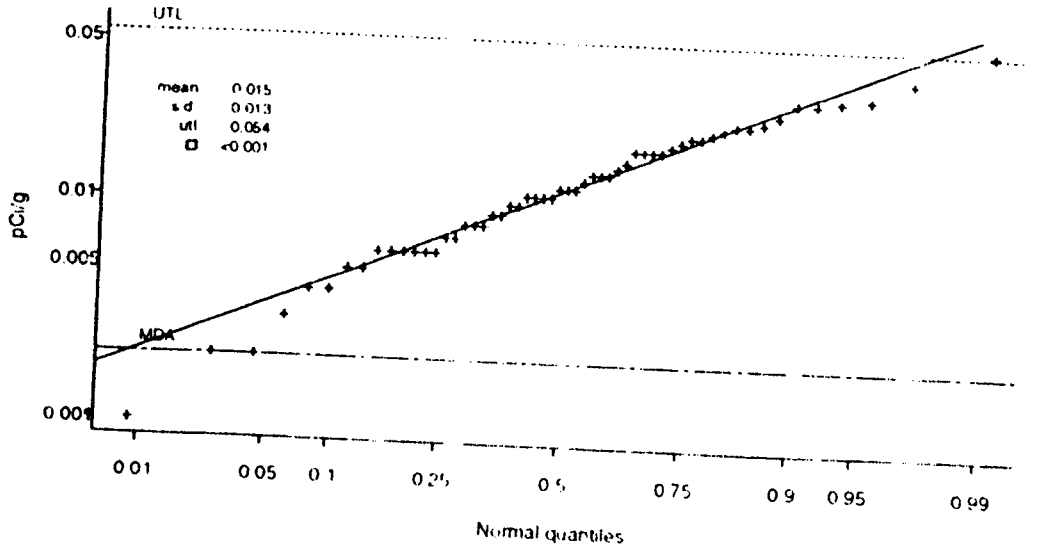
The recommended baseline data sets for $^{239+240}Pu$ consist of subsets of the data from the Marginal and Remote stations drom the most recent four years of sampling (1992-1995) All of the data for these four years are presented in Appendix A (Table A-5 and Figure A 5) Statistics based on this data set are presented in Table 7. One observation in the baseline data set (0.55 pCi/g at the Otowi Remote/Marginal station) exceeds the UTL. In Figure 11, this observation appears to be well in line with the remainder of the baseline data, so it is not considered an outlier.

Plutonium-239/240 results are reported to the nearest 0 001 pCi/g. Reported measurement errors are typically 0.001 or 0 002 pCi/g and the estimated MDA is 0.002 pCi/g (Appendix A) Most of the observations in both baseline data sets are well above the level of measurement error.

Table 8. Baseline Statistics for $^{239+240}Pu$

Years	1992-1995
Stations	Marginal/Remote
Number of observations	56
Mean	0.015 pCi/g
Standard deviation	0 013 pCi/g
Coefficient of variance	0 84
Smallest positive observation	0 001 pCi/g
Median	0 012 pCi/g
Maximum	0 055 pCi/g
(95, 95) UTL	0 054 pCi/g
Number of observations above the UTL	1

Figure 11. Lognormal Plot of Baseline Data for $^{239+240}Pu$



The difference between the Central and Marginal/Remote stations is statistically highly significant (P< 001). Table 3 suggests that this difference is due primarily to data from the TA-50 and TA-53 stations in the north-central part of the Laboratory, but three other Central stations also have one observation out of four above the baseline UTL. Therefore it is possible that the baseline data set based on Marginal and Remote data may not be representative for ER sites the central part of the Laboratory, but the effects of this on ER decisions should be negligible, as discussed in Section 3.3.

In addition to the significant elevation of the Central stations, there is a statistically significant difference between Marginal and Remote stations (P<0.05) for $^{239+240}Pu$ in the most recent time period. A (.95,.95) UTL based on the Remote data only is slightly smaller (0.044 pCi/g), a figure which is still exceeded by only the 1992 Otowi measurement A (.95,.95) UTL based on the Marginal data only is slightly larger (0.062 pCi/g), greater than the largest Marginal observation. In the interests of consistency with the other sections of this paper, the UTL based on the combined Marginal and Remote stations has been selected, but for sites near or within the Laboratory boundaries this number may be slightly low

4.6 Americium-241

Americium-241 (^{241}Am) was measured infrequently prior to the 1990s Recently it has been measured in 1992, 1994 and 1995 at most of the 23 stations listed in Table 2. No observations were reported from the R-Site Road or North Mesa stations in 1995 The1992 measurements are significantly higher than results reported in 1994 and 1995 Probably a different analytical technique (gamma spectroscopy) was used in 1992, reported measurement errors are also large compared to more recent years Consequently, only data from 1994 and 1995 are considered for the baseline data set Data for these two years are presented in Table A-6 and Figure A 6 of Appendix A

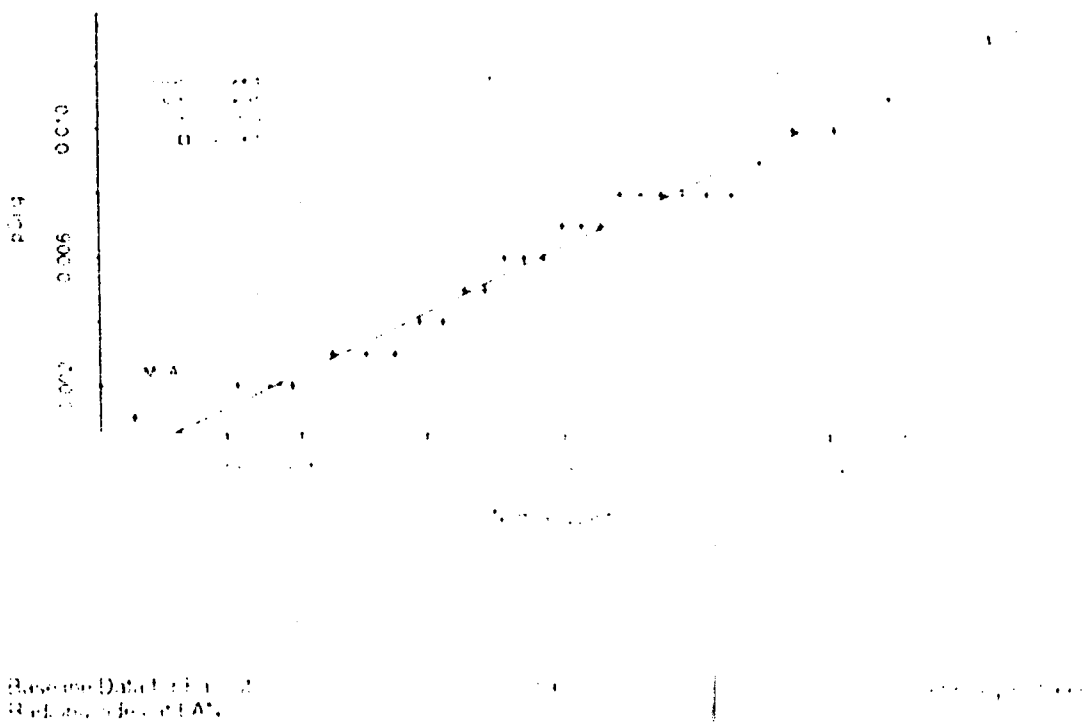
Table 9. Baseline Statistics for ^{241}Am

Years	1994, 1995
Stations	**Marginal/Remote**
Number of observations	27
Mean	0.0064 pCi/g
Standard deviation	0.0031 pCi/g
Coefficient of variance	0.49
Minimum positive value	0.001 pCi/g
Median	0.007 pCi/g
Maximum	0.013 pCi/g
(.95, .95) UTL	0.0133 pCi/g
Number of observations above the UTL	0

The recommended baseline data set for ^{241}Am consists of the most recent two years of data from the Marginal and Remote stations, a total of 27 observations. Statistics based on this data set are presented in Table 9. Unlike baseline data for the other isotopes, these data are very nearly normally distributed, so the statistics in Table 9 were calculated using normal assumptions. Figure 12 is a normal probability plot of the baseline data. The UTL is not exceeded by any observation in the baseline data set.

Americium-241 results are reported to the nearest 0.001 pCi/g with a reported measurement error of 0.001pCi/g. The estimated MDA is 0.002 pCi/g (Appendix A). Most of the observations in the baseline data sets are thus above the level of measurement error, although quantization effects are visible in the lower tail of the data in Figure 12.

Figure 12. Normal Plot of Baseline Data for ^{241}Am



In this time period, differences among Central, Marginal and Remote stations are statistically insignificant, although two high observations from TA-50 suggest that this station may be affected by the facilities at TA-50. There are no statistically significant differences between stations or years in the baseline data set.

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Appendix A. Environmental Surveillance Data

This appendix contains the data reported by the LANL Environmental Surveillance Program for the isotopes and years used to construct local background data sets. The data used in the baseline data sets are shown in bold type in Tables A-1 through A-6, while data from the Central stations and other data not used in the baseline set are shown in plain type. Complete data, except for 1995, can be found in the report by Fresquez et al. (1996). The 1995 data are available in the most recent Environmental Surveillance Program report (1996).

Analytical Methods

The methods used are documented by Gautier (1995). The LANL method numbers given below are from that document.

Tritium (^{3}H) is measured by a distillation and liquid scintillation counting procedure (LANL Method ER210). Tritiated water is distilled from the soil sample. An aliquot of 5 mL is pipetted into a polyethylene liquid-scintillation vial to which a luminescent scintillation cocktail is added. The decay of ^{3}H is observed by liquid scintillation, typically for a 60-minute count period. The MDA of 0.5 pCi/mL given in ER210 represents the "three-sigma" limit for background, and is exceeded by only one observation in the baseline data set. If the method detection limit were calculated by the more standard method of Currie (1968), its value would be closer to 0.75 pCi/L, close to the UTL reported in Section 4. The standard deviation ("one sigma") of the counting error associated with individual measurements is typically 0.3 pCi/L. Thus virtually all of the data in the baseline data set (Table 1.1 and Figure 1.1) are within the range of measurement "noise"

Strontium-90 (^{90}Sr) is measured by gas flow proportional beta counting (LANL Method ER190). Soil samples (typically a 2.0 g aliquot) are extracted twice, allowing an intervening Yttrium-90 (^{90}Y) ingrowth period of at least two weeks. Yttrium-90, the daughter product of ^{90}Sr decay, is beta-counted in the final dried filtrate for 200 minutes ER160 reports a detection limit of 1 pCi/g in soils, although recent improvements to the procedure will reduce this to about 0.5 pCi/g. The "one-sigma" error commonly reported with ESP measurements is on the order of 0.2 to 0.3 pCi/g. Thus about 85% of the data in the baseline data set (Table 1.2, Figure 1.2) are within the two standard deviations of zero.

Cesium-137 (^{137}Cs) is measured using gamma-ray spectrometry (LANL Method ER130) Soil samples are dried and milled, and a 100 g aliquot is counted One estimate of the detection limit for ^{137}Cs is 0.06 pCi/g. However, the associated "one-sigma" errors reported with ESP measurements are typically 0.05 to 0.1 pCi/g About two-thirds of the data in the background data set are above the detection limit

Plutonium-238 (^{238}Pu) and Plutonium-239/240 ($^{238/240}Pu$) are measured by radiochemistry and alpha spectrometry (LANL Method ER160) Americium-241 (^{241}Am) can also be obtained by this method if the appropriate tracer is added, or by the similar LANL Method ER110 which includes an additional solvent extraction step Ten-gram samples are ashed and plutonium is isolated by ion exchange and electrodeposited on stainless steel planchets to be counted by alpha spectroscopy The average MDA of 0.002 pCi/g, and the "one-sigma" errors reported with ESP measurements are typically 0.001 to 0.003 pCi/g Thus about half of the ^{238}Pu and ^{241}Am data and most of the $^{239/240}Pu$ data in the background data set are above noise levels.

Table A-1. Tritium (pCi/L)

	1991	1992	1993	1995
Bernalillo	-0.1	0.2	0.0	0.2
Cochiti	0.1	-0.1	0.2	0.2
Embudo	-0.2	0.0	-0.2	0.2
Jemez	-0.1	0.2	-1.6	0.3
North Mesa	0.9	0.3	0.3	0.2
Otowi	0.5	0.1	0.0	0.1
R-Site Road	0.8	0.6	1.2	0.4
Rendija Canyon	0.1	0.5	0.4	0.2
Rio Chama	0.2	0.4	0.2	0.1
Santa Cruz	-0.2	0.3	-0.1	0.4
TA-11	0.3	0.5	0.4	0.3
TA-21	1.4	0.4	0.5	0.2
TA-33	2.0	0.9	1.1	0.1
TA-36	0.3	0.2	-0.2	0.3
TA-49	-0.1	0.2	0.0	0.2
TA-50	1.2	0.5	0.1	0.2
TA-53	1.2	0.4	0.4	0.5
TA-54	0.2	0.2	9.5	0.0
TA-8	0.2	0.5	-0.2	-0.1
Test Well DT-9	-0.3	4.0	0.2	0.2
Tsankawi	0.1	0.2	-0.1	0.1
Two Mile Mesa	0.8	1.1	0.7	0.3
White Rock	0.1	0.1	-0.1	0.1

Figure A.1. Tritium by Station

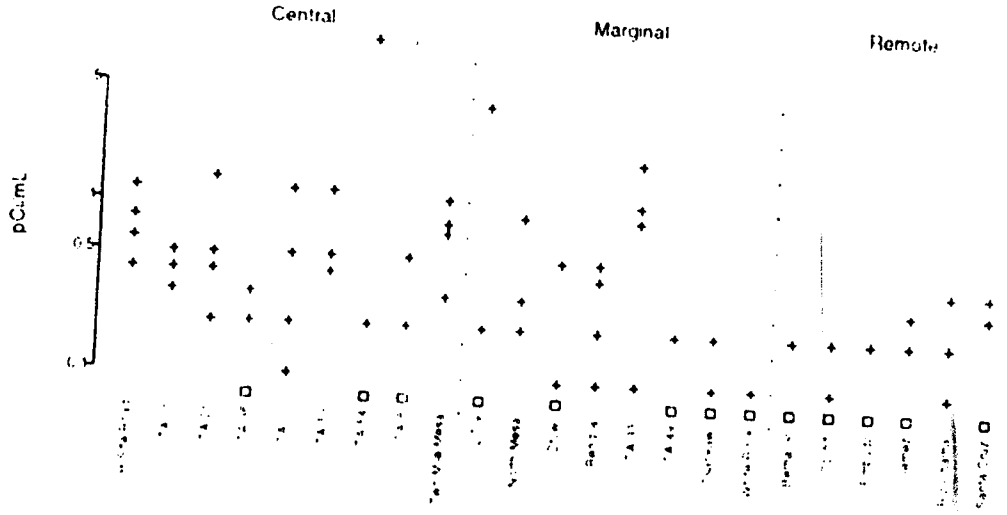


Table A-2. Strontium-90 (pCi/g)

	1992	1994	1995
Bernalillo	0.2	0.8	0.1
Cochiti	0.2	0.4	0.3
Embudo	0.3	0.4	0.1
Jemez	0.3	0.4	0.3
North Mesa	0.3	0.2	0.2
Otowi	0.0	0.7	0.5
R-Site Road	0.2	0.7	1.3
Rendija Canyon	0.3	0.3	0.8
Rio Chama	0.0	0.5	0.1
Santa Cruz	0.1	0.2	0.4
TA-11	0.3	0.5	1.1
TA-21	0.1	0.3	0.1
TA-33	0.2	1.1	0.2
TA-36	0.2	0.5	0.3
TA-49	0.2	0.4	0.3
TA-50	0.1	0.3	0.8
TA-53	0.2	0.4	0.2
TA-54	0.2	0.2	0.5
TA-8	0.4	1.1	0.5
Test Well DT-9	0.0	0.8	0.2
Tsankawi	0.4	0.7	0.4
Two Mile Mesa	0.3	0.4	0.6
White Rock	0.2	0.1	0.4

Figure A.2 Strontium-90 by Station

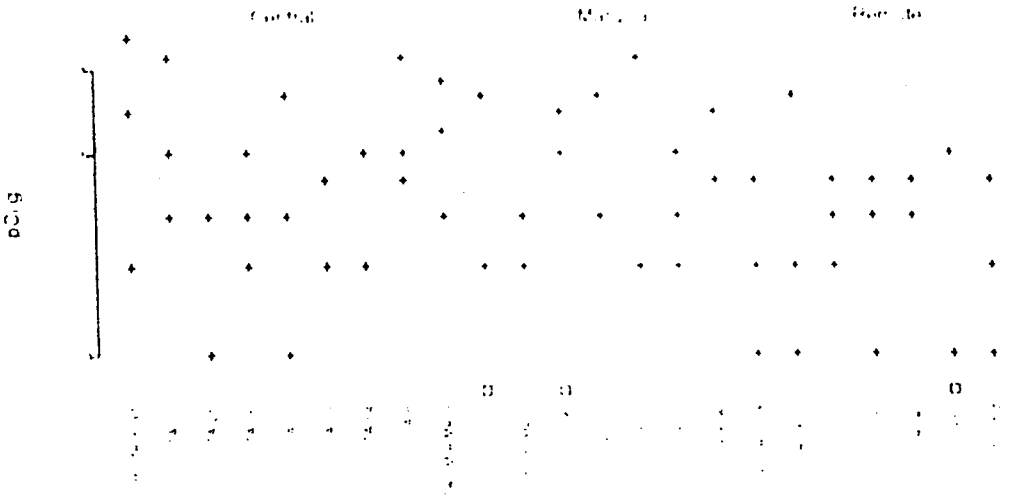


Table A-3. Cesium-137 (pCi/g)

	1991	1993	1994	1995
Bernalillo	0.3	0.1	0.03	0.24
Cochiti	1.7	0.1	0.22	0.10
Embudo	0.8	0.6	0.38	0.45
Jemez	0.2	0.1	0.33	0.50
North Mesa	0.3	0.1	0.07	0.32
Otowi	0.5	0.1	0.17	0.51
R-Site Road	0.5	0.4	0.31	0.57
Rendija Canyon	0.3	0.2	0.26	0.62
Rio Chama	0.2	0.7	0.19	0.25
Santa Cruz	1.1	0.1	0.31	0.46
TA-11	0.2	3.1	0.15	0.46
TA-21	1.9	0.1	0.01	0.18
TA-33	0.3	0.1	0.61	0.00
TA-36	0.3	0.2	0.20	0.30
TA-49	1.0	1.0	0.42	0.41
TA 50	0.1	0.2	0.08	0.62
TA-53	0.3	0.2	0.05	0.33
TA-54	0.2	2.0	0.10	0.10
TA-8	1.2	2.1	0.75	1.21
Test Well DT-9	0.3	1.0	0.94	0.32
Tsankawi	0.7	0.1	0.10	0.13
Two Mile Mesa	0.2	0.7	0.90	0.47
White Rock	0.7	0.0	0.21	0.30

Figure A.3. Cesium-137 by Station

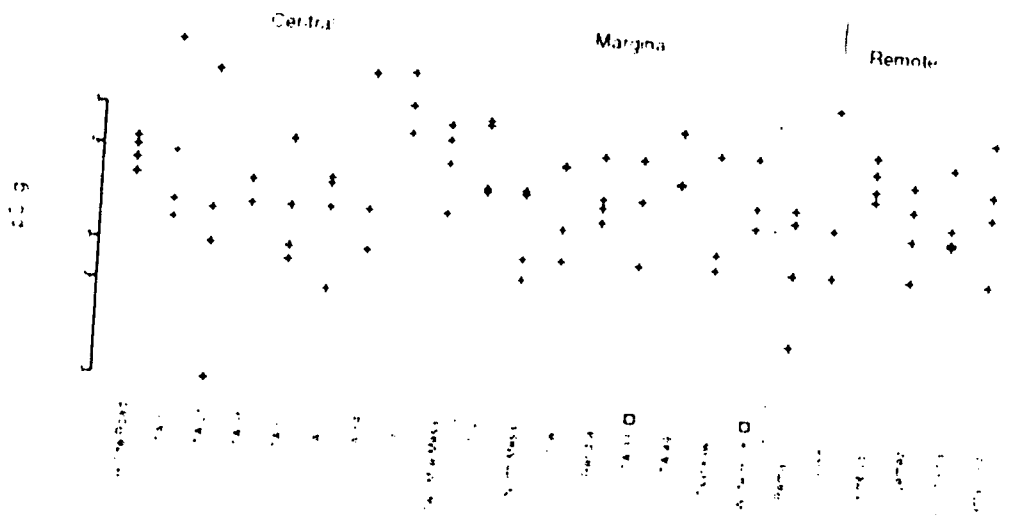


Table A-4. Plutonium-238 (pCi/g)

	1992	1993	1994	1995
Bernalillo	**0.002**	**0.003**	**0.006**	**0.011**
Cochiti	**0.004**	**0.010**	**0.009**	**0.007**
Embudo	**0.021**	**0.018**	**0.023**	**0.018**
Jemez	**0.007**	**0.006**	**0.013**	**0.012**
North Mesa	**0.016**	**0.015**	**0.005**	**0.018**
Otowi	**0.055**	**0.024**	**0.009**	**0.019**
R-Site Road	0.009	0.015	0.016	0.025
Rendija Canyon	**0.012**	**0.006**	**0.018**	**0.040**
Rio Chama	**0.005**	**0.027**	**0.008**	**0.006**
Santa Cruz	**0.008**	**0.004**	**0.010**	**0.021**
TA-11	0.005	0.022	0.007	0.024
TA-21	0.013	0.030	0.009	0.071
TA-33	**0.011**	**0.001**	**0.022**	**0.014**
TA-36	0.046	0.009	0.009	0.013
TA-49	**0.008**	**0.033**	**0.020**	**0.024**
TA-50	0.023	0.062	0.032	0.351
TA-53	0.061	0.030	0.019	0.030
TA-54	0.034	2.248	0.005	0.024
TA-8	0.022	0.068	0.000	0.045
Test Well DT-9	**0.032**	**0.031**	**0.031**	**0.014**
Tsankawi	**0.011**	**0.025**	**0.006**	**0.006**
Two Mile Mesa	0.017	0.024	0.039	0.021
White Rock	**0.014**	**0.002**	**0.011**	**0.012**

Figure A.4 Plutonium-238 by Station

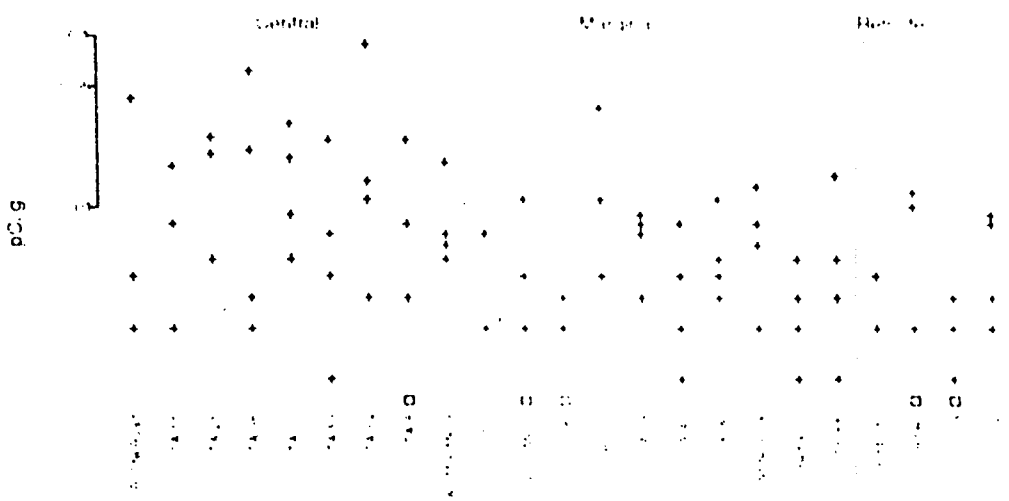


Table A-5. Plutonium-239/240 (pCi/g)

	1992	1993	1994	1995
Bernalillo	0.002	0.003	0.006	0.011
Cochiti	0.004	0.010	0.009	0.007
Embudo	0.021	0.018	0.023	0.018
Jemez	0.007	0.006	0.013	0.012
North Mesa	0.016	0.015	0.005	0.018
Otowi	0.055	0.024	0.009	0.019
R-Site Road	0.009	0.015	0.016	0.025
Rendija Canyon	0.012	0.006	0.018	0.040
Rio Chama	0.005	0.027	0.008	0.006
Santa Cruz	0.008	0.004	0.010	0.021
TA-11	0.005	0.022	0.007	0.024
TA-21	0.013	0.030	0.009	0.071
TA-33	0.011	0.001	0.022	0.014
TA-36	0.046	0.009	0.009	0.013
TA-49	0.008	0.033	0.020	0.024
TA-50	0.023	0.062	0.032	0.351
TA-53	0.061	0.030	0.019	0.030
TA-54	0.034	2.248	0.005	0.024
TA-8	0.022	0.068	0.000	0.045
Test Well DT-9	0.032	0.031	0.031	0.014
Tsankawi	0.011	0.025	0.006	0.006
Two Mile Mesa	0.017	0.024	0.039	0.021
White Rock	0.014	0.002	0.011	0.012

Figure A.5. Plutonium-239 by Station

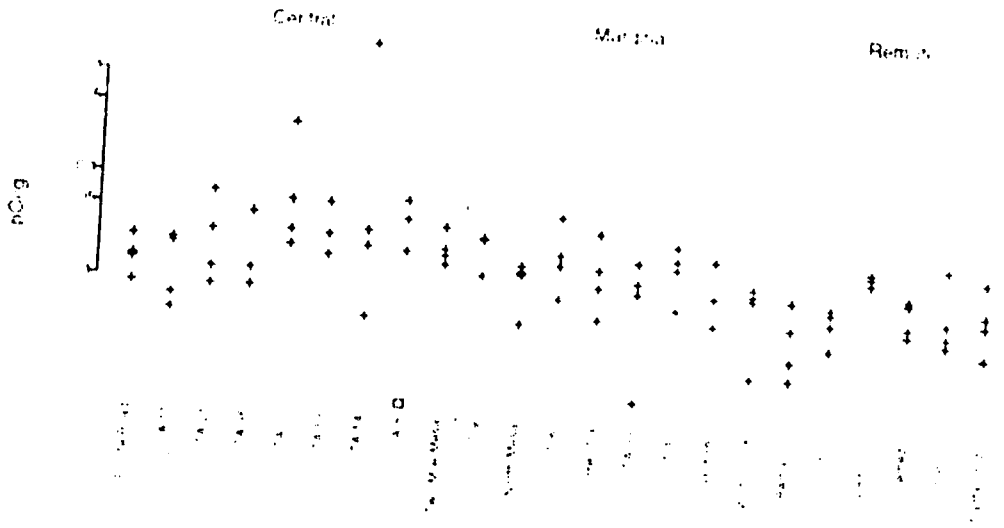
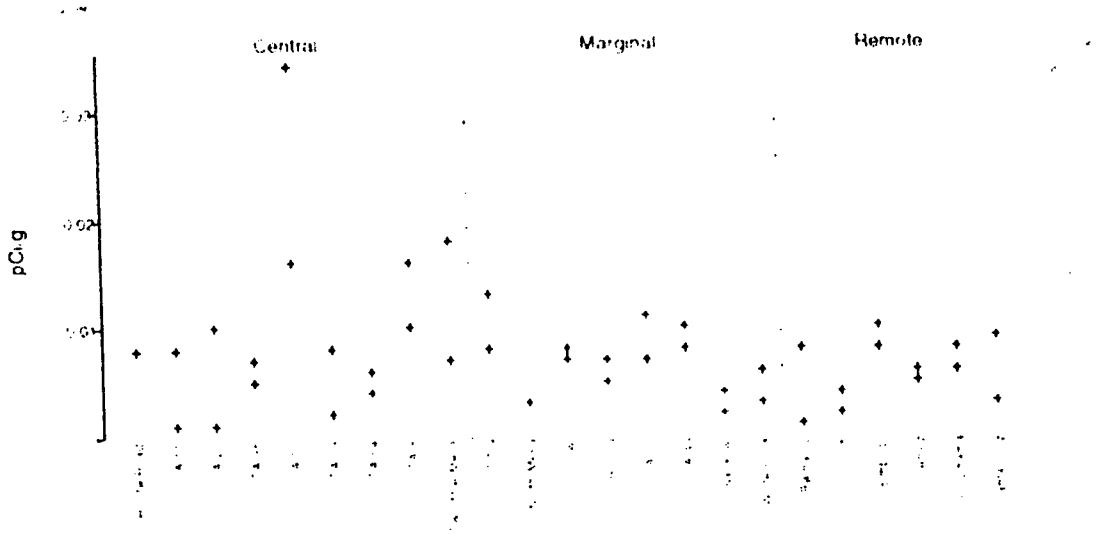


Table A-6. Americium-241 (pCi/g)

	1994	1995
Bernalillo	0.001	0.008
Cochiti	0.004	0.002
Embudo	0.008	0.010
Jemez	0.006	0.005
North Mesa	0.003	
Otowi	0.008	0.007
R-Site Road	0.008	
Rendija Canyon	0.005	0.007
Rio Chama	0.006	0.008
Santa Cruz	0.003	0.009
TA-11	0.001	0.008
TA-21	0.001	0.010
TA-33	0.011	0.007
TA-36	0.005	0.007
TA-49	0.008	0.010
TA-50	0.016	0.034
TA-53	0.002	0.008
TA-54	0.004	0.006
TA-8	0.010	0.016
Test Well DT-9	0.013	0.008
Tsankawi	0.002	0.004
Two Mile Mesa	0.018	0.007
White Rock	0.003	0.006

Figure A.6. Americium-241 by Station



APPENDIX B. STATISTICAL METHODS

This appendix discusses the statistical methods used in Section 4, where local background data sets are proposed and summarized.

Distributional Assumptions

All parametric statistical calculations (i.e., calculations of means, standard deviations, coefficients of variability, and (.95,.95) UTLs) reported in Section 4 are based on the assumption that the populations from which the local background data sets are drawn are lognormally distributed. A random variable X is said to be lognormally distributed if the random variable Y=log(X) is normally distributed. The lognormal distribution, supported on the positive real numbers (that is, $Pr(X \leq 0)=0$ if X is a lognormally distributed random variable) and skewed to the right, is frequently found to describe concentration data quite well

Section 4 includes lognormal probability plots for each of the selected local background data sets. When data from a lognormal distribution are plotted in this way, they should fall approximately along a straight line. Thus the lognormality of the local background distributions can be assessed informally by inspection of these plots.

Some problems are visible in some of these plots For example, quantization effects are very obvious for some of the radionuclides because many of them are being measured near the limits of the analytical methods used. Tritium, for example, is reported to the nearest 0.1 pCi/mL, and more than half of the observations are below 0.5 pCi/mL. The estimated measurement errors reported by Fresquez et al. (1996) are typically 0.2 - 0.4 pCi/mL, so there is certainly no point in reporting results to more than one significant digit, but the result, of course, is not a continuous lognormal distribution.

Even when the measured values are generally within the range of the analytical method used, some problems are often observed in the lower tails. For example, the lognormal probability plot for ^{137}Cs in Section 4.3 (Figure 7) shows that the observations below 0.05 pCi/g are smaller than would be expected if they came from the same lognormal distribution as the rest of the data As noted in Section 4.3, one possible reason for this is that these data are not reliable, since the standard deviation of the measurement errors is at least 0.03 pCi/g. This part of the data is not fit by the estimated lognormal distribution represented by the dashed line in Figure 7, which however provides a reasonable approximation to the bulk of the data. If these observations had been allowed to influence the estimate, then the estimate of the mean would have been slightly smaller, but the estimate of the standard deviation and hence the (.95, 95) UTL would have been much larger. The result would be a serious distortion of what the data are telling us about the local background. Robust estimation methods, described below, were used to minimize such problems.

Another possibility would be to define a more reasonable practical detection limit (on the order of 0.03 pCi/g for 137Cs, for example) and treat all measurements reported below this limit as non detects. This approach would give similar results, but estimates derived in this way are quite sensitive to the choice of the detection limit Methods for determining statistically defensible detection limits exist, but no such detection limits have been provided with the ESP data

Treatment of Zeros and Negative Values

Appendix A shows that there are zero and even negative reported observations for most of the radionuclides Negative values are sometimes reported because the reduction of the raw data requires that an instrumental background value be subtracted If the isotope being measured is

present only at levels that are effectively below the instrumental detection limit for the analytical method being used, this subtraction can result in a negative result The instrumental detection limit is generally much lower than the true method detection level at which results can be reliable quantified. As a result, even many of the reported positive results are statistically indistinguishable from zero, as indicated by reported errors (usually "two-sigma" errors) that are larger than the reported result

Zero and negative values do not need to be modified if the data are being used directly in two-sample tests comparing background and site distributions However, for the statistical calculations reported in Section 4, many of which are based on the assumption of lognormality, they must be replaced by positive values For these data, the choice of 0 5 times the minimum reported positive value is generally satisfactory (For data that are "censored" at a detection level, the use of 0.5 times the detection level is frequently recommended, but radionuclide data are not censored, and consistently calculated method detection levels or MDAs are not reported for the ESP data) Because robust estimation methods that minimize the effect of data in the tails of the distribution are being used, this choice actually has little effect on parameter estimates Its effects are primarily visible in the appearance of plots such as the lognormal probability plots in Section 4 and the point plots in Appendix A In both cases, values which have been substituted are displayed using a different symbol so that their presence is immediately obvious

Parameter Estimation

The estimates of the means and variances of the local background distributions that are reported in Section 4 are computed using the equations for minimum variance unbiased (MVU) estimation for lognormal populations described by Gilbert (1987) Section 13 1 1 The MVU estimate of the mean μ of a lognormally distributed random variable X is related to the estimates of the mean ν and the variance τ^2 of the random variable Y $\log(X)$ (where "log" denotes the natural logarithm, i e., logarithms to the base e) by

$$\hat{\mu} = \{\exp(\nu)\}\Psi_n\left(\frac{\tau^2}{2}\right) \qquad (B\ 1)$$

where n is the sample size and $\Psi_n(t)$ is a function defined in Section 13 1 1 of Gilbert (1987)

The MVU estimate of the variance σ^2 of X is given by

$$\hat{\sigma}^2 = \exp(2\nu)\{\Psi_n(2\tau^2) - \Psi_n\left(\frac{(n-2)\tau^2}{n-1}\right)\} \qquad (B\ 2)$$

The sample mean and variance of the logged data are used for ν and τ^2 in Eqs (B 1) and (B 2) However these conventional estimates are unsatisfactory for our data primarily because of the problems in the lower tails of the distributions described above Therefore a robust estimation method M-estimation is used to provide estimates of ν and τ^2

An M-estimate of location ν is the [illegible] of the equation

$$\sum \psi\left(\frac{Y_i - \nu}{\tau}\right) = 0 \qquad (B\ 3)$$

where the choice of the weighting function Θ is described below. When the scale parameter τ is also unknown, as is the case in all applications in Section 4, then Eq. (B.3) must be solved simultaneously with

$$\sum \Theta\left(\left(\frac{y - \nu}{\tau}\right)^2\right) = \beta , \tag{B.4}$$

where the correct choice for β depends on Θ. (See Huber's "Proposal 2", Huber 1981.) This system of two equations must usually be solved iteratively.

One common choice for $\Theta(t)$ is the "Huber estimator"

$$\Theta_1(t) = \begin{cases} t & \text{abs}(t) \le c \\ c\ \text{sgn}(t) & \text{abs}(t) > c \end{cases}$$

where c is a "tuning constant" chosen to optimize tradeoffs between minimizing the effects of outliers and optimizing the performance of the estimator in the normal case. If $c=\infty$, then the algorithm described by Eqs. (S-3, 4) produces the usual least squares estimates of ν and τ, which of course are the most efficient unbiased estimators for the normal case but are extremely sensitive to deviations in normality. The "bisquare" $\Theta(t)$ is

$$\Theta_2(t) = \begin{cases} \frac{1}{6}\left(c^2 - t^2\right)^2 & \text{if abs}(t) \le c \\ 0 & \text{if abs}(t) > c \end{cases}$$

Thus, the Huber estimator down-weights outliers and the bisquare estimator entirely ignores data that are extreme outliers. The version of the algorithm used in Section 4 first performs two iterations with the Huber function Θ, followed by four iterations with the bisquare function. (Calculations were performed using the function **robloc** in Splus™, a statistical computer package.) Extreme outliers, including a good many in the lower tail of the distribution that would have influenced the estimate of τ and the (.05, .95) UTLs upwards as noted above, were thus ignored by the final estimator.

Upper tolerance limits

Upper tolerance limits (UTLs) are calculated using the parametric method described in Section 11.3 of Gilbert (1987). For the lognormally distributed variables considered in Section 4, the UTLs take the form

$$\text{UTL}(p,1-\alpha) = \exp(\hat{\mu} + K'_{1-\alpha,p}\,\hat{\sigma})$$

where $K'_{1-\alpha,p}$ is a tabled factor that depends on the sample size (n), the quantile being estimated (p) and the level of confidence desired ($100(1-\alpha)\%$). In the limit as $n \to \infty$, $K'_{1-\alpha,p} = Z_p$, where Z_p is the $100p^{th}$ percentile of the standard normal distribution, but for finite n, $K'_{1-\alpha,p} > Z_p$. $K'_{1-\alpha,p}$ is tabled for $p = .95$, $100(1-\alpha)\% = 95\%$, and selected values of n in Appendix A of Gilbert (1987). Linear interpolation in the table has been used when necessary to find values for the actual sample sizes available.