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DISSERTATION

RADIONUCLIDES IN SOIL
AROUND THE ROCKY FLATS ENVIRONMENTAL TECHNOLOGY SITE
JEFFERSON COUNTY, COLORADO

Submitted by

Scott E. Hulse

Radiological Health Sciences Department

In partial fulfillment of the requirements

For the Degree of Doctor of Philosophy

Colorado State University

Fort Collins, CO

Spring, 2002

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WE HEREBY RECOMMEND THAT THE DISSERTATION PREPARED UNDER OUR SUPERVISON BY SCOTT E. HULSE ENTITLED RADIONUCLIDES IN SOIL AROUND THE ROCKY FLATS ENVIRONMENTAL TECHNOLOGY SITE JEFFERSON COUNTY, COLORADO BE ACCEPTED AS FULFILLING IN PART REQUIREMENTS FOR THE DEGREE OF DOCTOR OF PHILOSOPHY.

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ABSTRACT OF DISSERTATION

RADIONUCLIDES IN SOIL AROUND THE ROCKY FLATS ENVIRONMENTAL TECHNOLOGY SITE JEFFERSON COUNTY, COLORADO

Plutonium was released into the environment around Rocky Flats during routine operations, several building fires and remediation efforts at the 903 Barrel Storage Area after barrels containing contaminated oil leaked onto the soil surface. Soil concentrations of naturally occurring and anthropogenic radionuclides were therefore characterized. Mean soil concentrations of naturally occurring 232Th, 40K and 226Ra were 96 Bq kg-1, 720 Bq kg-1 and 63 Bq kg-1, respectively, around Rocky Flats and along Colorado's front range corridor. Rocky Flats is along regional concentration gradients that decrease at a mean rate of 4 Bq kg-1 km-1 for 40K and 0.4 Bq kg-1 km-1 for 232Th from Colorado Springs north to Fort Collins, CO. Variability in soil concentrations of 232Th, 226Ra and 40K is related to soil type, unconsolidated surficial deposits of alluvium and other debris and bedrock. Surface soil concentrations in the top 3 cm of soil and depositions of 241Am decreased in an easterly direction from more than 5.3 kBq kg-1 and 170 kBq m-2 respectively near the 903 pad to 1.3 Bq kg-1 and 0.8 + 0.7 kBq m-2 at distances more than 7 km from the 903 pad at Rocky Flats. Surface deposits of 137Cs from global fallout in the top 3 cm of soil and depositions averaged 0.12 kBq kg-1 and 4.7 kBq m-2 respectively. These deposits were unevenly distributed around Rocky Flats and at regional background locations. Deviations from the uniform exponential rate at which soil concentrations of 137Cs typically decreased with depth, -0.25 cm-1 at undisturbed sites, enabled us to determine that about 10% of sample sites had been disturbed by erosion, tillage or other factors. Soil concentrations of 241Am decreased with depth at a similar mean rate of –0.22 cm-1 close to the 903 pad where measurements were robust. The mean rate at which 239,240Pu decreased with depth throughout the study area was about the same, -0.23 cm-1. The ratio between soil depositions of 239,240Pu determined using radiochemical analyses and 241Am determined in-situ for locations close to the 903 pad was 5.4, which is about the same as the ratio of 5.5 between soil concentrations of 239,240Pu and 241Am in the top 3 cm of soil that was estimated from laboratory analyses.

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INTRODUCTION

The Radioecology Group at Colorado State University (CSU) has continued its effort to quantitatively assess human and ecological risk attributable to releases of radionuclides to the environment around Rocky Flats, Jefferson County, Colorado with an investigation consisting of three tasks:

- Characterize and quantify distributions of radionuclides in soil and biota around Rocky Flats,
- Develop an analytic model to predict doses and risks from radionuclides in the environment around Rocky Flats, and
- Implement a bioassay program to validate the analytic model.

The purpose of CSU's research, which began during the 1970s, is to independently assess health risks to the public from radionuclide releases from Rocky Flats to the environment. The research described in this dissertation, which is included in the first task to characterize the distribution and transport of radionuclides in CSU's risk assessment effort, is divided into three parts:

- Characterization of the distribution of naturally occurring ^{232}Th, ^{226}Ra and ^{40}K in soil around Rocky Flats and along Colorado's Front Range corridor,
- Characterization of the distribution of ^{241}Am and ^{137}Cs using results from laboratory analyses of soil samples with gamma spectroscopy, and comparison with distributions of ^{239,240}Pu in soil around Rocky Flats, and

- Comparison of soil concentrations and deposition of ^{241}Am and ^{137}Cs estimated from in-situ gamma spectroscopy measurements with concentrations and depositions estimated from laboratory analyses.

The first part of this dissertation describes research that characterized spatial and depth distributions of naturally occurring ^{232}Th, ^{226}Ra and ^{40}K in soil around Rocky Flats (Hulse et al. 2000). Natural radiation is the major source of human exposure and so soil distributions of ^{232}Th, ^{226}Ra and ^{40}K provide a basis for future comparisons with exposures to anthropogenic radiation.

Research described in the second part of this dissertation had two objectives (Hulse et al. 1999). The first was to compare soil concentrations, depth distributions and depositions of ^{241}Am estimated from laboratory gamma spectroscopy measurements with concentrations and depositions of ^{239,240}Pu determined from radiochemical analyses reported by Webb et al. (1994). The purpose of this comparison was to determine if soil concentrations of ^{241}Am measurements can be used to predict concentrations ^{239,240}Pu since Krey and Krajewski (1972) reported that the initial ratio of plutonium isotopes was constant in weapons grade plutonium used at Rocky Flats. The second objective was to determine how similar depth distributions of ^{137}Cs from global fallout were in soil profiles at different sample locations. The idea was that the total absorption peak from ^{137}Cs is relatively easy to measure and that diminished peaks could provide a way to rapidly identify sites where soil profiles have been altered by erosion or anthropogenic activity. In addition, ^{137}Cs in global fallout was deposited at about the same time as the ^{241}Am and ^{239,240}Pu released from Rocky Flats. Therefore, if it could be demonstrated that ^{137}Cs, ^{241}Am and ^{239,240}Pu have quantitatively similar behavior in soil around Rocky Flats, soil concentrations of these actinides could be normalized to ^{137}Cs to minimize effects from local variations in deposition and transport processes that influenced patterns of deposition.

The third part of this dissertation describes research that compared soil depositions of ^{137}Cs and ^{241}Am estimated from in-situ gamma spectroscopy measurements with depositions estimated from laboratory analyses. The purpose of this research was to determine the degree to which soil concentrations and deposition estimates made using in-situ measurements are correlated with concentrations and depositions estimated from laboratory analyses. Use of in-situ ^{137}Cs to identify sites affected by soil disturbance was another aspect of this research.

Research described in this dissertation had specific objectives; however, results undoubtedly have other applications. In addition to answering questions about historical operations at the Rocky Flats plant, results from this research may be useful during the current public debate about action levels and future land use for Rocky Flats.

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CONCENTRATIONS OF ^{232}Th, ^{226}Ra, AND ^{40}K IN SOIL AROUND ROCKY FLATS AND ALONG COLORADO'S FRONT RANGE CORRIDOR

BACKGROUND

Soil concentrations of ^{232}Th, ^{226}Ra and ^{40}K were measured during recent studies characterizing spatial distributions of ^{239,240}Pu (Webb et al., 1997), and ^{137}Cs and ^{241}Am (Hulse et al., 1999) in soil around the Rocky Flats Environmental Technology Site (Rocky Flats) and at regional locations along the Front Range urban corridor. The corridor, extending northward from Pueblo to Fort Collins along Interstate Highway I-25 in central Colorado (Fig. 1), is an area where the population is rapidly increasing. Components for nuclear weapons were manufactured at Rocky Flats between 1953 and 1989. These measurements were made because naturally occurring radionuclides from the ^{232}Th and ^{238}U decay series are major sources of human exposure to radiation, as is ^{40}K (NCRP, 1992). This is as true for populations living around Rocky Flats as it is for populations in areas unaffected by nuclear facilities. Natural radionuclide levels should be quantified as part of any evaluation of potential health effects from operations at these facilities.

ChemRisk (1994) reported that combined losses of thorium in airborne effluents from Rocky Flats were less than 32 kg during the life of plant operations. They indicated that losses consisted primarily of ^{232}Th ($T_{1/2}$ = 1.405 X 10^{10} y) with minor 'amounts' of ^{228}Th ($T_{1/2}$ = 1.9131 y) and that these were normal operational losses during metalworking processes. ChemRisk (1994) also reported that airborne releases of depleted and enriched uranium occurred during routine operations at Rocky Flats between 1953 and 1977. Litaor (1995) reported that ^{238}U was released from the 903 pad, which has been described by Krey and Hardy (1970), after cutting oil leaked from corroded drums. Litaor also described releases from the mound site where drums containing enriched and depleted uranium were buried, a pit where oil contaminated with uranium was burned, and several trenches where uranium contaminated oil and solid uranium wastes were buried. In addition, Ripple et al. (1996) estimated that 270 MBq of enriched and 560 MBq of depleted uranium were released during plant operations at Rocky Flats from 1953 through 1989. Depleted uranium used at Rocky Flats contained virtually no ^{226}Ra and therefore had no effect on soil inventories of ^{226}Ra.

Numerous geologic and pedogenic factors probably influenced natural concentrations of ^{232}Th, ^{238}U, ^{226}Ra and ^{40}K in soil around Rocky Flats. These factors included mineralogy of parent material that was altered by weathering. Although the local geology is fairly well known, relationships between natural concentrations of primordial radionuclides and soil type, or surficial and bedrock geology, are poorly understood. The veneer of sediments around Rocky Flats was formed by coalescing fans of alluvium made up of debris from the interior of Colorado's Front Range. Radionuclide concentrations in debris changed as the Rocky Mountains were uplifted and more deeply buried rocks with mineral assemblages different from those exposed earlier were uncovered by erosion. As a result, soil concentrations of ^{232}Th, ^{238}U, ^{226}Ra and ^{40}K should be expected to vary somewhat in these alluvial fans, which were deposited unconformably on erosional surfaces above Cretaceous carbonates, siltstones and sandstones in the Arapahoe, Laramie and the Fox Hills Formations beneath Rocky Flats. Malde (1955), Van Horn (1957; 1972; 1976), Spencer (1961), Hurr (1976), Shroba and Carrara (1994) and EG&G (1995a; 1995b) and others have described underlying sedimentary rocks as well as other geologic features, hydrogeology and geography that could have influenced distributions of ^{232}Th, ^{226}Ra and ^{40}K around Rocky Flats.

With the exception of airborne, soil-sample, and in-situ gamma spectroscopy measurements by Mallory and Bird (1953), Brown and Stehle (1956), EG&G (1974) and Boyns (1982; 1990) there have been few measurements of ^{232}Th, ^{226}Ra and ^{40}K in soil around Rocky Flats before this study. There have been several regional studies characterizing concentrations of ^{232}Th, ^{238}U, and ^{40}K in soil east of Colorado's Front Range. Some of these studies assumed a level of equilibrium between soil concentrations of ^{226}Ra and ^{238}U. These include studies by Grossman and Smith (1956) during a program to evaluate economic mineralization, Poponoe (1965; 1966) during the Airborne Radiological Measurement Surveys (ARMS), and EG&G Geometrics (1978; 1979) during the National Uranium Resource Evaluation (NURE) Program. Concentrations of ^{232}Th, uranium (adjusted for relative levels of equilibrium with ^{226}Ra in some cases) and ^{40}K in regional soil samples collected along Colorado's Front Range have been reported by Myrick et al. (1983), the United States Department of Energy (USDOE, 1993; 1995) and the United States Geological Survey (Wells, 1967; Bunker and Bush, 1968; Shacklette and Boerngen, 1984; Hoffman and Buttleman, 1994). Soil concentrations of ^{40}K can also be inferred from concentrations of K_2O along Colorado's Front Range

Urban Corridor (Severs and Tourtelot, 1994) because the natural abundance of ^{40}K (0.0118%) is nearly constant (Bunker and Bush, 1967).

Specific objectives of this study were to: 1) measure soil concentrations of ^{232}Th, ^{226}Ra and ^{40}K at sites east of Colorado's Front Range to determine the regional distribution of these radionuclides, 2) determine how concentrations of ^{232}Th, ^{226}Ra and ^{40}K are distributed in soil around Rocky Flats, 3) compare ratios between soil concentrations of ^{226}Ra, that originated from decay of natural ^{238}U, and ^{232}Th at sample locations around Rocky Flats with similar ratios for regional samples to determine if effects from low-level releases of ^{232}Th can be detected, and 4) discover correlations between soil concentrations of ^{232}Th, ^{226}Ra and ^{40}K, and soil type, surficial and bedrock geology, to identify pedogenic factors that affected spatial distributions of these radionuclides.

MATERIALS AND METHODS

Area of Investigation and Sample Locations

Rocky Flats, located 20 km northwest of Denver, CO, includes an industrial area of about 1.5 km^2 surrounded by a relatively undisturbed 26.5 km^2 buffer zone between the plant and neighboring locales. During 1993 and 1994, 1,309 soil samples were collected from 18 on-site and 24 off-site locations along four radial transects, each approximately 20 km long (Fig. 1). An effort was made to sample undisturbed areas. Six to thirteen 10 m by 10 m macroplots spaced at logarithmically increasing intervals along transects were sampled to assure more detailed information near Rocky Flats. Bearings of the transects, 60°, 90°, 120° and 150° relative to true North from the 903 pad, were selected based on directions of dominate strong winds. Regional concentration levels were estimated from samples taken in 20 communities between Colorado Springs and Fort Collins believed to be well outside the area that was measurably affected by contaminants from Rocky Flats.

Soil Sampling

Three types of soil samples were taken from randomly selected 1 m^2 microplots within each macroplot. The first was a scrape from the top 3 mm of soil over an area 1.0 m by 0.5 m. Terry (1994) has described this sampling method, used to characterize the soil fraction most susceptible to resuspension. Samples from 3 cm thick horizontal layers of soil were also taken from an area 25 cm long by 15 cm wide to a depth of 21 cm at each on-site macroplot and one of four microplots in each off-site macroplot. Composite samples from 0 to 21 cm soil columns were taken at three other microplots in each off-site macroplot and at regional locations. Rocks were removed from soil samples and weighed in the field. Rock volume in samples was estimated using an average density for crystalline and metasedimentary rocks of 2.65 g cm^{-3} (Carmichael, 1989). In-situ soil density estimates were made following protocols described in ASTM Method D2167-84 (1984).

Sample Preparation

Soil samples were air dried at room temperature, clods were crushed with a mortar and pestle, and rocks, plant roots and other large debris were removed using a sieve with 4.75 mm openings. The sieved soil was placed in steel cans and oven dried at 110° C for 24 hours, after which samples were mechanically sieved for 30 minutes using an ASTM #10 sieve with 1.65 mm openings. Dried samples passing the sieve were placed in seamless steel cans 10 cm in diameter and 6 cm high. Cans were sealed with silicone to assure an airtight seal, using a method that has been thoroughly tested at Colorado State University. Samples were then allowed to equilibrate for at least 30 days to bring short-lived natural radionuclides into equilibrium before gamma spectroscopy measurements were made.

Counting Standards

Secondary counting standards described by Rope and Whicker (1980), that were prepared from uranium mill tailings, were used to calibrate the detector response to ^{226}Ra. Masses of tailings in these standards ranged from 50 to 350 g to account for differences of geometry in cans containing field samples that were partially filled with soil. Rope and Whicker reported that concentrations of ^{226}Ra in their secondary standards were estimated by comparison with a soil standard spiked with an aqueous solution of ^{226}Ra from the National Institute of Standards and Technology Standard Reference Material 4950-D (NIST, Gaithersburg, MD) that was packaged in the same geometry as the mill tailings. Soil standards for ^{232}Th were prepared from ground silica sand with a particle size less than 2 mm that had been spiked during 1982 with a ^{232}Th solution that was traceable to NIST standards. Concentrations of ^{228}Ra in these standards had reached 77% of secular equilibrium with ^{232}Th. These standards were also packaged in the same geometry as field samples. Masses of sand in ^{232}Th standards ranged from 50 to 350 g. Calibration factors for both ^{226}Ra and ^{232}Th standards were verified by predicting concentrations of ^{226}Ra and ^{232}Th in a 160 g aliquot of the NIST Rocky Flats Soil #1 packaged with the same geometry as field samples. Results were compared with results for NIST Rocky Flats Soil #1 Standard Reference Material 4353. The total content of ^{226}Ra in the aliquot of Rocky Flats Soil #1 was reported to be 7.0 ± 0.5 Bq. The predicted content after calibrating with Rope and Whicker's standards was 6.0 ± 0.5 Bq. The total content of ^{232}Th was reported to be 11.3 ± 0.6 Bq. The predicted content was 12.8 ± 1.3 Bq.

The ^{40}K content in four sets of soil standards used during this study were estimated by comparison with the content of ^{40}K in the 160 g aliquot of the NIST Rocky Flats Soil #1 (117 ± 11 Bq). Concentrations of ^{40}K in these standards ranged from 300 to 1,000 Bq kg^{-1}. Soil masses in the different sets of standards ranged from 50 to 400 g.

Counting, Data Analysis and Statistical Methods

Samples were counted for at least 100 minutes on an HPGe (96% efficiency relative to a 3 inch by 3 inch NaI detector at 1.33 MeV) or a GeLi detector (30%

efficiency relative to a 3 inch by 3 inch NaI detector at 1.33 MeV). The Levenberg-Marquardt optimization algorithm (Marquardt, 1963) in SPSS' PeakFit™ (Version 4.0) software was used to obtain a least-squares fit of a Gaussian distribution to each total absorption peak. After removing counter background effects from the spectra the Gaussian provided the estimate for the total absorption peak area.

Soil concentrations of ^{226}Ra were measured indirectly by averaging concentrations of ^{214}Bi estimated from the areas under total absorption peaks at 609 and 1,120 keV. Estimates for ^{232}Th concentrations were made using the assumption, based on a review by Harbottle and Evans (1997), that ^{232}Th in undisturbed soils is generally in secular equilibrium with its entire decay chain including ^{228}Ac, ^{212}Pb and ^{208}Tl. Concentrations of ^{232}Th were measured indirectly by averaging concentrations of ^{208}Tl estimated from the areas under total absorption peaks at 583 and 2,614 keV and the concentration of ^{228}Ac estimated from the area under a total absorption peak at 911 keV. These estimates were made after accounting for disequilibrium between ^{228}Ra and ^{232}Th in standards. We assumed that ^{214}Bi was in secular equilibrium with ^{226}Ra, and that ^{208}Tl and ^{228}Ac were in secular equilibrium with ^{228}Ra after holding the sealed samples for more than 30 days before counting. Soil concentrations of ^{40}K were estimated directly from counts under the 1,461 keV total absorption peak. We attempted to estimate soil concentrations of ^{238}U indirectly from activity concentrations of ^{234m}Pa and ^{234}Th, and to directly estimate concentrations of ^{235}U; however, low photon emission rates and interference from other more abundant radionuclides made these estimates impractical.

A method originally proposed by Currie (1968) with modifications recommended in ANSI Standard Method N13.30 (ANSI, 1989) was used to estimate lower limits of detection for a 250 g sample, which was the most prevalent sample size. These limits were 6 Bq for ^{40}K, 0.5 Bq for ^{226}Ra and 0.3 Bq for ^{232}Th when measurements were made with the HPGe detector system. Lower limits of detection were 15 Bq for ^{40}K, 1.4 Bq for ^{226}Ra and 0.8 Bq for ^{232}Th when the GeLi detector was used. The critical level described by Currie is the level used to distinguish between samples that contain an analyte and samples in which any analyte concentration is below the minimum detectable amount. Rather than use a blank paired with each sample as described by Currie, one blank consisting of an empty steel can like those used for soil samples was counted for each ten samples because variability between blank counts was negligible throughout this investigation.

Currie's critical level enables a decision as to whether a sample contains an analyte; however, the critical level does not provide information about whether the analyte concentration is in excess of normal background levels. A second statistic, the decision level, estimated with a method described by Borak and Kirchner (1995), provided a benchmark to distinguish environmental samples containing background concentration levels of ^{232}Th and ^{226}Ra from those containing elevated concentrations. Decision levels, estimated after setting α levels for Type I errors to 0.20, were 130 Bq kg^{-1} for ^{232}Th and 96 Bq kg^{-1} for ^{226}Ra. Soil concentrations for ^{232}Th and ^{226}Ra at 20 regional locations and sample locations more than 7 km from the 903 pad at the east ends of the 60°, 90° and 120° transects were used to estimate decision levels. These samples were included to assure a sufficiently large statistical sample (n ~ 30) as recommended by Borak and Kirchner (1995).

When the assumptions that independent populations had normal distributions with common variance were valid, t-tests were used to compare population means. Means for independent samples were compared with two-sample t-tests; for example, comparisons of on-site and off-site samples. Means for matched samples were compared with paired t-tests as was the case for split samples or recounts for the same samples. The Anderson-Darling test (Ryan and Joiner, 1976) was used to test the assumption that populations had normal distributions. Results of two sample t-tests were corroborated using the non-parametric Mann-Whitney two sample rank test and confidence interval and the non-parametric Kruskal-Wallis test (Ott, 1988). The 0.05 level was used as the measure for significance of p-values. These p-values provide estimates for the probability of observing an outcome that was more contradictory of a null hypothesis than the result that was actually observed; for example, when testing whether the slopes of regression lines or differences between two means were significantly different from zero. Differences between two-sample t-tests and non-parametric tests were generally negligible so only results for t-tests have been reported. Partial correlation coefficients (pcc) were used to characterize the relationship between soil concentrations of two radionuclides while holding the concentration of the third constant. Each of the two concentrations that were to be compared was regressed against the concentration that was held constant and residuals were computed. Pearson Correlation Coefficients for the two sets of residuals provided pcc estimates for the correlation between respective soil concentrations while controlling for the third. All statistics were calculated using algorithms in the Minitab™ (Version 11.12) statistical software package.

Quality Assurance and Quality Control

Stone et al. (1994) has described quality assurance and quality control measures intended to assure representative and reproducible field samples. Systematic counts of an empty can were made before counting each batch of 11 field samples to estimate and monitor counter background. One hundred and nineteen randomly selected samples were recounted to evaluate variability within samples caused by changes of geometry in containers that were partly filled with soil. Sample volumes varied because a constant volume was sampled with a fixed geometry; however, the amount of soil depended on the volume of rocks that were removed when samples were sieved. Mean concentrations of ^{40}K, ^{226}Ra and ^{232}Th for both the original and recounted populations were not significantly different. Seventy-one replicates, prepared by splitting randomly chosen samples, were analyzed in the blind to assess variability between samples taken from identical locations. Differences between concentration estimates for ^{40}K, ^{226}Ra and ^{232}Th in split samples were also insignificant.

RESULTS AND DISCUSSION

Regional Levels

Mean soil concentrations of ^{232}Th, ^{226}Ra and ^{40}K for 20 regional locations (Table 1) sampled during this study that were beyond any probable influence from Rocky Flats had normal distributions. The USDOE (1995) reported that soil concentrations of both

^{226}Ra and ^{238}U in samples from regional locations that they sampled east of Colorado's Front Range had approximately lognormal distributions.

The range of our concentration estimates for ^{232}Th (Table 2) was higher than the mean concentration for 200 widely spaced samples from across North America (Lowder et al., 1964; NCRP, 1991; 1992). We believe this difference occurred because parent material for soil along Colorado's Front Range is made up of Precambrian igneous rocks and sedimentary rocks like those under the Eastern Plains that were metamorphosed during uplift and intrusion by Tertiary granites. Phair and Gottfried (1964) reported that area weighted average concentrations of ^{238}U and ^{232}Th in these Tertiary granites were about 60 Bq kg^{-1} and 90 Bq kg^{-1}, respectively (^{238}U:^{232}Th = 0.67). The NCRP (1992) has reported that concentrations of ^{238}U and ^{232}Th in granites (crustal averages) were 40 Bq kg^{-1} and 70 Bq kg^{-1}, and that concentrations in sedimentary rocks ranged from <10 to 40 Bq kg^{-1} and <8 to 50 Bq kg^{-1}, respectively. The ratio between mean crustal concentrations of ^{238}U and ^{232}Th in granites (0.57) is, coincidentally, the same as the ratio estimated from concentrations of ^{238}U and ^{232}Th certified by NIST (1980) in the Rocky Flats #1 Soil.

The mean concentration of ^{40}K for North America that was reported by Lowder et al. (1964) was in the lower quartile of the range for soil concentrations of ^{40}K measured during this study. Tertiary granites in the Front Range Mineral Belt west of Rocky Flats also contained higher concentrations of potassium feldspar than intruded rocks. In addition, potassium was concentrated in wall rocks surrounding granites as a result of alteration during the intrusion process (Lovering and Goddard, 1950; Tooker, 1963, Phair and Jenkins, 1975). Debris and sediments from these rocks were therefore expected to contain relatively high concentrations of ^{40}K since the natural abundance is nearly constant (Bunker and Bush, 1967). The NCRP (1992) has reported that the average concentration of ^{40}K in granite is 1000 Bq kg^{-1} or more while concentrations in sedimentary rocks range from 70 to 900 Bq kg^{-1}.

Surface Distributions

Partial correlation coefficients were used to compare spatial distributions of ^{232}Th, ^{226}Ra and ^{40}K in soil at different locations. The pccs between northerly Universal Transverse Mercator (UTM) coordinates and soil concentrations, -0.44 for ^{232}Th, -0.37 for ^{226}Ra and -0.54 for ^{40}K, estimated while holding easterly UTM coordinates constant, indicate that soil concentrations decrease in a northerly direction. Levels of ^{40}K were found to decrease at about 4 Bq kg^{-1} km^{-1} ($p = 0.007$) between Colorado Springs and Fort Collins, CO by regressing soil concentrations on northerly UTM coordinates. Levels of ^{232}Th decreased at about 0.4 Bq kg^{-1} km^{-1} ($p = .004$) between the same locations. Estimates for the rate of change in soil concentrations of ^{226}Ra from south to north lacked significance ($p = 0.6$). Detection of decreasing northerly gradients in soil concentrations of ^{40}K and ^{232}Th may have been possible because sampled locations were distributed across 210 km between Colorado Springs and Fort Collins. Partial correlation coefficients between soil concentrations of ^{232}Th (0.15), ^{226}Ra (0.23) and ^{40}K (0.35) and easterly UTM coordinates, estimated while holding northerly UTM coordinates constant, were weak. This may have been because the distribution of samples was not optimized to test for an easterly gradient: samples were taken across an easterly UTM interval of about 20 km.

Mean soil concentrations for ^{40}K were expected to decrease with distance from massive features like the Pikes Peak batholith exposed to the south of Denver between Colorado Springs and Sedalia, which contains potassium rich granites (Barker et al., 1975; Wobus, 1976). A similar trend occurred in soil concentrations of K_2O reported by Severson and Tourtelot (1994) along Colorado's Front Range Corridor between Colorado Springs and Fort Collins. There is also a slight decrease in soil concentrations of total potassium in a northwesterly direction in eastern Adams County where soil concentrations of ^{40}K range from 600 to 1,200 Bq kg^{-1} (Bunker and Bush, 1968).

We don't know why ^{232}Th concentrations decrease in a northerly direction. There appears to be a weakly decreasing trend in data from pressurized ion chamber measurements between Monument (about 30 km north of Colorado Springs) and Fort Collins that was reported by Stone et al. (1999). However, measurements made by Lowder et al. (1964) indicate that the highest soil concentrations of ^{232}Th occur near Denver and that concentrations of ^{232}Th were lower where their samples were taken in Colorado Springs and Fort Collins.

Concentrations of ^{232}Th in soil from depths of 0 to 3 cm were regressed on the log-transform of distance from the 903 pad to determine if contamination released from Rocky Flats had an effect. The estimated slope of the regression equation lacked significance ($p = 0.3$). Thus, we found no evidence that soil concentrations of ^{232}Th

decrease with distance from Rocky Flats as should be expected if there were significant releases from the site.

A regression procedure similar to that used for ^{232}Th was used to determine that mean concentrations of ^{226}Ra in soil from depths of 0 to 3 cm increased at a significant rate from on-site to off-site locations along transects ($p < 0.001$ for the slope of the regression equation). The mean soil concentration of ^{226}Ra in samples from depths of 0 to 3 cm at regional locations was higher than at off-site transect locations ($p = 0.001$ for a two-sample t-test). Concentrations of ^{226}Ra in soil from depths of 3 to 6 cm ($p = 0.01$), 9 to 12 cm ($p =< 0.001$), 12 to 15 cm ($p = 0.01$) and 18 to 21 cm ($p = 0.02$) at each sample location also increased at significant rates from on-site to off-site locations. With the exception of the 0 to 3 cm interval, concentrations in soil from separate 3 cm depth intervals were not measured at regional locations. The mean concentration of ^{226}Ra in 21 cm soil columns at regional locations was higher than the mean for 0 to 21 cm intervals at on-site locations ($p < 0.001$), however. Differences between concentrations in 21 cm depth intervals along transects at off-site and regional locations were insignificant ($p = 0.98$). We believe that natural uranium is the most likely source of all ^{226}Ra detected around Rocky Flats since we have found no contradictory evidence.

Variations With Depth

Mean soil concentrations for ^{40}K and ^{232}Th increased by about 20% from the surface to a depth of 6 cm and were constant (720 ± 180 and 96 ± 19 Bq kg^{-1} respectively) at greater depths (Fig. 2). There were no significant differences between mean soil concentrations of ^{40}K and ^{232}Th for samples from depths greater than 3 cm at individual on-site, off-site and regional locations. There were only marginal differences between mean concentrations of ^{40}K in surface layers at on-site and off-site locations ($p = 0.02$ for the 0 to 3 mm layer and $p = 0.03$ for the 0 to 3 cm layer). Concentrations of ^{40}K and ^{232}Th within separate 3 cm sample intervals for all locations were therefore pooled to estimate mean rates at which soil concentrations of each radionuclide changed with depth. Soil concentrations of both radionuclides in each depth interval varied by ±15% to ±20%. More than one-half of the variability was attributable to differences in concentrations between locations. Average standard deviations for soil concentrations of ^{40}K and ^{232}Th in samples from all depths between 6 and 21 cm at individual locations were generally less than 3% and 9% respectively.

Although there were significant differences between mean soil concentrations of ^{226}Ra in five of the seven depth intervals as described previously, differences between mean concentrations in 21 cm soil columns were marginal. Soil concentrations of ^{226}Ra in samples from separate 3 cm depth intervals at all locations were therefore also pooled to estimate the mean rate at which soil concentrations of ^{226}Ra changed with depth. Like ^{40}K and ^{232}Th, mean soil concentrations for ^{226}Ra increased from the surface to a depth of about 6 cm (Fig. 2). Soil concentrations remained nearly constant (63 ± 14 Bq kg^{-1}) from 6 to 21 cm. We observed that mean soil concentrations for ^{226}Ra in 3 cm sample intervals varied by ±15% to ±20% and like ^{232}Th, about one-half of the variability was attributable to nearly constant differences between locations. The average standard deviation for soil concentrations of ^{226}Ra in samples from all depths between 6 and 21 cm at individual locations was generally less than 9%.

The ratio between the concentrations of ^{232}Th or ^{226}Ra in plants and soil is significantly less than one. We believe, based on field descriptions of soil characteristics recorded when samples were taken, that there is an inverse relationship between increased concentrations of both ^{232}Th and ^{226}Ra and decreased content of organic matter with depth. We also determined that soil density increased from 0.9 ± 0.2 g cm^{-3} in the 0 to 3 cm depth interval to 1.2 ± 0.2 g cm^{-3} in the 3 to 6 cm interval. The apparent peak in ^{226}Ra concentrations between depths of 3 to 6 cm is probably a statistical aberration without significance.

Ratios Between ^{226}Ra and ^{232}Th

With a few exceptions, we observed no significant differences between the mean ratio of ^{226}Ra and ^{232}Th in soil around Rocky Flats (0.67 ± 0.10). This ratio is the same as the ratio between concentrations of ^{238}U and ^{232}Th reported by Phair and Gottfried (1964) for granite intrusions throughout Colorado's Front Range (0.67). One exception was a site near Leyden close to a location where Gude and McKeown (1952) reported carnotite ($K_2(UO_2)_2(VO_4)_2 \cdot 3H_2O$) on fractures along shallow coal seams. We also observed ratios that were significantly higher than the mean ratio between soil concentrations of ^{238}U and ^{232}Th in samples near streambeds down gradient from uranium deposits.

Although by no means a complete list, nearby uranium deposits include the Schwartzwalder mine 6 km southwest of the 903 pad (Downs and Bird, 1965; Heyse, 1971; Maslyn, 1978; Wallace and Karlson, 1985), and the Aubrey, Ladwig and Ascension properties 14 km south-southwest of Rocky Flats (Sheridan et al., 1967; Ferris and Bennet, 1977; Wallace, 1979). Goldstein (1957) reported uraninite in the Dakota Formation further to the south near Morrison. Derzay and Bird (1976) and Van Horn (1976) described uraniferous rocks in Ralston Creek 7 km south-southwest of the 903 pad. Higher than average concentrations of ^{226}Ra and ^{238}U have also been reported in numerous silica-rich Tertiary porphyry dikes along the eastern margins of Colorado's Front Range throughout the central part of the Front Range Mineral Belt immediately west of Rocky Flats (Sims et al., 1963). There is, in addition, a massive thorium-rich dike near Central City about 30-km southwest of Rocky Flats (Young and Sims, 1961) near the headwaters of Clear Creek and Coal Creek. Clear Creek flows in an easterly direction south of Rocky Flats. Coal Creek, located north of Rocky Flats, also flows to the east. Other natural occurrences of enhanced uranium and thorium deposits around Rocky Flats have been described by Adams and Gude (1953), Merwin (1956), Phair and Gottfried (1964), Childers (1974), Reade (1978), Nelson-Moore et al. (1978), Carpenter et al. (1979; 1995) and Harshman and Adams (1981), among others.

We believe that ratios between soil concentrations of ^{226}Ra and ^{232}Th are not particularly useful for delineating the extent of contamination from operations at Rocky Flats because of the variability associated with naturally enhanced concentrations of these radionuclides. Litaor (1995) came to a similar conclusion after examining ratios between concentrations of uranium isotopes in soil around Rocky Flats.

Soil Concentrations, Geography, Geology and Proximity

The hypothesis that soil concentrations of ^{232}Th, ^{226}Ra and ^{40}K were not significantly different at the various sites that were sampled irrespective of soil type and surficial or bedrock geology was tested with one-way analysis of variance (AOV). Soil types from the study area in which concentrations of ^{232}Th, ^{226}Ra or ^{40}K were significantly different from the overall means for the various radionuclides included the Hargrave fine sandy loam. The sample upon which this observation is based is from a coal mining area near Marshall that has been described by Soister (1974). The relationship between lignite, coal and uranium in the western Great Plains has been described by Denson et al. (1959) and so it is no surprise that this sample contained the highest overall concentrations of ^{226}Ra (106 Bq kg^{-1}) that were detected during this study. Samples of the Valmont and Leyden-Primen-Standley cobbly clay loam, which contained relatively high concentrations of ^{232}Th (130 Bq kg^{-1}) and slightly elevated concentrations of ^{226}Ra (71 Bq kg^{-1}) were taken along the 150° transect near Leyden 5 km south of the 903 pad in another area where coal was mined. Relatively high concentrations of ^{40}K were detected in soil samples from the Denver-Kutch, Nunn and Ulm urban land complex (1100 Bq kg^{-1}). The lowest concentrations of ^{232}Th, ^{226}Ra and ^{40}K were measured in samples from the Samsil shingle complex.

Soil concentrations of both ^{232}Th and ^{226}Ra were significantly higher in soil covering surficial deposits of Piney Creek alluvium along Woman Creek, southeast of Rocky Flats. In addition, concentrations of ^{40}K were higher at one location where residual soil and detritus over the Arapahoe formation was sampled. Soil concentrations of both ^{232}Th and ^{40}K were also relatively high in samples taken over the Pierre Shale at two locations. This observation is based on a comparison of the mean for only two samples; however, the comparison has considerable statistical power since the sample used to estimate the overall mean was much larger (n = 51). We also observed that mean soil concentrations of ^{40}K in all soil types over the Arapahoe formation (n = 14) were lower than the overall mean, an observation that is inconsistent with the higher ^{40}K in surficial deposits of residual soil and detritus over the Arapahoe formation (n = 1).

Soil types at sampling locations were identified using maps published at scales of 1:20,000 and 1:24,000 by the Soil Conservation Service of the United States Department of Agriculture (Sampson and Baber, 1974; Moreland and Moreland, 1975; Price and Amen, 1983). Characteristics of the regolith, which include surficial alluvium and residual detritus, and lithology of bedrock at sampling locations were identified using geologic maps published by the United States Geological Survey at a scale of 1:24,000 (Malde, 1955; Van Horn, 1957; Spencer, 1961; Wrucke and Wilson, 1964; Wells, 1967; Sheridan et al., 1967; Gable, 1968; Scott, 1972; Romero and Hampton, 1972; Van Horn, 1972; Pierce and Schmidt, 1975; Trimble, 1975; Hurr, 1976; Van Horn, 1976; Colton and Anderson, 1977; Machette, 1972; Lindvall, 1978; Lindvall, 1979; Lindvall, 1980; Shroba, 1980; Shroba and Carrara, 1994) and a thesis by Schwochow (1972). Soil concentrations at regional sample locations in surrounding communities within 20 km of Rocky Flats were included with data from transect locations for this analysis; however, data for more distant regional locations near Colorado Springs and Fort Collins were omitted because our data in those areas was sparse.

The strength of correlations between soil concentrations of ^{232}Th, ^{226}Ra and ^{40}K were compared by estimating partial correlation coefficients for two of these radionuclides while controlling the third (Ott, 1988); for example, comparing the correlation between ^{232}Th and ^{226}Ra while controlling ^{40}K. The strongest correlation was between concentrations of ^{226}Ra and ^{232}Th (0.60). Correlations between concentrations of ^{232}Th and ^{40}K (0.44), and ^{226}Ra and ^{40}K (0.40) were weaker. These results are consistent with the idea that ^{238}U, the source of ^{226}Ra after radioactive decay, and ^{232}Th originated from a common host, possibly the granite intrusions scattered throughout Colorado's Front Range. The weaker correlations of ^{226}Ra and ^{232}Th with ^{40}K undoubtedly reflect the ubiquitous distribution of ^{40}K in marine sedimentary rocks underlying the study area as well as in granite intrusions. Differences in the strength of these correlations may also be due to similarities in the geochemical behavior of ^{232}Th and ^{238}U and dissimilarities between the behavior of these and the geochemical behavior of potassium.

CONCLUSIONS

The following conclusions are drawn from this analysis.

- Soil concentrations of ^{232}Th around Rocky Flats range from 40 to 140 Bq kg^{-1} with a mean of 96 Bq kg^{-1}. Soil concentrations of ^{232}Th decrease in a northerly direction from Colorado Springs to Fort Collins at a rate of about 0.4 Bq kg^{-1} km^{-1}. No significant differences were found between mean concentrations of ^{232}Th at sample sites on and around Rocky Flats in soil from depths of 0 to 21 cm.

- Mean soil concentrations of ^{226}Ra at individual locations range from 30 to 110 Bq kg^{-1} with an overall mean of 63 Bq kg^{-1} around Rocky Flats. We found no evidence that mean concentrations of ^{226}Ra differ significantly between on-site and off-site locations in soil from depths of 0 to 21 cm. No spatial patterns in soil concentrations of ^{226}Ra were observed along Colorado's Front Range.

- No significant decreases in soil concentrations of ^{226}Ra or ^{232}Th with distance from the 903 pad at Rocky Flats were found.

- Ratios between soil concentrations of ^{226}Ra and ^{232}Th are nearly constant around Rocky Flats except where there is evidence for naturally enhanced uranium and thorium concentrations. Mean ratios around Rocky Flats are about the same as the ratios for Tertiary granite intrusions in Colorado's Front Range (0.67). Ratios decrease between Colorado Springs and Fort Collins due to the decreasing northerly gradient in soil concentrations of ^{232}Th.

- Mean concentrations of ^{40}K in soil from depths of 0 to 21 cm around Rocky Flats range from about 360 to 1200 Bq kg^{-1} with a mean of 720 Bq kg^{-1}. Soil concentrations of ^{40}K decrease in a northerly direction from Colorado Springs to Fort Collins at about 4 Bq kg^{-1} km^{-1}.

- We found a moderate correlation (pcc = 0.60) between concentrations of ^{226}Ra and ^{232}Th in soil. We also found that there are correlations between ^{232}Th, ^{226}Ra and ^{40}K, and specific soil types, surficial deposits and at least one kind of bedrock, particularly where soil occurs over coal deposits near Marshall and southeast of Rocky Flats near Leyden along the 120° transect.

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Table 1. Mean concentrations of ^{232}Th, ^{226}Ra and ^{40}K in soil from 3 mm surface scrapes, separate 3-cm intervals from the surface to 21 cm and for the entire 21 cm depth interval.

	On-site[1]			Off-site[2]			Regional*		
	n	mean Bq kg^{-1}	std dev Bq kg^{-1}	n	mean Bq kg^{-1}	std dev Bq kg^{-1}	n	Mean Bq kg^{-1}	std dev Bq kg^{-1}
					^{232}Th				
0 to 3 cm	77	88	15	107	91	20	25	90	39
3 to 6 cm	83	94	16	24	100	22	*	*	*
6 to 9 cm	71	96	15	27	98	21	*	*	*
9 to 12 cm	73	95	15	26	100	24	*	*	*
12 to 15 cm	78	97	15	23	98	23	*	*	*
15 to 18 cm	72	97	14	25	100	25	*	*	*
18 to 21 cm	71	95	16	24	103	24	*	*	*
					^{226}Ra				
0 to 3 cm	77	58	11	107	65	15	25	73	30
3 to 6 cm	83	59	15	24	68	13	*	*	*
6 to 9 cm	71	59	12	27	65	14	*	*	*
9 to 12 cm	73	59	13	26	73	13	*	*	*
12 to 15 cm	79	61	12	23	70	13	*	*	*
15 to 18 cm	72	62	14	25	67	13	*	*	*
18 to 21 cm	71	61	15	24	69	14	*	*	*
					^{40}K				
0 to 3 cm	77	710	180	107	660	100	25	760	480
3 to 6 cm	83	720	180	24	680	90	*	*	*
6 to 9 cm	71	730	180	27	690	120	*	*	*
9 to 12 cm	73	750	180	26	700	120	*	*	*
12 to 15 cm	78	740	200	23	680	130	*	*	*
15 to 18 cm	72	750	180	25	690	120	*	*	*
18 to 21 cm	71	730	190	24	680	120	*	*	*

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[1] On-site locations were within the buffer zone at the Rocky Flats Environmental Technology site along the 60°, 90°, 120° and 150° transects.

[2] Off-site locations were outside the Rocky Flats Environmental Technology site buffer zone along the 60°, 90°, 120° and 150° transects.

* Samples were not taken from the specified interval.

Table 2. Summary of some soil concentrations for ^{232}Th, ^{238}U, ^{226}Ra and ^{40}K that have been reported in literature.

Investigators	Location	^{232}Th n	^{232}Th range	^{238}U n	^{238}U range	^{226}Ra n	^{226}Ra range	^{40}K n	^{40}K range
			Bq kg^{-1}		Bq kg^{-1}		Bq kg^{-1}		Bq kg^{-1}
Boyns (1990)	Rocky Flats & vicinity	64	30-86	64	23-52	*	*	64	270-970
Bunker & Bush (1968)	Eastern Adams County, CO	83	30-80	83	10-40	*	*	83	640-1,200[3]
Bush & Abston[4] (1995)	Front Range Corridor, CO	2,010	Trace-3.7 x 10^5	1,631	Trace-3.8 x 10^5	*	*	1,677	Trace- 4,000[5]
Citizens Environmental Sampling Committee (1996)	Rocky Flats & vicinity	*	*	78	41-230	*	*	*	*
Duvall et al[5] (1989)	Front Range Corridor, CO	*	*	Aerial	40-70	*	*	*	*
Duvall et al[14] (1990)	Front Range Corridor, CO	Aerial	20-110	*	*	*	*	Aerial	240-710[11]
Duvall[6] (1995)	Front Range Corridor, CO	Aerial	20-110	Aerial	40-70	*	*	Aerial	240-710[11]
Hoffman & Buttleman[7] (1994)	Front Range Corridor, CO	3,720	Trace-2,100	13,556	Trace-2,800	*	*	3,716	Trace-2,400[11]

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[3] *Estimate made by multiplying potassium concentrations by 0.0118%, the natural abundance for ^{40}K (Bunker and Bush, 1967)*

[4] Combined soil and outcrop sample data from the National Radioelement Data Base

[5] Compilation of gridded data for aerial measurements from the National Uranium Resource Evaluation Program

[6] Compilation of profiles along flight-lines for aerial measurements from the National Uranium Resource Evaluation Program

[7] Analyses of soil and stream sediment collected during the National Uranium Resource Evaluation Program

Table 2. (cont.) Summary of some soil concentrations for ^{232}Th, ^{238}U, ^{226}Ra and ^{40}K that have been reported in literature.

Investigators	Location	^{232}Th		^{238}U		^{226}Ra		^{40}K	
		n	range	n	range	n	range	n	range
			Bq kg^{-1}		Bq kg^{-1}		Bq kg^{-1}		Bq kg^{-1}
Litaor (1995)	Rocky Flats & vicinity	*	*	118	30-285	*	*	*	*
Myrick et al. (1983)	Colorado along I-70 & I-25, & SW Colorado	20	4-115	32	17-111	32	18-126	*	*
NIST (1980)	903 pad at Rocky Flats	1	69	1	39	1	43	1	723
Phair & Gottfried[8] (1964)	Rocky Mountains, CO	392	92	392	57	*	*	*	*
Phair & Jenkins[9] (1975)	Rocky Mountains, CO	250	1-2,020	250	5-1,700	*	*	250	192-3,720[11]
Revzan[10] et al. (1988)	North-Central Colorado	*	*	*	*	Aerial	33	*	*
USDOE (1993)	Rock Creek, CO NE of Rocky Flats	*	*	16	33-56	10	28-41	18	*[11]
USDOE (1995)	East of Colorado's Front Range	*	*	20	27-96	20	43	21	*[19]
Severs and Tourtelot (1994)	Front Range Corridor, CO	*	*	*	*	*	*	845	670-1,300[11]

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[8] Reported concentrations are area weighted means for granitic intrusions in the Colorado Front Range

[9] Concentrations are for samples of granitic rocks

[10] Mean surface concentrations of radionuclides for six 1° X 2° aerial radiometric quadrangles

[11] Incomplete dissolution of potassium feldspars during analysis was reported

Table 2. (cont.) Summary of some soil concentrations for ^{232}Th, ^{238}U, ^{226}Ra and ^{40}K that have been reported in literature.

Investigators	Location	^{232}Th		^{238}U		^{226}Ra		^{40}K	
		n	range	n	range	n	range	n	range
			Bq kg^{-1}		Bq kg^{-1}		Bq kg^{-1}		Bq kg^{-1}
This Study	Rocky Flats and vicinity	42	40-130	*	*	42	32-86	42	360-860
This Study	East of Colorado's Front Range	19	40-160	*	*	19	42-110	19	290-2,000
		n	mean	n	mean	n	mean	n	mean
Lowder et al[12] (1964)	United States	200	37	200	66	*	*	200	400
NCRP[13] (1991)	United States	*	*	*	22	*	*	*	*

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[12] These soil concentrations are also reported by the NCRP (1992)

[13] Mean concentration for an unspecified number of samples

Table 3. Mean soil concentrations of ^{232}Th, ^{226}Ra and ^{40}K associated with soil type, soil over surficial deposits and soil over bedrock around Rocky Flats.

Number Of Locations	^{232}Th mean Bq kg^{-1}	^{232}Th SD Bq kg^{-1}	^{226}Ra mean Bq kg^{-1}	^{226}Ra SD Bq kg^{-1}	^{40}K mean Bq kg^{-1}	^{40}K SD Bq kg^{-1}	Description
Concentrations in Various Soil Types							
1	130	17	106	12	930	90	Hargrave Fine Sandy Loam
6	130*	11	71	8	840	60	Valmont & Leyden-Primen-Standley Cobbly Clay Loam
4	120	11	74	9	850	60	Standley-Nunn Gravelly Clay Loam
4	120	12	74	9	1100*	70	Denver-Kutch, Nunn & Ulm Urban Land Complex
17	120	8	67	6	880	50	Denver, Denver-Kutch-Midway, Englewood & Nunn Clay Loam
2	110	14	64	10	880	76	Heldt Clay
1	110	14	70	10	920	80	Gravelly Soil On Shale Outcrop
8	100	11	69	8	890	60	Haverson & Platner Loam
7	93	10	60	7	850	50	Flatirons & Nederland Cobbly Sandy Loam
1	54	17	36	12	480	90	Samsil Shingle Complex
Concentrations in Soil Over Surficial Deposits							
2	130*	14	85*	11	830	80	Piney Creek Alluvium
1	120	16	81	12	800	90	Louviers Alluvium
8	120	13	64	10	900	70	Rocky Flats Alluvium
4	110	14	61	11	870	80	Loess-stratified clay, silt and sand with gravel lenses
7	110	7	74	5	830	40	Verdos Alluvium
17	110	7	71	6	860	40	Residual Soil and Detritus over the Laramie Formation
6	110	14	61	10	820	80	Older Stoney to Clayey Colluvium
1	100	20	63	15	1200	110	Residual soil and Detritus over the Arapahoe Formation
4	90	10	75	8	760	57	Terrace Gravel
1	90	20	54	15	720	110	Undifferentiated Alluvium
Concentrations in Soil Over Bedrock Types							
2	150*	21	73	16	1200*	120	Pierre Shale
35	93	7	64	5	740	40	Laramie Formation
14	85	8	70	6	600*	50	Arapahoe Formation

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* Soil concentrations that are different from the overall mean for the particular radionuclide at the 0.05 level of significance.

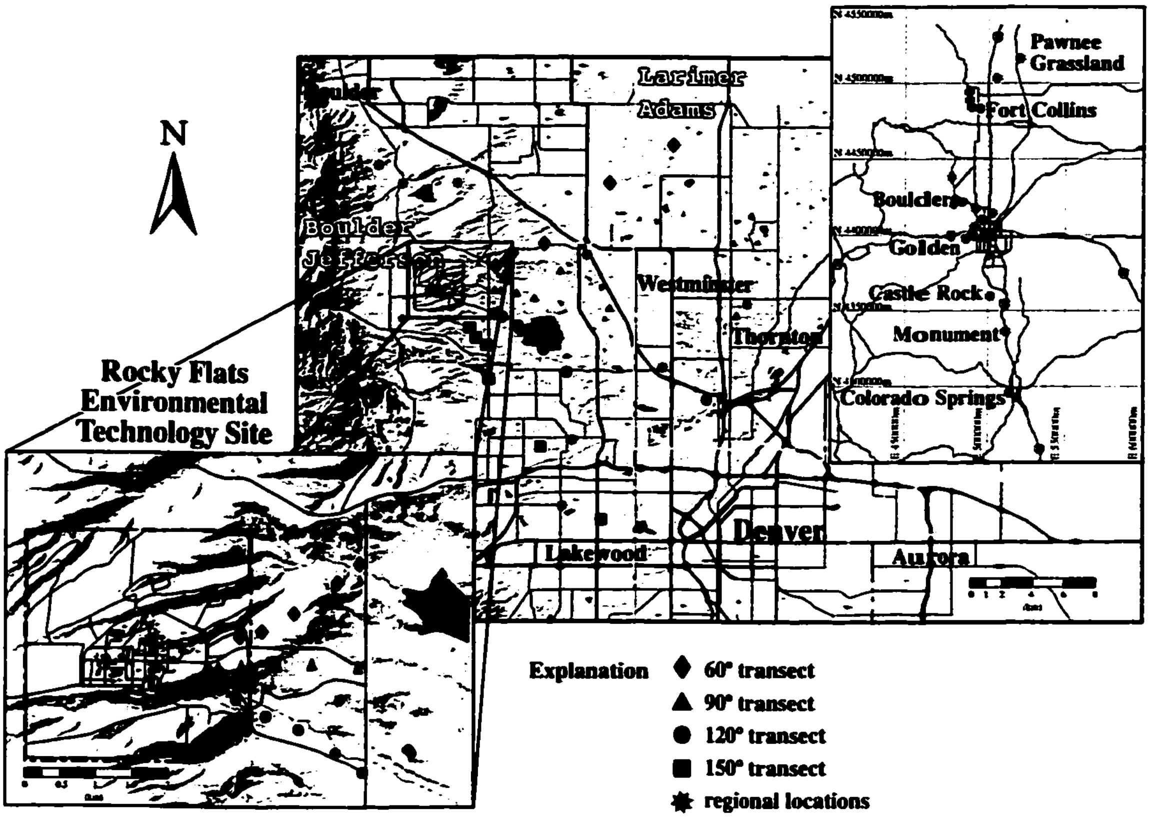
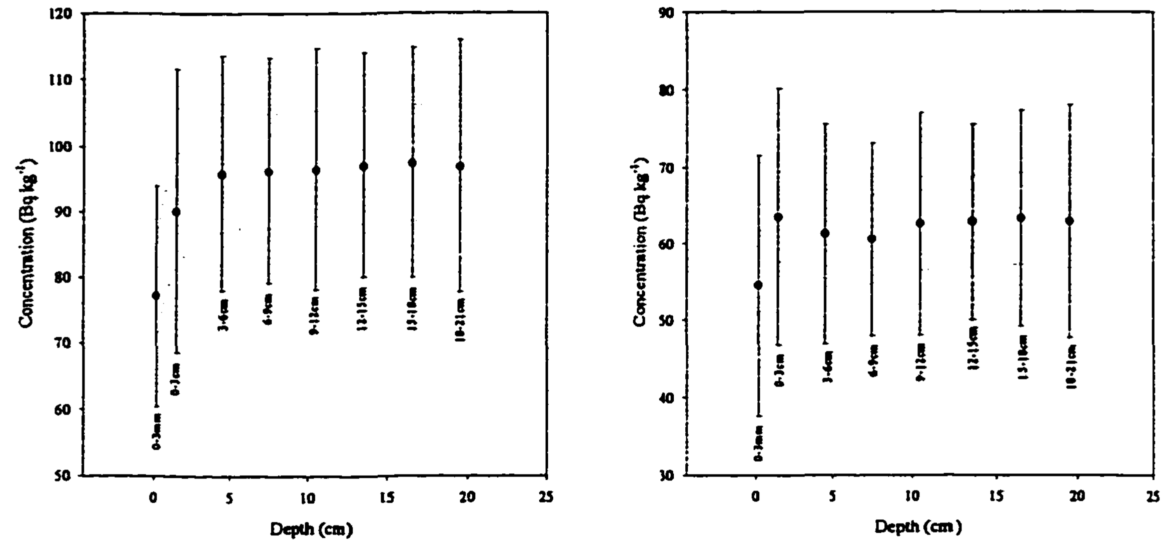
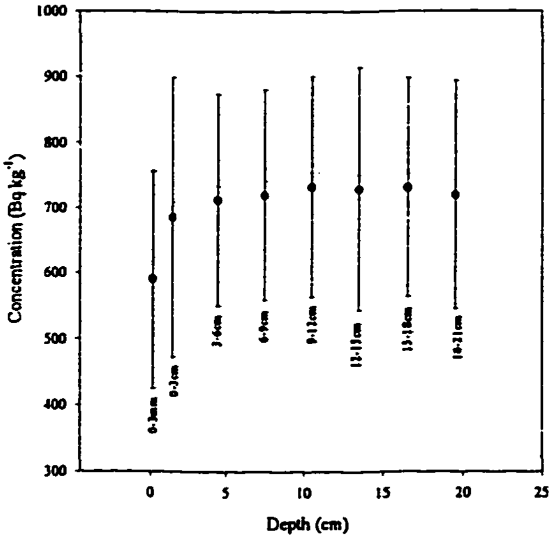


Figure 1. Sample locations around Rocky Flats in Jefferson, Boulder and Adams Counties, and along Colorado's Front Range Corridor from Colorado Springs to Fort Collins, Colorado.



^{232}Th

^{226}Ra



^{40}K

Figure 2. Mean concentrations of ^{232}Th, ^{226}Ra and ^{40}K within incremental depth intervals in soil around Rocky Flats, Jefferson County, CO. Error bars are for the standard deviation of the mean concentration for all on-site and off-site locations. Dotted lines are for the average standard deviation for individual locations.

COMPARISON OF ^{241}Am, ^{239,240}Pu AND ^{137}Cs CONCENTRATIONS IN SOIL AROUND ROCKY FLATS

BACKGROUND

Releases of weapons grade plutonium from Rocky Flats[14] to the environment occurred after cutting oil contaminated with plutonium leaked from corroded drums into soil at the 903 pad (Little and Whicker, 1978; Mongan et al, 1996a), and as the result of fires in 1957 (Mongan et al, 1996b) and 1969 (ChemRisk, 1994). Releases during routine operations were relatively insignificant (Ripple et al, 1996). Most of the contamination was in soil within the Rocky Flats site boundary. Although there was almost no ^{241}Am with weapons grade plutonium after initial purification, there has been an in-growth of ^{241}Am following decay of ^{241}Pu ($T_{1/2}$ = 14.4 y). Krey and Krajewski (1972) and Martell (1975) reported that weapons grade plutonium used at Rocky Flats contained about 0.58% ^{241}Pu. We have estimated that peak soil concentrations of ^{241}Am will occur during the year 2032, which is consistent with the estimate of 2033 by Krey et al. (1976a) after accounting for uncertainties in annual releases. Soil concentrations of ^{241}Am and ^{239,240}Pu have been used in numerous studies to characterize the dispersion of weapons grade plutonium from these releases. With the exception of a companion study by Webb et al.(1997), these relied on relatively small numbers of samples because of the time and expense of radiochemical separation and alpha spectroscopy measurements. We used

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[14] The Rocky Flats plant site and the surrounding buffer zone have been renamed by the United States Department of Energy. The area is now referred to as the Rocky Flats Environmental Technology Site.

gamma spectroscopy measurements of ^{241}Am to further explore patterns of contaminant deposition in soil around Rocky Flats. Previous reports describing the distribution of weapons grade plutonium in the environment around Rocky Flats based on soil concentrations of ^{241}Am include those by Poet and Martell (1972), Krey et al.(1972; 1976a; 1976b; 1977), Litaor et al.(1994; 1996), Schierman (1994), Hardy and Krey (1995), Ibrahim et al.(1995; 1996), Litaor (1995a; 1995b), Litaor and Allen (1996) and the Citizens' Environmental Sampling Committee (1996).

Redistribution caused by disturbance and natural erosion following initial dispersion has probably affected current deposition patterns of ^{241}Am and ^{239,240}Pu around Rocky Flats. Measurements of ^{137}Cs from global fallout can be used to assess the net result of these processes because like ^{241}Am and ^{239,240}Pu, ^{137}Cs is strongly adsorbed to small soil and clay particles. Measurements of ^{137}Cs have been used successfully to assess soil/sediment erosion and deposition patterns because of this property (Ritchie and McHenry, 1973; 1990; Whicker et al, 1994; Heit and Miller, 1987). The peak deposition of ^{137}Cs from global fallout occurred during 1964 (Krey et al, 1990) at about the same time as major actinide releases from Rocky Flats between 1957 and 1969 (Ripple et al, 1996; Mongan et al, 1996a; 1996b). These radionuclides have therefore all been present in local soils for roughly similar time periods (~ 30 ± 5 y).

Measurements of ^{137}Cs could reveal much about the expected behavior of ^{241}Am and ^{239,240}Pu if it could be demonstrated that these actinides have quantitatively similar behavior to that of ^{137}Cs in soil around Rocky Flats. Shapes of depth profiles for soil concentrations could be compared to assess the similarity in behavior of these radionuclides. Soil concentrations of ^{241}Am and ^{239,240}Pu could be normalized to concentrations of ^{137}Cs in the same samples if the depth profiles are statistically indistinguishable, possibly enabling considerable refinement in our understanding of actinide deposition and potential migration around Rocky Flats. Use of ^{137}Cs as a surrogate to predict distributions of ^{241}Am and ^{239,240}Pu could be advantageous because ^{137}Cs is generally more abundant than ^{241}Am in soil around Rocky Flats except near the 903 pad. In addition, more samples could be analyzed with gamma spectroscopy, which is relatively fast and inexpensive when compared to radiochemical separations and alpha spectroscopy.

Specific objectives of this study were to:

- Characterize background concentrations and depositions of ^{137}Cs and ^{241}Am that are unrelated to Rocky Flats immediately east of Colorado's Front Range,
- Determine how concentrations of ^{241}Am, ^{239,240}Pu and ^{137}Cs change with depth in soil around Rocky Flats,
- Examine the strength of correlations between depth-normalized ^{241}Am, ^{239,240}Pu and ^{137}Cs concentrations in soil samples,
- Examine the effect of soil disturbance on the distribution of ^{137}Cs and thus ^{241}Am and ^{239,240}Pu, and
- Estimate ratios of ^{241}Am and ^{239,240}Pu to ^{137}Cs, and examine the relationship of these ratios to distance and direction from the 903 pad.

METHODS AND MATERIALS

Area of Investigation and Sample Locations

The Rocky Flats Environmental Technology Site, located 20 km northwest of Denver, CO, includes an industrial area of about 1.5 km^2 surrounded by a 26.5 km^2, relatively undisturbed buffer zone between the plant and neighboring locales. During 1993 and 1994, 1,309 soil samples were collected from the buffer zone, moderately populated outlying residential communities, densely populated urban areas, and undeveloped open-space. Soil samples were collected from 18 on-site and 24 off-site locations along four radial transects, each approximately 20 km long (Fig. 1). An effort was made to sample six to thirteen 10-m by 10-m macroplots in undisturbed areas along each transect (Stone, 1994). Macroplots were spaced at logarithmically increasing intervals along transects to assure more detailed information near the 903 pad, the most likely source of contamination. The transect azimuths corresponded to the direction of prevailing strong winds with bearings of 60°, 90°, 120° and 150° from the 903 pad relative to true North. Samples were also taken from 10 sites in adjacent communities and 10 other sites from Colorado Springs to Fort Collins believed to be well outside the area measurably affected by contaminants from Rocky Flats.

Soil Sampling

Three types of soil samples were taken from randomly selected 1-m^2 microplots within each macroplot. The first was a scrape from the top 3 mm of soil over an area 1.0-m by 0.5-m. This sampling method, used to characterize the soil fraction most susceptible to resuspension, has been described by Terry (1994). Samples from 3 cm thick horizontal layers of soil were also taken from an area 25 cm long by 15 cm wide to a depth of 21 cm from the surface at each on-site macroplot and one of four microplots in each off-site macroplot. Composite samples from 0 to 21 cm soil columns were taken at three other microplots in each off-site macroplot and at background locations. Rocks larger than 3 cm in diameter were removed from soil samples and weighed in the field. Rock volume in samples was estimated using an average density of 2.65 g cm^{-3} (Carmichael, 1989), since most were either crystalline or metasedimentary rocks. In-situ soil density estimates were made following protocols described in Method D2167-84 (ASTM, 1984). Soil densities were adjusted to account for the decreased sample bulk density and volume after rocks were removed. This adjustment was significant where rocks occupied a large fraction of the sampled volume, since these rocks probably contained no detectable ^{137}Cs or ^{241}Am (Higley, 1994).

Sample Preparation

Soil samples were air dried at room temperature; clods were crushed with a mortar and pestle, and rocks, plant roots and other large debris were removed using a sieve with 4.75 mm openings. The sieved soil was placed in steel cans and oven dried at 110° C for 24 hours, after which samples were mechanically sieved for 30 minutes using an ASTM #10 sieve with 2 mm openings. Dried samples were placed in steel cans 10 cm in diameter and 6 cm high. Cans were sealed with silicone and stored for at least 30 days to bring short lived natural radionuclides into equilibrium and to allow the Compton region to stabilize before gamma spectroscopy measurements were made. Concentrations of ^{232}Th, ^{226}Ra and ^{40}K in the same samples were also measured during an on-going study to characterize soil concentrations of natural radionuclides. We assumed surfaces of rocks, plant material and debris removed from samples did not retain significant activities of ^{137}Cs and ^{241}Am. This assumption was based in part on very low soil-to-plant concentration ratios for ^{241}Am that have been reported (Cline, 1967 cited in Price, 1973). Higley (1994) observed that activity concentrations of ^{241}Am in the gravel fraction (>2mm) of soil accounted for less than one percent of the total ^{241}Am in samples from the Rocky Flats buffer zone. Soil-to-plant concentration ratios for ^{137}Cs have been estimated to be about 0.01 by Ng et al.(1982). In addition, there have been numerous studies demonstrating that the bulk of ^{137}Cs in soil is bound to clay particles, particularly micaceous clay like illite (Francis and Brinkley, 1976; Evans et al, 1983; Cremers et al, 1988; He and Walling, 1996).

On-site samples generally contained 150 g of soil in an attempt to limit the total volume of material that was removed from Rocky Flats because these samples were subject to stringent inventory and future waste disposal requirements. Most off-site samples contained 250 g of soil because lower concentrations of ^{241}Am were expected.

Counting Standards

Sets of counting standards were prepared by spiking uncontaminated soils with solutions containing known activities of either ^{137}Cs or ^{241}Am. Each set included standards with soil masses ranging from 50 to 400-g in 50-g increments to account for geometry and self-absorption differences. Both ^{137}Cs and ^{241}Am soil standards were prepared with a range of activities for each of the different soil masses. Activity concentrations in ^{241}Am standards ranged from 15 to 7,500 Bq kg^{-1}. Activity concentrations in ^{137}Cs standards ranged from 10 to 1,000 Bq kg^{-1}. Both sets included soil that was not spiked. This enabled construction of a three-dimensional calibration matrix that accounted for soil mass, level of activity, and energy. The efficacy of these standards was verified by comparing predicted activity concentrations of ^{137}Cs (1.93 Bq ± 8% predicted, 2.01 Bq ± 4.5% reported) and ^{241}Am (0.14 Bq ± 21% predicted, 0.19 Bq ± 7.3% reported) in a 160-g sample of the Rocky Flats Soil #1[15] with activity concentrations reported by NIST (1980) that were adjusted for decay. The soil sample was packaged with the same geometry as field samples.

An aqueous solution containing ^{137}Cs, obtained from the National Institute of Standards Technology Reference Materials Program[16], was used to prepare counting standards for ^{137}Cs. Soil for the ^{241}Am counting standards was spiked with an internally calibrated aqueous solution. Specific activity in the ^{241}Am solution was determined by evaporating known masses of the solution to dryness on four platinum planchets and estimating areas under the ^{241}Am peaks in the alpha spectra measured with a surface barrier detector. Alpha counting efficiency was determined with electroplated mixed Pu standards traceable to NIST (Ibrahim, 1995).

Counting, Data Analysis and Statistical Methods

Samples were counted for at least 100 minutes on an HPGe (95.8% efficiency relative to a 3 inch by 3 inch NaI detector at 1.33 MeV) or a GeLi detector (30% efficiency relative to a 3 inch by 3 inch NaI detector at 1.33 MeV). The Levenberg-Marquardt optimization algorithm (Marquardt, 1963) in SPSS' PeakFit™[17] software was used to obtain a least-squares fit of a Gaussian distribution to each photopeak. After removing counter background effects from the spectra the Gaussian provided the estimate

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[15] NIST Rocky Flats Soil #1. Gaithersburg, MD; National Institute of Standards and Technology; Standard Reference Material 4353; 1980.

[16] NIST Cesium-137 radioactivity standard. Gaithersburg, MD; National Institute of Standards and Technology; Standard Reference Material 4233-D; 1996.

for the photopeak area. Depositions (Bq m^{-2}) of ^{137}Cs and ^{241}Am at all on-site and one microplot in each off-site macroplot were estimated by summing incremental contributions from each 3 cm depth interval. Depositions were estimated directly for samples from other off-site microplots and background locations using concentration data for composite samples from 0 to 21 cm soil columns.

A method originally proposed by Currie (1968) with modifications recommended in ANSI Standard Method N13.30 (ANSI, 1989) was used to estimate lower limits of detection for 250 g samples. Lower limits of detection were 0.07 Bq for ^{137}Cs and 0.1 Bq for ^{241}Am when measurements were made with the HPGe detector system. Lower limits of detection were 0.2 Bq for ^{137}Cs and 0.3 Bq for ^{241}Am when the GeLi detector was used. The critical level described by Currie is the level used to distinguish between samples that contain an analyte and samples in which any analyte concentration is below the minimum detectable amount. Rather than use a blank paired with each sample as described by Currie, one blank consisting of an empty steel can like those used for soil samples was counted for each ten samples because variability between blank counts was negligible throughout this investigation.

Currie's critical level enables a decision as to whether a sample contains an analyte; however, the critical level does not provide information about whether the analyte concentration is in excess of normal background levels. A second statistic, the decision level described by Borak and Kirchner (1995), was used to distinguish environmental samples containing background concentrations of ^{241}Am from those containing contaminants from Rocky Flats (Table 1). We set α levels for Type I errors to

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[17] SPSS, Inc. PeakFit Version 4.0, © 1995, 2592 Kerner Blvd. San Rafael, CA 94901

0.20 when estimating decision levels listed in Table 2. We believe these error levels were reasonable after considering unavoidable uncertainties from sampling and measurements. Soil concentrations for ^{241}Am and ^{137}Cs at 20 background locations and seven locations at the east ends of the 60°, 90° and 120° transects were used to estimate decision levels. The seven extra locations where soil concentrations and depositions of ^{137}Cs, ^{239,240}Pu and ^{241}Am were within one standard deviation of the mean concentration for background locations and effects from Rocky Flats were believed to be minimal, were included to assure a sufficiently large statistical sample ($n \sim 30$) as recommended by Borak and Kirchner (1995).

When the assumptions that independent populations had normal distributions with common variance were valid, t-tests were used to compare population means. Means for independent samples were compared with two-sample t-tests; for example, comparisons of on-site and off-site samples. Means for matched samples were compared with paired t-tests as was the case for split samples or recounts for the same sample. The Anderson-Darling test (Ryan and Joiner, 1976) was used to test the assumption that populations had normal distributions. Results of two sample t-tests were corroborated using the non-parametric Mann-Whitney two sample rank test and confidence interval (Minitab, 1996) and the non-parametric Kruskal-Wallis test (Ott, 1988). The 0.05 level was used as the measure for significance during this study. Differences between two-sample t-tests and non-parametric tests were generally negligible so only results for t-tests have been reported. The median was used as a measure of central tendency for log-normal distributions rather than the geometric mean and the 95% non-parametric sign confidence interval (Gibbons, 1976; Hollander and Wolfe, 1973) was used rather than the standard deviation in several cases. The sign confidence interval provides a good estimate of the confidence interval for non-symmetric as well as a variety of symmetric distributions (Minitab, 1996). Non-parametric statistics were used to facilitate comparisons with soil concentrations that were reported by others to have non-normal distributions. Statistics were calculated using algorithms in the Minitab™[18] statistical software package.

Quality Assurance and Quality Control

Stone et al.(1994) has described quality assurance and quality control intended to assure representative and reproducible field samples. Systematic counts of an empty can were made before counting each batch of 11 field samples to estimate and monitor counter background. One hundred and nineteen randomly selected samples were recounted to evaluate variability within samples caused by changes of geometry in containers that were partly filled with soil. Sample volumes varied because a constant volume was sampled with a fixed geometry; however, the amount of soil depended on the volume of rocks that were removed when samples were sieved. Mean concentrations of both ^{137}Cs and ^{241}Am for the original and recounted populations were not significantly different. Seventy-one replicates, prepared by splitting randomly chosen samples, were analyzed in the blind to assess variability between samples taken from identical locations. Differences between concentration estimates for both ^{137}Cs and ^{241}Am in split samples were also insignificant.

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[18] Minitab, Inc. Minitab Version 11.12 © 1996, 3081 Enterprise Drive, State College, PA 16801-3998

RESULTS AND DISCUSSION

Background Concentrations and Depositions

Background activity concentrations of ^{241}Am in 26 soil samples from depths of 0 to 3 cm were approximately log-normally distributed with a median (95% sign confidence interval) of 1.3 (1.1 to 1.6) Bq kg^{-1} along Colorado's Front Range (Table 1). Depositions of ^{241}Am in 0 to 21 cm soil columns at 20 background locations were also approximately log-normally distributed with a median (95% sign confidence interval) of 0.20 (0.15 to 0.30) kBq m^{-2}. The USDOE (1995) reported that ^{241}Am concentrations in fifty soil samples from depths of 0 to 5 cm along Colorado's Front Range ranged from 0.04 to 0.9 Bq kg^{-1} and that concentrations in similar samples from Rock Creek, north-northeast of Rocky Flats, ranged from 0.4 to 1.3 Bq kg^{-1}.

The median concentration (95% sign confidence interval) of ^{137}Cs in soil from depths of 0 to 3 cm at all our sample locations was 120 (70 to 140) Bq kg^{-1}(Table 1). The highest concentrations and depositions of ^{137}Cs were in soil from sites more than 50 km south of Rocky Flats, and at on-site and off-site locations northeast and off-site locations east and southeast of the 903 pad. Depositions of ^{137}Cs in 0 to 21 cm soil columns were approximately log-normally distributed with a median (95% sign confidence interval) of 6.4 (5.4 to 8.4) kBq m^{-2}.

Global fallout from weapons testing during the 1950's and 1960's, with minor additions during the 1970's following weapons tests by the Chinese (Rees et al, 1977 cited in Yoder, 1977) and the Chernobyl accident (Volchock and Chieco, 1986), appear to be the only confirmed sources of ^{137}Cs around Rocky Flats. Depositions of ^{137}Cs in soil east of Colorado's Front Range have been reported to range from 0.06 to 7.4 kBq m^{-2}

during the Fort St. Vrain Nuclear Generating Station environmental monitoring program (Anonymous, 1975), and by Perkins and Thomas (1980), Purtyman et al.(1990), and the USDOE (1993; 1995), among others. Soil depositions of ^{137}Cs in the Denver area around Rocky Flats reported by Michels (1974), Thomas (1974), Colorado Department of Health (1977), Harley (1977), Johnson (1977), Murri (1977), Yoder (1977), Gogolak and Miller (1978) and the Citizens' Environmental Sampling Committee (1996) are similar to those for other areas east of Colorado's Front Range. The greatest depositions have been reported north-northwest and south-southwest of Rocky Flats on east facing slopes of the Front Range. The wide range in depositions can probably be attributed to variations in topography, wind patterns, differences in precipitation with elevation, and differences in physical properties of soils at various locations along the eastern flanks of the Rocky Mountains (Perkins and Thomas, 1980; Whicker and Schultz, 1982; Arnalds et al, 1989).

Depth Distributions

Comparison of ^{137}Cs, ^{241}Am and ^{239,240}Pu

Soil concentrations, C(z) in Bq kg^{-1}, of ^{241}Am, ^{239,240}Pu and ^{137}Cs at depth, z in cm, decreased from the surface to a depth of 21 cm at rates that can be characterized with exponential functions having the form,

$$C(z) = C_1 e^{mz}, \qquad \text{(Equation 1)}$$

where C_1 is the concentration in Bq kg^{-1}for soil from depths of 0 to 3 cm. At each macroplot, the coefficient, m (cm^{-1}), was estimated as the slope of a linear regression of the natural logarithms of concentrations on depth.

Estimated values of m for ^{137}Cs were nearly constant with a mean of

-0.25 ± 0.03 cm^{-1} at on-site locations and -0.25 ± 0.06 cm^{-1} off-site. Mean estimates of m for $^{239,240}Pu$ were -0.23 ± 0.04 cm^{-1}on-site and -0.22 ± 0.18 cm^{-1} off-site using data from Webb et al.(1994; 1997). Webb et al.(1993) also reported an estimate of m for $^{239,240}Pu$ of -0.23 cm^{-1} at a location 225-m east-southeast of the 903 pad.

Rates at which soil concentrations of ^{137}Cs and $^{239,240}Pu$ decreased with depth at each macroplot were compared by regressing differences between natural logarithms of soil concentrations for ^{137}Cs and $^{239,240}Pu$ on depth. Although there were local variations, mean slope estimates for the differences were not significantly different from zero (p = 0.16), leading to the conclusion that movement of ^{137}Cs and $^{239,240}Pu$ in soil profiles occurs at rates that are nearly indistinguishable. In addition, soil concentrations of both ^{137}Cs and $^{239,240}Pu$ were strongly correlated when these were normalized to their respective mean concentrations in soil from depths of 0 to 3 cm at each location (Fig. 2).

Estimated values of m for ^{241}Am averaged -0.22 ± 0.04 cm^{-1} at the four sample locations closest to the 903 pad. The estimated value of m for ^{241}Am became closer to zero with increasing distance at locations more than 0.5 km from the 903 pad. We believe this resulted from inherent errors in near-background estimates for soil concentrations of ^{241}Am, particularly in deeper samples. Rates at which of ^{241}Am decreased with depth at the four close-in sample locations were compared with rates at which ^{137}Cs and $^{239,240}Pu$ changed by regressing differences between the natural logarithms of concentrations on depth as was done for ^{137}Cs and $^{239,240}Pu$. Mean slope estimates were not significantly different from zero (p = 0.26 for the difference with ^{137}Cs and p = 0.88 for the difference with $^{239,240}Pu$). This observation led to the conclusion that the mean rate at which soil concentrations ^{241}Am changed with depth was nearly indistinguishable from corresponding rates for ^{137}Cs and ^{239,240}Pu at these locations. This is consistent with the observation by Mboulou et al.(1998) that 8 years after release, fractions of ^{241}Am and ^{239,240}Pu decreased at about the same rate with depth in soil around the village of Kopachi several km from a damaged reactor at Chernobyl in the Ukraine. There is also a strong correlation between normalized concentrations of ^{137}Cs and ^{241}Am in samples from all depths at the four sample locations closest to the 903 pad (Fig. 3).

Soil disturbance

Rates at which soil concentrations of ^{137}Cs decreased with depth, as described by estimates of m, were similar at all macroplots around Rocky Flats except where surface soil had been disturbed. On-average, about 60% of ^{137}Cs was retained in the top 3 cm of soil (95% confidence interval from 52% to 65%). About 86% was within 6 cm of the soil surface (95% confidence interval from 79% to 88%). Our estimates for the fraction of ^{137}Cs in the top 6 cm of soil are lower than those by Price (1991), who estimated that 99% of ^{137}Cs was in the top 5 cm of soil at one sample location on the Hanford site in Washington and at an off-site location that was 25 km upwind from the site. These inconsistencies might be caused by differences in soil characteristics, annual precipitation or other factors. Where surface soil was undisturbed at the two locations, it is significant that most ^{137}Cs was in the top 5 to 6 cm of soil because of the tendency of ^{137}Cs to bind tightly to small soil particles. This property, and the ubiquitous distribution of ^{137}Cs from global fallout, provided a gauge for determining if surface soil had been disturbed. Distributions of ^{137}Cs have been used to measure soil losses from erosion (Ritchie and McHenry, 1990) and enhanced deposition of sediments (Heit and Miller, 1987; Whicker et al, 1994).

Soil concentrations of ^{137}Cs were relatively uniform throughout 0 to 21 cm soil columns at 4 of 42 macroplots. These sites were therefore excluded from spatial analyses and comparisons during this study. In addition to uniform soil concentrations of ^{137}Cs in samples from all depths, there was visual evidence of disturbance in the 0 to 21 cm soil column at these sites; for example, nails and other man-made debris as well as compaction at the base of plowed layers (plow soles) in previously tilled fields. Tillage that followed deposition of ^{137}Cs appeared to be the primary cause of disturbance at these sites, as has been observed in other agricultural communities throughout the United States (Cline and Rickard, 1972; Ritchie and McHenry, 1973).

Areal Distribution

Soil concentrations of ^{241}Am

Soil concentrations of ^{241}Am in samples from depths of 0 to 3 cm, $C_1(X, \theta)$ in kBq kg^{-1}, along transects bearing θ degrees from true North, decreased with distance, X in km from the 903 pad, at rates described by the equation,

$$C_1(X,\theta) = B(\theta)\, X^{M(\theta)}, \qquad \text{(Equation 2)}$$

as shown in Fig. 4. Soil concentrations of ^{241}Am at depths from 0 to 3 mm were similar to concentrations at depths of 0 to 3 cm and so could also be described by Equation 2, as could ^{241}Am depositions. Exponents, $M(\theta)$, and constants, $B(\theta)$, for equations describing these relationships are listed in Table 3 with p-values for the estimates, which were less than 0.001 except those for the 150° transect south-southeast of Rocky Flats where concentrations and depositions of ^{241}Am were not significantly different from background levels. Soil concentrations of ^{241}Am in samples from depths of 0 to 3 mm and 0 to 3 cm decreased most rapidly along the 90° and 120° transects, at rates that were nearly proportional to the inverse square of distance. Concentrations of ^{241}Am were indistinguishable from background levels more than 7 km from the 903 pad.

Soil concentrations of ^{239,240}Pu

Soil concentrations of ^{239,240}Pu decreased similarly with distance from the 903 pad (Webb et al, 1997); however, concentrations were above our estimates of the decision level for ^{239,240}Pu (3 Bq kg^{-1} in samples from depths of 0 to 3 cm) up to 10 km away. The theoretical activity concentration of ^{241}Am in weapons grade plutonium released from Rocky Flats was about 19% of that for ^{239,240}Pu during 1994, based on isotopic ratios for weapons grade plutonium reported by Krey and Krajewski (1972). These relative concentration differences probably account for the different distances at which concentrations of ^{241}Am and ^{239,240}Pu were distinguishable from background. When estimating the theoretical percentage for the activity concentration of ^{241}Am in soil relative to ^{239,240}Pu we assumed that cutting oil contaminated with weapons grade plutonium leaked from about the same number of drums that were stored at the 903 pad during each year from 1958 through 1964. We also assumed that the weapons grade plutonium in the drums was purified mid-year during the year of storage. In-growth of ^{241}Am began immediately following purification. The ratio between activity concentrations of ^{241}Am and ^{239,240}Pu was estimated by summing effects of releases from the 903 pad (ChemRisk, 1994; Mongan et al, 1996a), annual releases during operations between 1953 and 1989 (Ripple et al, 1996), and releases during the 1957 (ChemRisk, 1994; Mongan et al, 1996b) and 1969 (ChemRisk, 1994) fires.

Relationships Between ^{241}Am and ^{239,240}Pu In Soil

Relationships between ^{241}Am and ^{239,240}Pu in soil were investigated using our data and ^{239,240}Pu data from splits of the same samples reported by Webb (1996) and Webb et al.(1994; 1997). The data indicate a very strong relationship between ^{241}Am and ^{239,240}Pu in soil from depths of 0 to 3 cm, as expected (Fig. 5). A log-log regression yielded

$$[^{239,240}\mathrm{Pu}] = 5.5\,[^{241}\mathrm{Am}]^{1.1}, \qquad \text{(Equation 3)}$$

with an r-squared value of 0.87 for locations where soil concentrations exceeded decision levels.

The ratio between ^{241}Am and ^{239,240}Pu in soil samples from depths of 0 to 3 cm, which was estimated by regression, was 0.18 for all on-site locations. The ratio for off-site locations was 0.36. The ratio between concentrations of ^{241}Am and ^{239,240}Pu in soil from depths of 0 to 3 mm for all on-site locations was 0.22. This ratio is the same as that reported by Mongan et al.(1996a) in airborne effluents from Rocky Flats and almost the same as the ratio (0.21) reported by Litaor and Allen (1996) in soil from depths of 0 to 6 mm at their on-site sample locations. These results are also consistent with ratios calculated by Litaor et al.(1998) for concentrations in soil from depths of 0 to 20 cm and 20 to 40 cm at locations close to the 903 pad.

Schierman (1994) and Ibrahim et al.(1995; 1996) used radiochemical separation and alpha spectroscopy to independently estimate the mean ratio between activity concentrations of ^{241}Am and ^{239,240}Pu in splits from sixty-three soil samples that we analyzed with gamma spectroscopy. Their samples were taken from five locations along the 90° and 120° transects closest to the 903 pad within the buffer zone at Rocky Flats.

The Pearson correlation coefficient between concentrations of ^{241}Am estimated by Schierman from alpha spectroscopy measurements and estimates that we made with soil-sample gamma spectroscopy measurements was 0.83. Schierman and Ibrahim et al.estimated that the mean ratio between ^{241}Am and ^{239,240}Pu was 0.19 during 1994. We calculated the same ratio of 0.19 when soil concentrations were limited to splits of samples that Schierman and Ibrahim et al.analyzed.

Non linear regression was used to estimate parameters for a Hill four-parameter sigmoidal[19] model that predicts the ratio [^{241}Am]:[^{239,240}Pu] in soil from depths of 0 to 3 cm (Y) at distance (X in km) from the 903 pad. Parameters for the model, which has the form,

$$Y = Y_0 + (aX^b)(c^b + X^b)^{-1}, \qquad \text{(Equation 4)}$$

Y_0, a, b, and c^b have values of 0.21, 0.21, 5.16 and 1.16 respectively. The model predicted that the ratio (95% confidence interval) should be 0.21 (0.15 to 0.29) close to the 903 pad and 0.41 (0.35 to 0.51) at distances of more than 50 km. The r-squared value for the sigmoidal curve fit was 0.45. The predicted ratio of 0.41 at background locations was higher than the mean ratio of 0.36 estimated directly from soil concentrations in samples from off-site locations. This should be expected since some of the off-site samples from locations closest to Rocky Flats were affected by releases of weapons grade plutonium. In addition, ratios between soil concentrations at background locations were not included in the off-site estimate. The predicted ratio was higher than the ratio of 0.32 obtained by adjusting the ratio estimated by Krey et al.(1976b) for additions of ^{241}Am from the decay of ^{241}Pu in global fallout since 1974. It is also higher than the ratio of

0.29 ± 0.17 that we estimated using concentrations in background soil samples reported by the USDOE (1995). Litaor and Allen (1996) reported, however, that the mean ratio between ^{241}Am and ^{239,240}Pu in soil from depths of 0 to 6 mm at off-site locations around Rocky Flats was 0.56. Clearly, these ratios should be used judiciously when predicting concentrations of ^{239,240}Pu from soil concentrations of ^{241}Am to delineate the extent of contamination from Rocky Flats.

Soil Concentrations of ^{137}Cs along Sampling Transects

Median soil concentrations and ranges for ^{137}Cs in soil from depths of 0 to 3 mm and 0 to 3 cm and depositions in 0 to 21 cm soil columns along transects around Rocky Flats are listed in Table 4. There were no significant differences between mean concentrations of ^{137}Cs in soil from depths of 0 to 3 mm ($p=0.23$) and 0 to 3 cm ($p=0.33$) at on-site ($n=77$) and off-site ($n=107$) locations. Mean depositions of ^{137}Cs along the 60°, 90° and 120° transects were about twice the mean deposition along the 150° transect. Concentrations and depositions of ^{137}Cs concentrations were compared with mean annual rainfall at the nearest reporting stations[20]; however, there was no apparent correlation. We speculate that increased soil erosion, either by wind or surface runoff, may have removed near-surface soil along with ^{137}Cs south-southeast of Rocky Flats along the 150° transect. Accumulations of wind-blown sand along fences and a relatively dense dendritic pattern of stream channels provide circumstantial evidence that erosion may have occurred, although a soil survey report for the area (Price and Amen, 1980) gave no reason to expect more erosion than in surrounding areas.

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[19] SPSS, Inc. Sigma Plot Version 4.0, © 1995, 2591 Kerner Blvd. San Rafael, CA 94901

[20] Rainfall during the past thirty years at reporting stations along Colorado's Front Range is listed by the Office of Nolan Doeskin, Colorado State Climatologist, HTTP://ULYSSES.ATMOS.Colorado.edu.

Spatial variations in soil depositions of ^{137}Cs were compared to determine if distance from or position along Colorado's Front Range had any effect. Partial correlation coefficients (pcc) between depositions and northerly UTM coordinates were estimated while holding easterly UTM coordinates constant by first regressing deposition on easterly UTM coordinates and estimating residuals, then regressing northerly UTM coordinates on easterly UTM coordinates and estimating residuals. Pearson correlation coefficients for the two sets of residuals provided pcc estimates. The same procedure was used to estimate the pcc for depositions and easterly UTM coordinates while holding northerly UTM coordinates constant. There was a slight tendency for ^{137}Cs deposition to decrease (pcc = -0.18) in an easterly direction away from the foothills and a slightly stronger tendency (pcc=0.4) for ^{137}Cs deposition to increase to the north. The significance of these findings is questionable; however, since the sampling pattern was not originally designed for a critical test of such trends. Furthermore, erosion may have confounded these results.

A test was also made for correlations between soil concentrations of ^{137}Cs and direct distance from the 903 pad, which should be expected if ^{137}Cs was released from that site. There was no apparent correlation between ^{137}Cs depositions and proximity to the 903 pad at Rocky Flats (pcc = -0.06). Global fallout was therefore the most likely source of ^{137}Cs in the area.

Normalization of ^{241}Am and ^{239,240}Pu Data to ^{137}Cs

In an attempt to refine our understanding of ^{241}Am and ^{239,240}Pu distribution patterns in soil, concentrations were normalized to ^{137}Cs. The log-transformed ratios, [^{241}Am]:[^{137}Cs] and [^{239,240}Pu]:[^{137}Cs], were regressed on the log transform of distance for locations 0 to 21 km from the 903 pad (Figs. 6 and 7) and straight-line segments were fit to the data. A similar procedure was used to predict background ratios of 0.007 between ^{241}Am and ^{137}Cs (Fig. 6) and 0.020 between ^{239,240}Pu and ^{137}Cs (Fig.7) from data for locations at greater distances.

The ratios for the 60°, 90° and 120° transects were pooled to estimate mean rates at which the ratios, [^{241}Am]:[^{137}Cs], and [^{239,240}Pu]:[^{137}Cs], changed with distance from the 903 pad at Rocky Flats. These rates can be described with power functions that are similar to Equation 2. Exponents for these equations, -2.1 ± 0.1 for the mean ratios, [^{241}Am:^{137}Cs], and -2.4 ± 0.1 for the mean ratios, [^{239,240}Pu:^{137}Cs], indicate that like activity concentrations in soil, the ratios decrease at rates that are nearly proportional to the inverse square of distance from the 903 pad. R-squared values for linear fits to the combined data for the 60°, 90° and 120° transects out to 21 km from the 903 pad were 84% for the ratio [^{241}Am]:[^{137}Cs] and 81% for the ratio [^{239,240}Pu]:[^{137}Cs]. In comparison, r-squared values for linear fits to the combined ^{241}Am concentrations along the 60°, 90° and 120° transects out to 21 km from the 903 pad were 0.74 for ^{241}Am and 0.75 for ^{239,240}Pu. Data for the 150° transect was omitted from this comparison since soil concentrations of ^{241}Am and ^{239,240}Pu were not significantly different from background levels. The improvement of the fit after normalizing to ^{137}Cs concentrations was particularly evident for soil concentrations of ^{241}Am in samples of gravelly soil from depths of 0 to 3 cm at a location 0.7 km from the 903 pad along the 90° transect on the edge of a borrow pit in the buffer zone at Rocky Flats (Figs. 4 and 6).

There is a strong correlation of relative soil concentrations for both ^{241}Am and ^{239,240}Pu with ^{137}Cs (Figs. 2 and 3). This does not imply that ^{241}Am, ^{239,240}Pu and ^{137}Cs originated from the same source, but it does indicate that these radionuclides move at similar rates and are probably transported by similar mechanisms in soil around Rocky Flats. These correlations also indicate that use of the log-transformed ratios, [^{241}Am]:[^{137}Cs] and [^{239,240}Pu]:[^{137}Cs] rather than log-transformed soil concentrations for ^{241}Am and ^{239,240}Pu may attenuate effects from spatial variability in deposition attributable to wind patterns, precipitation, adsorption properties of soil, erosion and possibly other factors.

Perkins and Thomas (1980) reported that the mean ratio between total plutonium and ^{137}Cs in global fallout was relatively constant throughout the period when atmospheric tests of nuclear weapons were conducted. Miller and Michels (1973) estimated that the mean ratio between concentrations of ^{239,240}Pu and ^{137}Cs from global fallout in soil on the Great Plains east of the Rocky Mountains was 0.021 ± 0.0015. This corresponded to a ratio of about 0.032 during 1994 after adjusting for decay of ^{137}Cs concentrations since 1973. The ratio was about 0.028 in 1994 after adjusting an estimate by Beck and Krey (1983) for decay. Lee et al.(1996; 1997) calculated the mean ratio between concentrations of ^{239,240}Pu and ^{137}Cs in Korean soil was 0.023 ± 0.006 during 1994. Soil concentrations at background locations along Colorado's Front Range reported by USDOE (1995) were used to estimate that the mean ratios, [^{241}Am]:[^{137}Cs] and [^{239,240}Pu]:[^{137}Cs], from global fallout were 0.011 ± 0.004 and 0.041 ± 0.012 respectively during 1994. Hodge et al.(1996) independently analyzed splits of samples from 15 of 50 samples analyzed by the USDOE (1995) and estimated the mean ratio between soil concentrations of ^{239,240}Pu and ^{137}Cs was 0.026 ± 0.003. Cooper et al.(1998) found that the mean ratio between concentrations of ^{239,240}Pu and ^{137}Cs in Arctic sea ice entrained sediments, 0.014 ± 0.007, was relatively constant but suggested that the lower ratio was caused by the addition of ^{137}Cs from secondary sources.

CONCLUSIONS

The following conclusions are drawn from this analysis:

- Background concentrations of ^{137}Cs in soil from depths of 0 to 3 cm and total depositions in soil to depths of 21 cm along Colorado's Front Range vary from 70 to 140 Bq kg^{-1} and 3 to 8 kBq m^{-2} respectively. We believe that the lower levels were found in areas where there has been significant soil erosion and that soil concentrations of ^{137}Cs in soil profiles can therefore be used to screen for soil disturbance. No relationship was found between deposition of ^{137}Cs and distance from the 903 pad at Rocky Flats.

- Approximately 60% of ^{137}Cs was retained in the top 3 cm of soil and 86% was within the top 6 cm except where tillage, erosion and other kinds of disturbance had occurred.

- Background activity concentrations of ^{241}Am in soil from depths of 0 to 3 cm and total depositions in soil to depths of 21 cm had relatively narrow ranges from 0.8 to

  1.6 Bq kg^{-1} and 0.15 to 0.30 kBq m^{-2} respectively.

- Soil depositions of ^{241}Am at on-site locations ranged from 0.36 to 170 kBq m^{-2}. Soil depositions ranged from .15 to .75 kBq m^{-2} at off-site locations within 20 km of the 903 pad at Rocky Flats.

- Activity concentrations of ^{241}Am in the top 3 cm of soil decreased most rapidly in easterly and east-southeasterly directions at rates that were nearly proportional to the inverse square of distance from the 903 pad.

- Activity concentrations of ^{137}Cs, ^{241}Am and ^{239,240}Pu decreased exponentially with depth in undisturbed soil at rates ranging from –0.18 to –0.27 cm^{-1} with no statistically significant differences between mean rates for the three radionuclides at locations where soil concentrations of ^{241}Am were above decision levels.

- Activity concentrations of ^{137}Cs, ^{241}Am and ^{239,240}Pu were very strongly correlated when normalized to corresponding concentrations in samples from depths of 0 to 3 cm.

- Normalizing activity concentrations of ^{241}Am and ^{239,240}Pu in soil to activity concentrations for ^{137}Cs provided a clearer picture of geographical dispersion patterns from the 903 pad at Rocky Flats than normalization to soil mass.

- The ratio between soil concentrations of ^{241}Am and ^{239,240}Pu in the top 3 cm of soil appears to increase from 0.18 at on-site locations to 0.36 at off-site locations. This may be an artifact introduced by inherent errors in concentration estimates; however, these ratios are consistent with ratios reported by other investigators and may reflect differences in composition of weapons grade plutonium released from Rocky Flats and plutonium in global fallout.

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Table 4. Median and 95% sign confidence intervals for concentrations and depositions of ^{137}Cs and ^{241}Am in soil from 20 regional background locations along Colorado's Front Range between Colorado Springs and Fort Collins.

	^{137}Cs		^{241}Am	
	Median	95% C.I.	Median	95% C.I.
0 to 3 mm Soil Fraction (Bq kg^{-1})	95	70 – 130	1.1	0.8 – 1.7
0 to 3 cm Soil Sample (Bq kg^{-1})	120	70 – 140	1.3	1.1 – 1.6
21 cm Depositions (kBq m^{-2})	6.4	5.4 – 8.4	0.20	0.15 – 0.30

Table 5. Decision level estimates for soil concentrations in 250-g samples. Depositions of ^{137}Cs and ^{241}Am had normal distributions. Concentrations had log-normal distributions except for ^{137}Cs concentrations in soil from depths of 0 to 3; however, the distribution was treated as if it were log-normal for the purpose of this estimate.

	^{137}Cs	^{241}Am
0 to 3 mm Soil Fraction (Bq kg^{-1})	150	2
0 to 3 cm Soil Sample (Bq kg^{-1})	150	3
21 cm Depositions (kBq m^{-2})	8.4	0.35

Table 6. Constants, exponents and p-values of estimates for equations describing variations of ^{241}Am concentrations (kBq kg^{-1}) in soil from depths of 0 to 3 mm, 0 to 3 cm and depositions (kBq m^{-2}) in 21 cm soil columns with distance from the 903 pad at Rocky Flats.

	Exponent M(θ)	p-value	Constant B(θ) kBq kg^{-1} or kBq m^{-2}	p-value
60° Transect				
0 to 3 mm Soil Fractions	-1.24 ± 0.16	<.001	0.016 ± 0.001	0.001
0 to 3 cm Sample Interval	-1.29 ± 0.14	<.001	0.018 ± 0.001	<.001
21 cm Deposition	-0.69 ± 0.19	0.005	980 ± 40	<.001
90° Transect				
0 to 3 mm Soil Fractions	-1.75 ± 0.14	<.001	0.10 ± 0.01	<.001
0 to 3 cm Sample Interval	-1.89 ± 0.12	<.001	0.13 ± 0.01	<.001
21 cm Deposition	-1.45 ± 0.24	<.001	6,600 ± 270	<.001
120° Transect				
0 to 3 mm Soil Fractions	-1.83 ± 0.10	<.001	0.043 ± 0.002	<.001
0 to 3 cm Sample Interval	-1.75 ± 0.10	<.001	0.047 ± 0.002	<.001
21 cm Deposition	-1.48 ± 0.20	<.001	4,470 ± 160	<.001
150° Transect				
0 to 3 mm Soil Fractions	-0.17 ± 0.19	0.38	0.0012 ± 0.0001	0.003
0 to 3 cm Sample Interval	-0.08 ± 0.15	0.60	0.0014 ± 0.0002	0.013
21 cm Deposition	-0.46 ± 0.20	0.087	400 ± 30	0.007

Table 7. Median concentrations and depositions of ^{137}Cs in soil along transects east of Rocky Flats. Concentrations in soil from depths of 0 to 3 mm and 0 to 3 cm had log-normal distributions. Depositions had normal distributions.

Transect	0 to 3 mm Soil Fraction (Bq kg^{-1})			0 to 3 cm Sample Interval (Bq kg^{-1})			21 cm Deposition (kBq m^{-2})		
	n	Median	95% C.I.	n	Median	95% C.I.	n	Median	95% C.I.
60° Transect	47	99	88-116	49	116	87-129	11	4.1	3.4-5.2
90° Transect	51	85	78-97	56	115	89-128	12	4.7	3.4-6.5
120° Transect	57	44	35-59	56	51	36-70	13	4.1	2.8-4.6
150° Transect	27	29	16-36	27	40	18-55	6	2.2	1.3-3.5

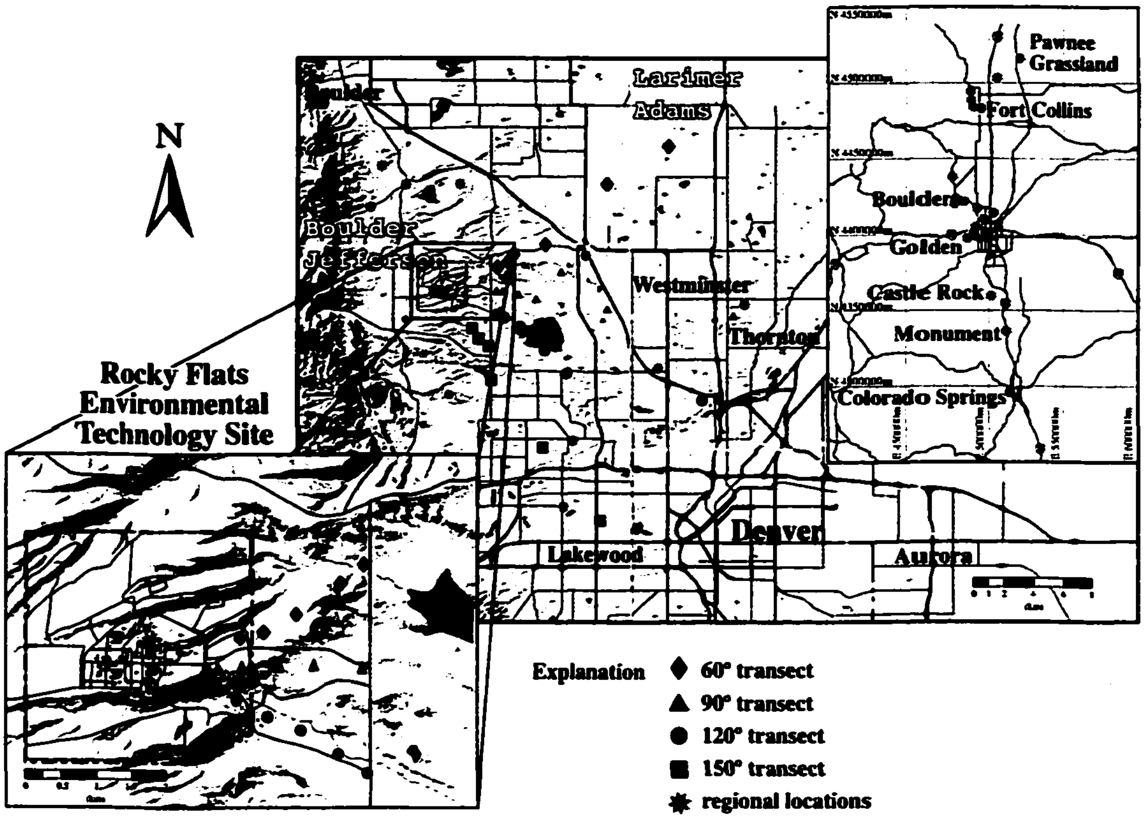

Figure 3. Sample locations around Rocky Flats in Jefferson, Boulder and Adams Counties, Colorado.

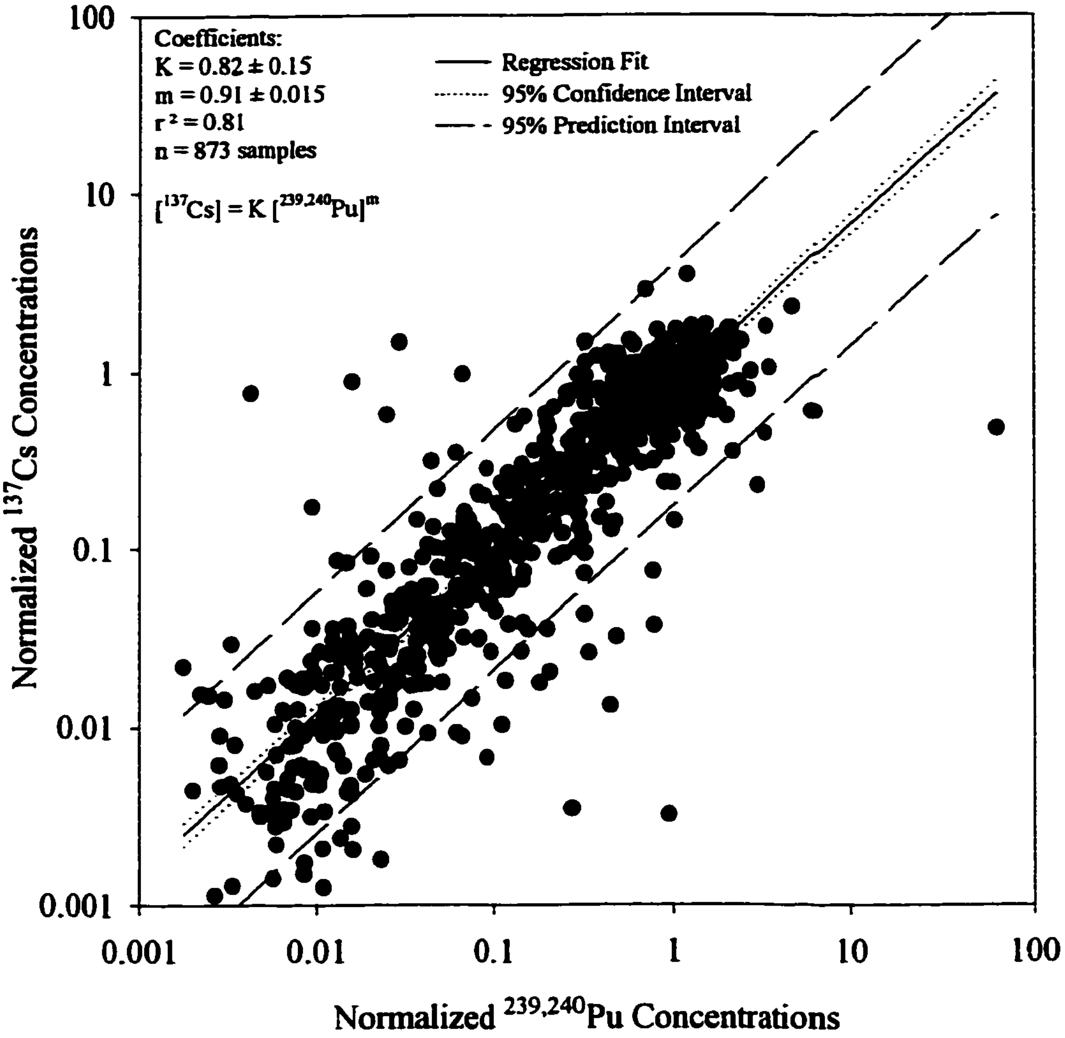

Figure 4. Correlation between concentrations of ^{239,240}Pu and ^{137}Cs in soil from all 3 cm depth intervals at on-site and off-site locations to depths of 21 cm around Rocky Flats. Concentrations were normalized to respective mean soil concentrations in samples from depths of 0 to 3 cm.

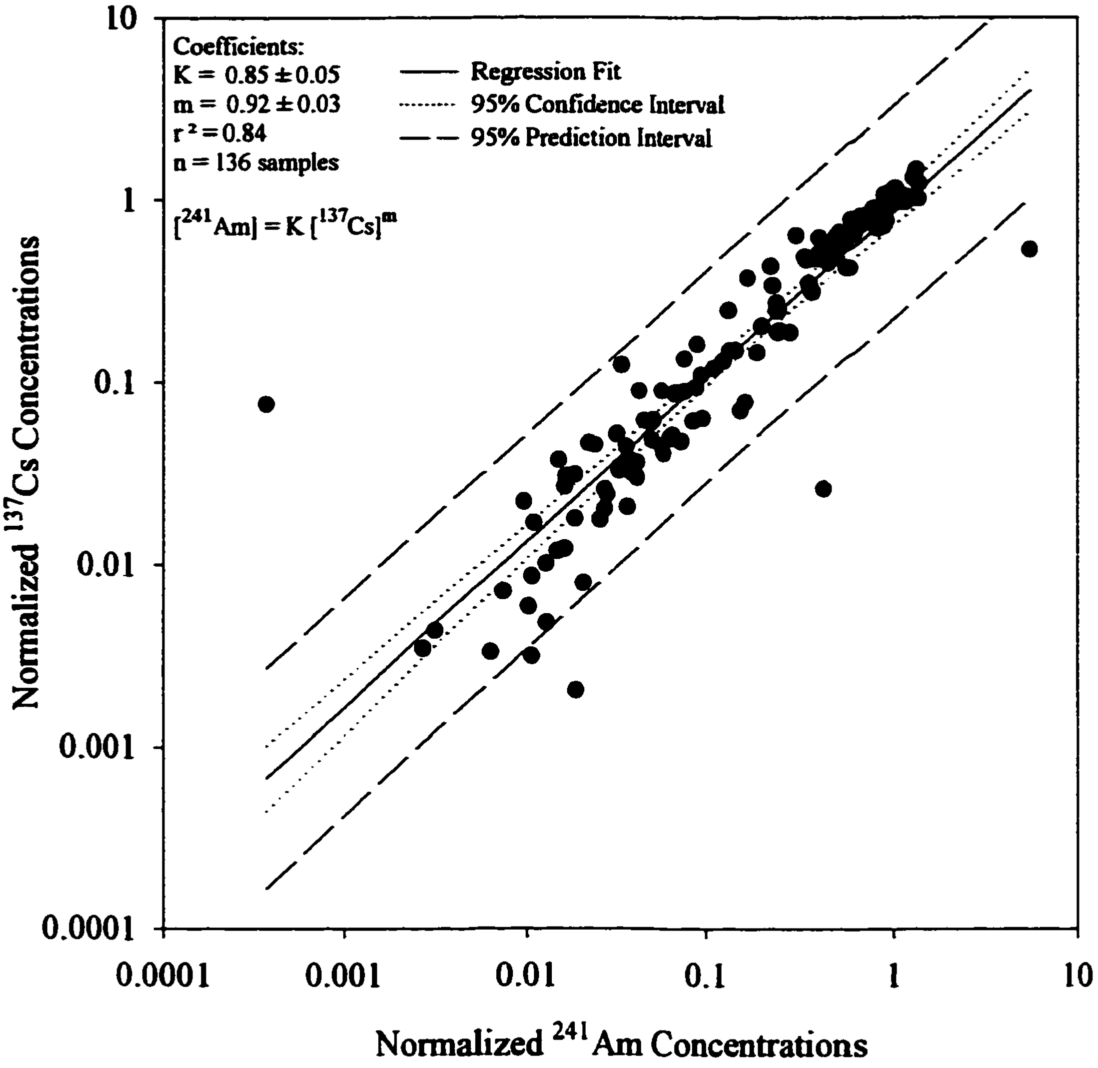

Figure 5. Correlation between concentrations of ^{241}Am and ^{135}Cs in soil from all 3 cm depth intervals at on-site and off-site locations to depths of 21 cm within 0.5 km of the 903 pad. Concentrations were normalized to respective mean soil concentrations in samples from depths of 0 to 3 cm.

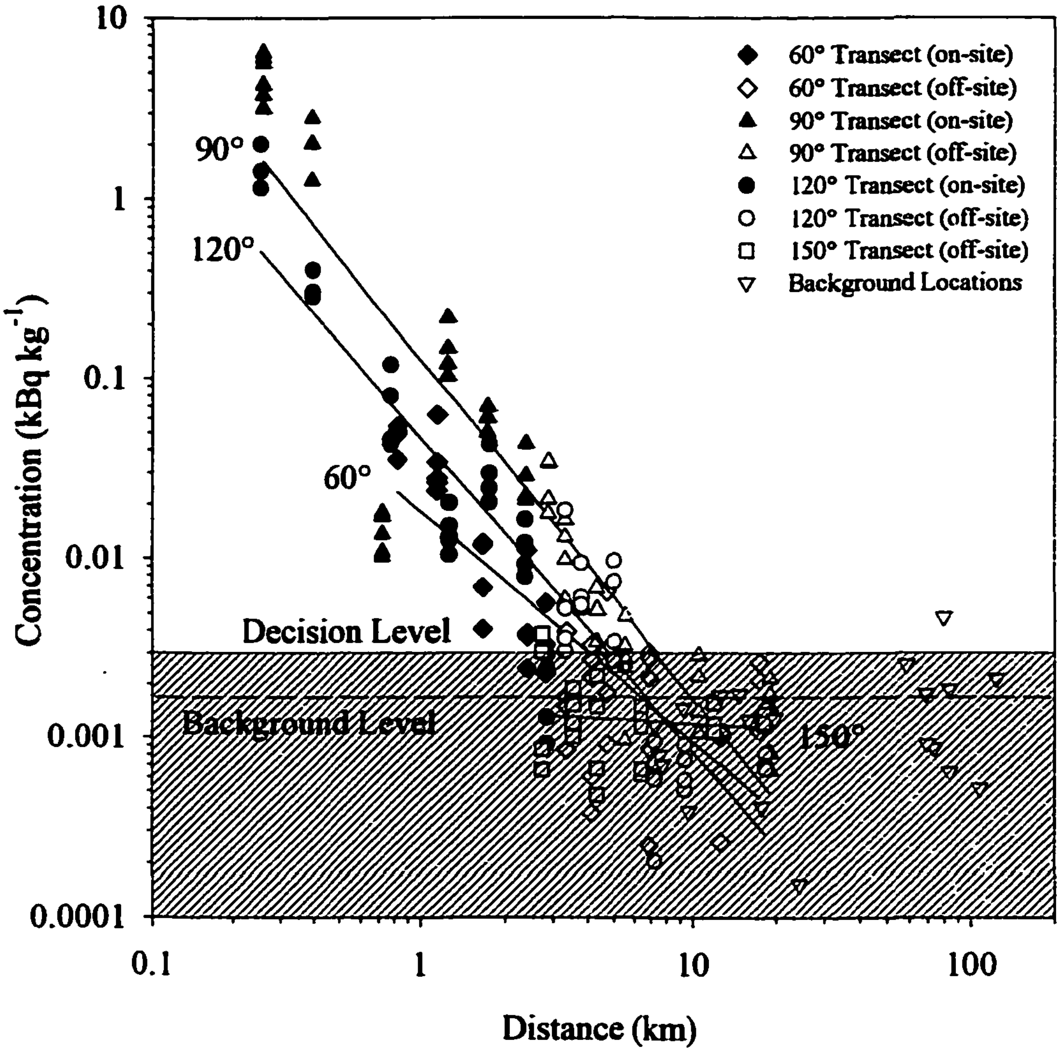

Figure 6. Variations of ^{241}Am concentrations in soil from depths of 0 to 3 cm with distance from the 903 pad east of Rocky Flats.

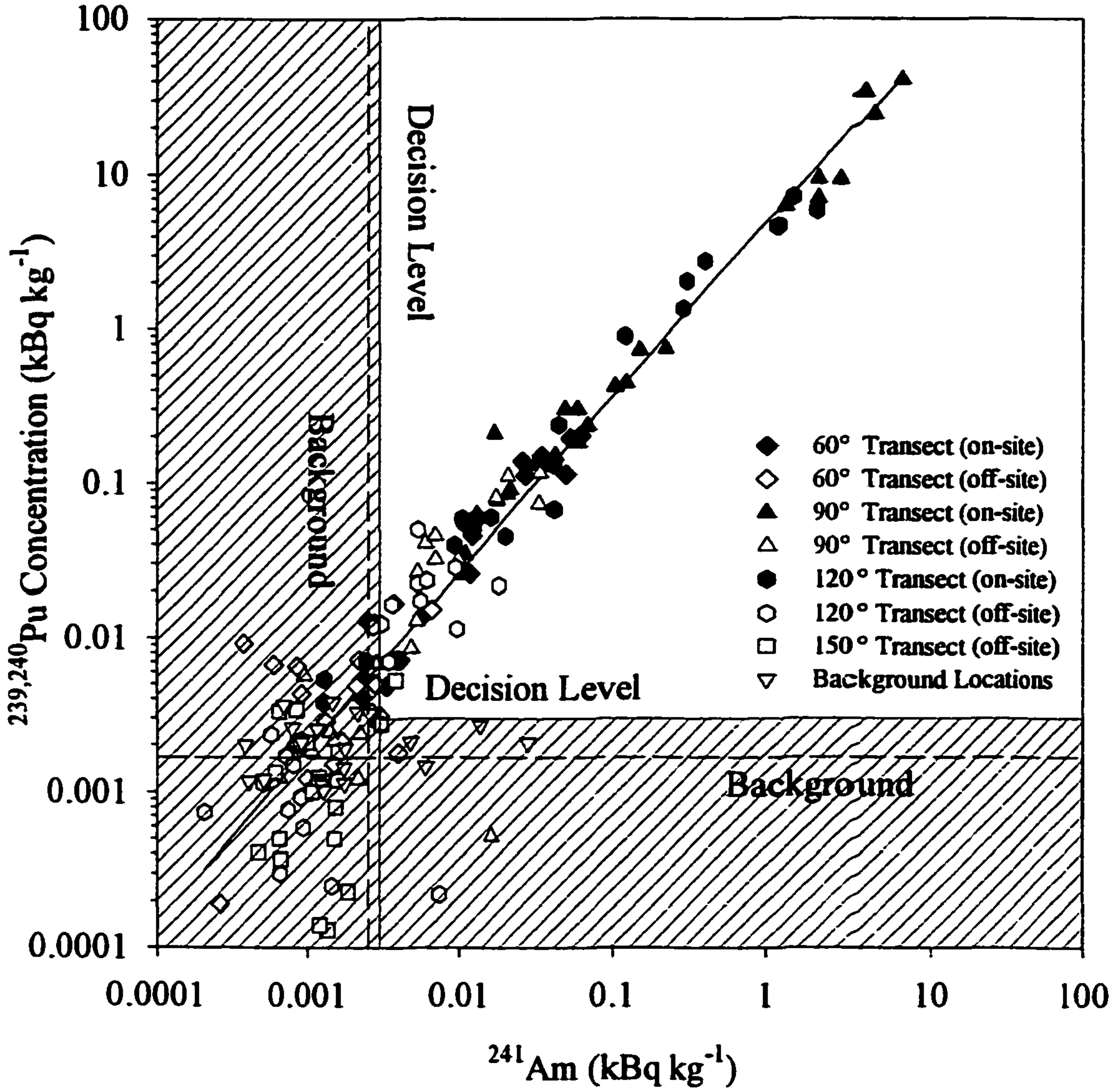

Figure 7. Correlation between ^{241}Am and ^{239,240}Pu in soil from depths of 0 to 3 cm at on-site and off-site locations east of the 903 pad around Rocky Flats.

Explanation

A - Ratio (0.011) estimated from USDOE (1995) data for the Colorado Front Range
B - Ratio (0.007) predicted during this study for areas around Rocky Flats

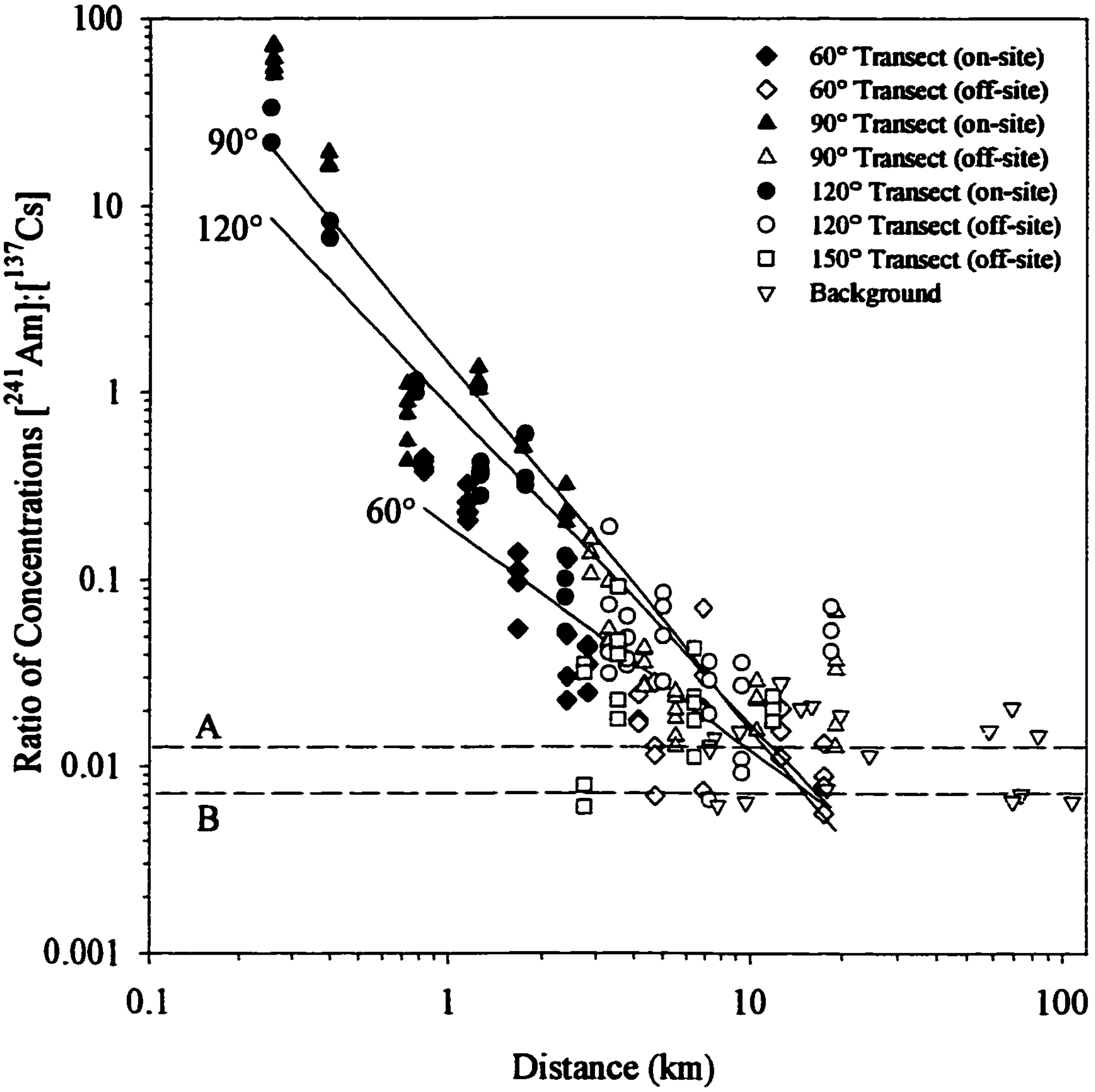

Figure 8. Ratios between concentrations of ^{241}Am and ^{137}Cs in soil from depths of 0 to 3 cm with distance from the 903 pad east of Rocky Flats. Estimated ratios between ^{241}Am and ^{137}Cs in global fallout are for 1994.

Explanation

A - Ratio (0.032) estimated by Miller and Michels (1973) for the Great Plains
B - Ratio (0.041) estimated from USDOE (1995) data for the Colorado Front Range
C - Ratio (0.026) estimated by Hodge et al.(1996) for Rocky Flats
D - Ratio (0.023) estimated by Lee et al.(1996) for Korean soils
E - Ratio (0.020) predicted during this study for areas around Rocky Flats

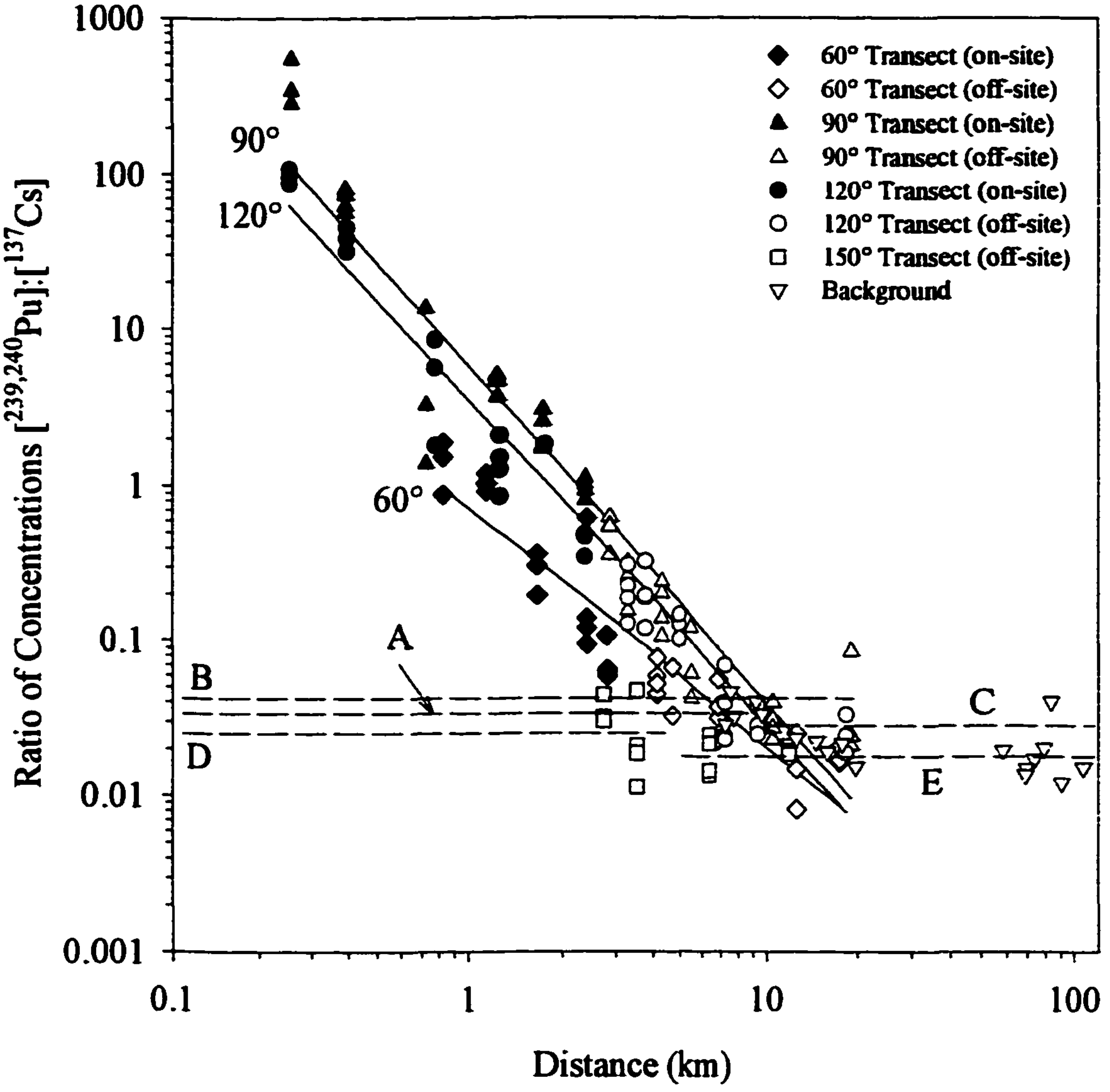

Figure 9. Ratios between concentrations of ^{239,240}Pu and ^{137}Cs in soil from depths of 0 to 3 cm with distance from the 903 pad east of Rocky Flats. Estimated ratios between ^{239,240}Pu and ^{137}Cs in global fallout are for 1994.

COMPARISON OF ^{137}CS AND ^{241}AM SOIL DEPOSITIONS ESTIMATED FROM IN-SITU AND LABORATORY GAMMA SPECTROSCOPY MEASUREMENTS

BACKGROUND

Soil depositions of ^{137}Cs and ^{241}Am at locations around the Rocky Flats Environmental Technology Site (Figure 1) were compared to discern the degree of correlation between estimates from in-situ and laboratory gamma spectroscopy measurements. The ratio of ^{239,240}Pu to ^{241}Am was then calculated to determine how well ^{241}Am soil depositions estimated from in-situ gamma spectroscopy measurements actually predicted soil levels of plutonium determined from radiochemical analyses of ^{239,240}Pu soil concentrations reported by Webb et al. (1994). This study follows-up on Ibrahim et al. (1995) who compared soil concentrations of ^{239,240}Pu and ^{241}Am determined with radiochemical analyses of samples from seven on-site locations at Rocky Flats, and preliminary ^{241}Am soil concentration estimates made with in-situ gamma spectroscopy at sampled locations. The work by Ibrahim et al. led to the conclusions that refining calibration procedures and accounting for environmental factors like soil moisture and rock content could reduce variability in soil deposition estimates for ^{241}Am, thus lowering detection limits. This comparison of soil deposition estimates for ^{241}Am made using in-situ and laboratory gamma spectroscopy and assessment of the ratio between ^{239,240}Pu and ^{241}Am depositions is also part of a larger study that characterized soil concentrations and depositions of ^{239}Pu (Ibrahim et al. 1996; Webb, et al. 1997), ^{137}Cs and ^{241}Am (Hulse et al. 1999), and ^{232}Th and ^{238}U (Hulse et al. 2000) around Rocky Flats.

Use of ^{241}Am soil deposition estimates from in-situ gamma spectroscopy measurements to predict levels of plutonium in soil is preferable because radiochemical separation and analysis for ^{239,240}Pu require considerable time and are labor intensive. In-situ measurements could reduce the need for soil sampling, custodial control of samples, sample preparation, and sample disposal. In addition, in-situ gamma spectroscopy measurements intrinsically average the detector response to radionuclides distributed uniformly over relatively large areas of 100 m^2 or more, potentially reducing overall variation in soil concentration and deposition estimates (Helfer and Miller, 1988; ICRU, 1994). Soil samples taken for laboratory analysis are generally from much less than a one meter square area.

Most of the ^{241}Am detected during this study was transported by wind from the 903 pad at Rocky Flats where oil contaminated with plutonium leaked from corroded drums into soil (Mongan et al. 1996a). A small portion of the ^{241}Am is from plutonium released during fires at Rocky Flats in 1957 (Mongan et al. 1996b) and 1969 (ChemRisk 1994). Hulse et al. (1999) analyzed soil samples in the laboratory with gamma spectroscopy and reported that mean concentrations of ^{241}Am in the top 3 cm of soil decreased in an easterly direction from more than 5.3 kBq kg^{-1} to background levels of 1.3 Bq kg^{-1} at measurement locations more than 7 km from the 903 pad. The rate of decrease was approximately proportional to the inverse square of distance. Webb et al. (1997) reported a similar trend in soil concentrations of ^{239,240}Pu determined with radiochemical analysis of splits from the samples that Hulse et al. (1999) analyzed. Hulse et al. concluded that soil concentrations of ^{241}Am and ^{239,240}Pu in the top 3 cm of soil around Rocky Flats are correlated, which was expected because Krey and Krajewski (1972), and Martel (1975) reported that the initial ratio between isotopes was nearly constant in plutonium used at Rocky Flats. By inference, the ratio between soil concentrations of ^{239,240}Pu and ^{241}Am can be used to characterize near-surface distributions of plutonium around Rocky Flats.

Global fallout from atmospheric testing of nuclear weapons is the most likely source of ^{137}Cs throughout the Rocky Mountain region (Rees et al. 1977 cited in Yoder 1977; Volchock and Chieco 1986). Krey et al. (1990) reported that peak deposition of ^{137}Cs from global fallout took place during 1964 at about the same time as major actinide releases from Rocky Flats. Johnson (1977) speculated that locally higher background levels of ^{137}Cs contamination might have come from Rocky Flats. However, Hulse et al. (1999) reported that, unlike ^{241}Am and ^{239,240}Pu, there is no apparent correlation between soil concentrations of ^{137}Cs and distance from Rocky Flats as should be expected if there had been a release of ^{137}Cs from the site. Hulse et al. also reported that the highest soil depositions of ^{137}Cs were in samples from more than 50 km south of Rocky Flats, as well as samples from on-site and off-site locations northeast of Rocky Flats, and off-site locations east and southeast of the 903 pad. We have speculated that increased soil deposition of ^{137}Cs occurred where drifted snow accumulated and where natural landscape features collected precipitation and dust.

Hulse et al. (1999) has shown that there are no statistically significant differences between the mean rates at which soil concentrations of ^{137}Cs, ^{239,240}Pu and ^{241}Am change with depth in soil around Rocky Flats, where concentrations of these radionuclides are above detection limits. This was expected because even though the source of ^{137}Cs is different from that for ^{239,240}Pu and ^{241}Am, deposition of these three radionuclides took place at about the same time. This relationship between ^{137}Cs, ^{239,240}Pu and ^{241}Am is fortuitous because the ^{137}Cs (661.7 keV) total absorption peak is easily detected with gamma spectroscopy. It was therefore possible to use in-situ gamma spectroscopy to screen for diminished surface soil concentrations and depositions of ^{137}Cs, thus identifying soil sample and measurement sites where erosion and other natural processes have occurred or where anthropogenic activities have taken place.

The relationship between ^{241}Am and ^{241}Pu in plutonium is well known. However, the efficacy of using soil depositions of ^{241}Am determined from in-situ gamma spectroscopy measurements as a surrogate for predicting soil concentrations and depositions of isotopic plutonium mixtures from the expected ratio of ^{239,240}Pu to ^{241}Am, has only begun to be tested; for example, at nuclear test sites in Australia (Burns et al. 1994) and at the Nevada Test Site (Fong and Alvarez 1997). Ibrahim et al. (1995) has indicated that if these predictions are to be accurate differences in attenuation of low-energy ^{241}Am photons (59.5 keV) between locations caused by heterogeneous soil moisture, rock content and other factors affecting soil density may have to be taken in to account. In addition, compensation for interference from ^{234}Th (63.3 keV) may be necessary. Objectives of this study were to:

- Characterize soil depositions of ^{137}Cs and ^{241}Am immediately east of Colorado's Front Range and around Rocky Flats using in-situ gamma spectroscopy measurements and compare results with estimates made from laboratory analyses of soil samples,
- Examine effects from varying amounts of soil moisture and rock mass on soil deposition estimates for ^{137}Cs and ^{241}Am made from in-situ gamma spectroscopy measurements,
- Determine how well soil depositions of ^{241}Am estimated from in-situ

gamma spectroscopy measurements predict soil depositions of ^{239,240}Pu.

MATERIALS AND METHODS

Area of Investigation and Measurement Locations

In-situ gamma spectroscopy measurements were initially made at the same 18 on-site and 24 off-site locations along four straight-line transects that are radial to the 903 pad at Rocky Flats and at 20 regional locations where soil samples were taken for laboratory analyses. Transects extended in the direction of prevailing winds with bearings of 60 °, 90 °, 120 ° and 150 ° relative to true North (Figure 1). Other in-situ measurements were made at 29 on-site and 45 off-site locations between the four transects as well as 15 additional regional locations.

Locations of soil samples and in-situ gamma spectroscopy measurements were chosen along transects at approximately exponentially increasing distances from the 903 pad. Locations of in-situ measurements between transects were also at exponentially increasing distances from the 903 pad; however, these were not along directional transects. A number of sample locations were relocated after assessing site access when in-situ measurements indicated levels of ^{137}Cs in soil profiles were diminished and therefore the possibility of soil disturbance existed, or when physical site characteristics indicated that normal vertical profiles of ^{137}Cs, ^{239,240}Pu and ^{241}Am in soil might have been altered. Locations where soil samples were taken and in-situ gamma spectroscopy measurements were made, site selection procedures, and factors that influenced site-selection were described in an earlier report by Stone et al. (1994). Both in-situ measurements and soil sampling were carried out between 1993 and 1996.

In-situ and Laboratory Measurements

In-situ gamma spectroscopy measurements used to estimate soil depositions were made with the detector at the center of ten-meter square macroplots. Soil samples were taken from up to four randomly selected one meter square microplots within each macroplot. Total absorption peaks used to estimate soil depositions of ^{137}Cs and ^{241}Am were measured in-situ with a coaxial n-type high purity germanium detector (model GC5022) manufactured by Canberra Industries, Inc.[21] An n-type detector was used to take advantage of that type of detector's intrinsic sensitivity to low energy X-rays and gamma rays; for example, 59.5 keV photons from ^{241}Am. Measurements were made with the detector mounted on a tripod one meter above the soil surface. The detector had a length of 63 mm, a diameter of 68 mm and an efficiency of 58% at 1.33 MeV relative to a 3 inch by 3 inch NaI detector. Hulse et al. (1999) has described laboratory procedures used to analyze soil samples, which, with the exception of sample preparation and instrument calibration, are similar to those used to analyze in-situ measurements.

The spectra of in-situ counts were typically recorded for 100 minutes in 4,096 channels over an energy range from 10 to 3,800 keV. Counts for the same energy spectra were recorded across 8,192 channels with the laboratory spectrometers. The Levenberg-Marquardt optimization algorithm (Marquardt, 1963) in SPSS' PeakFit[22] software was used to obtain least square fits of Gaussian distributions to the ^{137}Cs (661.7 keV) and ^{241}Am (59.5 keV) total absorption peaks after removing background effects from the spectra. Background-corrected Gaussians provided initial estimates for total absorption

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[21] Canberra Industries, 800 Research Parkway, Meriden, Connecticut 06450

[22] SPSS, Inc., PeakFit Version 4.0, ©1995, 2592 Kerner Blvd., San Rafael, California 94901

peak areas.

Interferences

Interferences from ^{214}Bi (665.4 keV) with the ^{137}Cs total absorption peak (661.7 keV) and ^{234}Th (63.3 keV) with the ^{241}Am total absorption peak (59.5 keV) were removed before deposition estimates were made. Net counts attributable to ^{214}Bi were estimated using the ^{214}Bi peak at 609 keV, which has a photon yield of 46.1% per transition, and adjusting for the yield from ^{214}Bi at 665.4 keV (1.6%). Likewise, net counts attributable to ^{234}Th were estimated using net counts from ^{234m}Pa at 1001 keV, which has a yield of 0.7% per transition but occurs in a portion of the spectrum that is devoid of interference. The photon yield from ^{234}Th at 63.3 keV (3.8%) was estimated by adjusting for the difference from the yield of ^{234m}Pa. We originally tried to use the ^{234}Th peak at 92.6 keV (5.4%) to estimate net counts from ^{234}Th at 63.3 keV; however, the spectra near the 92.6 keV total absorption peak is difficult to resolve because ^{228}Ac peaks in the ^{232}Th series occur at 90.0 keV (3.4%), 93.4 keV (5.6%) and 99.6 keV (1.3%). Miller et al. (1994) found that in most cases estimates for ^{234}Th made using the 1001 keV total absorption peak are similar to those obtained from the 92.6 keV total absorption peak, after accounting for measurement errors, which supports our decision to use the ^{234m}Pa total absorption peak to estimate ^{234}Th counts.

Soil deposition estimates from in-situ measurements for four background locations were excluded when calculating mean and median ^{241}Am background levels because these biased estimates to unreasonably negative levels. Negative background levels occur as statistical aberrations, particularly where there are large uncertainties in estimates of total absorption peak areas; however, the overall mean and median should be close to zero since ^{241}Am is an anthropogenic radionuclide and soil deposition of ^{241}Am from global fallout is normally close to or below detection limits.

Soil Deposition Estimates

A procedure developed by Beck et al. (1972) and Helfer and Miller (1988) was used with modifications described in ICRU Report 53 (1994) to estimate soil depositions from in-situ gamma spectroscopy measurements. Hulse (2001) has described how the calibration process was implemented for the gamma spectrometer used during this study. Soil depositions (kBq m^{-2}) were estimated from laboratory analyses using soil dry bulk densities to convert ^{137}Cs and ^{241}Am (Hulse et al. 1999), and ^{239,240}Pu soil concentrations (Webb et al. 1994) for 3 cm depth intervals to incremental depositions. Incremental soil depositions for each microplot were summed from the surface to a depth of 21 cm. The average of soil depositions for microplots within each macroplot provided the estimate for soil deposition. Soil concentrations of ^{137}Cs and, ^{241}Am in the 18 to 21 cm sample interval at most locations were at or below detection limits where soil samples were taken. Webb et al. (1994) reported that soil concentrations of ^{239,240}Pu were also close to or below the detection limit in the 18 to 21 cm sample interval.

Effects of Soil Moisture and Rock Content On Soil Deposition Estimates

Density variations caused by soil moisture had a negligible effect during laboratory data analyses because samples were oven dried before counting. Soil concentration estimates for ^{137}Cs and ^{241}Am (Hulse et al. 1999), as well as ^{239,240}Pu (Webb 1996), made from laboratory analyses were adjusted to account for the rock fraction. Soil deposition estimates based on laboratory analyses were calculated so that they were independent of the rock fraction, however.

Beck et al. (1972) reported that for photons with an energy of 60 keV, mass attenuation coefficients varied by less than 6% within a soil moisture range from 0% to 25%. Beck et al. also reported that at 650 keV the attenuation for the same soil moisture range varied by less than 4%. However, as described in ICRU Report 53 (1994), the relationship between mass attenuation and soil deposition estimates made from in-situ gamma spectroscopy measurements is non-linear. Local variations in soil density may therefore have an unpredictable effect on soil deposition estimates; however, the range of uncertainties in deposition estimates can be estimated.

Wet bulk densities were used to estimate soil depositions from in-situ measurements at most locations to account for variations in surface soil moisture and rocks, which undoubtedly affected in-situ measurements. The mean wet bulk density for all locations was used to estimate soil depositions where measured soil densities were unavailable. Soil densities, measured using procedures described in ASTM Method D2167-84 (1984), have been tabulated by Stone et al. (1994).

Relative effects of soil moisture and rocks on in-situ soil depositions at each location (i) were estimated by calculating differences between depositions determined using wet bulk densities (D_{wet}), and depositions determined using dry bulk densities (D_{dry}) and dry soil densities (D_{soil}). Dry bulk densities, which included effects from rocks and other debris, were measured after samples had been oven dried. Samples were then sieved with an ASTM #10 sieve and the density of the dry soil fraction passing through 2mm openings was measured.

The difference, $\Delta(i)_{wet}$, estimated with Equation 1,

$$\Delta(i)_{wet} = \frac{D_{dry} - D_{wet}}{D_{wet}}, \qquad (1)$$

is the fractional effect from soil moisture on soil deposition estimates. The difference, $\Delta(i)_{dry}$, estimated with Equation 2,

$$\Delta(i)_{dry} = \frac{D_{soil} - D_{wet}}{D_{wet}}, \qquad (2)$$

is the combined fractional effect from both soil moisture and rocks on soil deposition estimates. The difference, $\Delta(i)_{dry}$ - $\Delta(i)_{wet}$, is an estimate of the effect of rock mass on soil deposition estimates.

The relative effect of soil moisture on soil deposition estimates was determined by regressing $\Delta(i)_{wet}$ against the soil moisture fraction for each location. Likewise, the relative effect of rocks on soil deposition estimates was determined by regressing the difference, $\Delta(i)_{dry}$ - $\Delta(i)_{wet}$, against the rock fraction. Slopes of the regression lines provided estimates of rates at which soil deposition estimates varied with differences in soil moisture and rock content.

Lower Limits of Detection (LLD), Background and Decision Levels

Hulse et al. (1999) reported that the LLD for ^{137}Cs in the top 3 cm of soil ranged from 0.07 Bq for estimates made from laboratory analyses with an HPGe detector to 0.2 Bq for estimates made from measurements with a GeLi detector. Lower limits of detection (LLD) were estimated using a method described by Currie (1968). Although there is no reason to expect that contaminants from Rocky Flats contained ^{137}Cs, Hulse et al. (1999) used another statistic described by Borak and Kirchner (1995) to estimate the decision level for ^{137}Cs in laboratory samples (8.4 kBq m^{-2}). The decision level provided a guideline threshold for deciding if a sample contained radioactivity levels that were significantly higher than background levels.

Hulse et al. (1999) reported that the corresponding LLD estimates for ^{241}Am in soil samples analyzed with laboratory gamma spectroscopy ranged from 0.1 Bq when an HPGe detector was used to 0.3 Bq when using a GeLi detector. The decision level for ^{241}Am, estimated from laboratory analyses, was 0.35 kBq m^{-2} (Hulse et al. 1999).

Estimates of the LLD for soil depositions of ^{137}Cs or ^{241}Am determined with in-situ measurements were impractical because of the difficulty obtaining suitable "blanks." However, background levels of these radionuclides were estimated, thus enabling decision level estimates. Data for all locations were used to estimate median (95% sign confidence intervals; Gibbons, 1976; Hollander and Wolfe, 1973) ^{137}Cs background levels since global fallout is the only known source of ^{137}Cs around Rocky Flats. As was the case for laboratory analyses, background levels of ^{241}Am were characterized using soil deposition estimates for regional locations and depositions along transects at off-site locations believed to be unaffected by contaminants from Rocky Flats. As Hulse et al. (1999) reported for laboratory analyses, α levels for Type I errors were set to 0.20 when estimating decision levels for in-situ measurements.

Type I and Type II errors were set to 0.05 when establishing critical levels used to determine if a soil sample contained various analytes because it is possible to limit background levels and interference at relatively low cost in the laboratory. Type I errors for decision levels used to distinguish between samples containing environmental contaminants and those containing background levels estimated using results from in-situ measurements were set at 0.20. This level was selected because low resolution related to attenuation and variability associated with low count rates and interference from natural radionuclides cannot be controlled in the field.

Data Comparisons and Statistical Methods

Soil depositions of ^{137}Cs and ^{241}Am estimated from laboratory analyses were initially plotted as a function of estimates made from in-situ measurements to qualitatively evaluate relationships. Deposition estimates based on laboratory analyses were then regressed against estimates from in-situ measurements to quantify relationships. Pearson correlation coefficients were also calculated in most instances to assess the degree of linear relationship between the sets of deposition estimates. Soil depositions of ^{241}Am estimated from in-situ measurements were compared with depositions of ^{239,240}Pu determined from radiochemical analyses by Webb (1996) using a similar procedure. A similar procedure was also used to assess spatial variations.

The non-parametric Mann-Whitney test (McKean and Ryan 1977 in Minitab 1999) was used to compare median ^{137}Cs soil depositions estimated from in-situ measurements and laboratory analyses for each transect. This test was used because ^{137}Cs and ^{241}Am depositions estimated from laboratory analyses did not have normal distributions. While the data were partially paired, partial pairing was not taken into account.

Amounts of random variation in ^{137}Cs, ^{241}Am and ^{239,240}Pu soil deposition estimates attributable to microplots within macroplots and variations between macroplots were assessed using a mixed-effects model (Ott 1988). Variations between microplots within macroplots and variations between macroplots were expected to be about the same for background locations. Significantly larger variations between macroplots were expected than between microplots within macroplots where there were spatial variations in contamination as occurs around Rocky Flats.

Quality Assurance and Quality Control

In-situ counts were made at the same location on the Colorado State University (CSU) campus at the beginning of each day when field measurements were made to verify system operation. A similar count was made at the same location upon arrival back at CSU after completing daily measurements. Peak count rates in selected channels from the two daily spectra were compared with peak count rates in the same channels from previous counts to assure comparability. Count rates at the CSU location varied by less than one percent throughout this study. In addition, a ^{137}Cs source was used to verify the channel number of the 661.7 keV peak in the spectra prior to each in-situ measurement. No significant variations in the channel number of the ^{137}Cs total absorption peak were observed during this study. Stone et al. (1994) has described other quality assurance and quality control procedures used to assure selection of representative and reproducible field samples. Hulse et al. (1999) has described additional procedures that were used to assure data quality during laboratory analyses including recounts and analyses of split samples.

RESULTS AND DISCUSSION

Background Deposition Levels

The mean background level of ^{137}Cs estimated using in-situ measurements at 134 locations around Rocky Flats and along Colorado's Front Range was 4.3 ± 1.5 kBq m^{-2}. Soil depositions used to estimate ^{137}Cs background levels from in-situ measurements had an approximately normal distribution. Hulse et al. (1999) reported that the distribution of background levels estimated using laboratory analyses of samples from 61 of the same locations where in-situ measurements were made was approximately lognormal with a median (95% sign confidence interval) of 6.4 (5.4 to 8.4) kBq m^{-2}. To facilitate comparisons with results from laboratory analyses, the median background level for ^{137}Cs depositions estimated from the 135 in-situ measurements was 4.3 (1.2 to 7.8) kBq m^{-2}.

Deposition estimates for ^{137}Cs made from in-situ measurements (n = 96) and laboratory analyses (n = 33) of samples from locations within 10 km of the 903 pad at Rocky Flats both had approximately normal distributions. However, while deposition estimates for ^{137}Cs made from in-situ measurements (n = 39) at locations more than 10 km from the 903 pad had an approximately normal distribution, deposition estimates for ^{137}Cs made from laboratory analyses of samples from 22 of the same locations had an approximately log-normal distribution. About 36% of deposition estimates from laboratory analyses of samples from locations that were more than 10 km from the 903 pad were one standard deviation above the mean and one-half of those were more than two standard deviations above the mean, which explains the lognormal distribution for laboratory analyses of ^{137}Cs. None of the in-situ measurements were more than one standard deviation above the mean.

Background ^{241}Am soil deposition levels estimated from in-situ measurements at 39 locations beyond the influence of contamination from Rocky Flats had an approximately normal distribution and a mean of 0.8 ± 0.5 kBq m^{-2}. Hulse et al. (1999) reported that the distribution of ^{241}Am depositions estimated using laboratory analyses of 22 background samples from the same locations where in-situ measurements were made had an approximately lognormal distribution with a median (95% sign confidence interval) 0.20 (0.15 to 0.30) kBq m^{-2}. For comparison, the median background level for ^{241}Am determined from in-situ measurements at 42 locations, which included some of the same sites where soil samples were taken for laboratory analyses, was 0.8 (-0.1 to 1.9) kBq m^{-2}.

Decision Levels

The decision level for soil depositions of ^{137}Cs estimated using in-situ gamma spectroscopy measurements was 5.8 kBq m^{-2}, which is lower than the corresponding decision level estimate made from laboratory analyses (8.4 kBq m^{-2}). The lower decision level for ^{137}Cs estimated from in-situ measurements is related to both decreased variability and lower background levels, which may be attributable to insipient averaging by in-situ measurements of local heterogeneity in soil depositions within macroplots.

The decision level for soil depositions of ^{241}Am estimated from in-situ measurements was 1.3 kBq m^{-2}, which is significantly higher than the decision level estimated from laboratory analyses (0.35 kBq m^{-2}). The ^{241}Am total absorption peaks were recorded across twice as many channels during laboratory analyses as the corresponding total absorption peaks measured in-situ. It was therefore easier to discriminate between the ^{241}Am (59.5 keV) and ^{234}Th (63.3) peaks during laboratory analyses, leading to increased resolution and decreased variability. In addition to a higher mean background level for ^{241}Am, variability was also higher than that for estimates made with laboratory analyses. The higher decision level for ^{241}Am soil deposition estimates made from in-situ measurements is therefore attributable to both the higher background level and higher variability.

Soil Moisture and Rocks

The fractions of wet bulk soil density attributable to moisture and rocks have means of 0.12 (range from 0.02 to 0.26) and 0.12 (range from 0.03 to 0.24) respectively. Soil densities for the 0 to 3 cm and 3 to 6 cm depth intervals were only measured at a single location within each macroplot so information about variability within macroplots is unavailable. Differences in attenuation within and between locations attributable to variations in soil density caused by soil moisture and rocks were therefore a concern. This was particularly true for low energy photons from ^{241}Am as is apparent from the attenuation curves in Figure 2. Relative effects attributable to soil moisture, rocks and combined effects from both soil moisture and rocks in the top two 3 cm thick layers on deposition estimates were therefore compared. Effects from both individual layers were evaluated; however, the average density for the top two 3 cm thick layers provided the least variability.

The difference in soil deposition estimates for ^{137}Cs measurements (Figure 3) increased about 0.5% for each percent of mass attributable to soil moisture ($p<0.001$) and rocks ($p<0.001$). Corresponding differences in soil deposition estimates for ^{241}Am (Figure 4) were estimated to increase by about 0.7% and 0.6% respectively for each percent of mass attributable to soil moisture ($p<0.001$) and rocks ($p<0.001$).

Comparison of Soil Deposition Estimates

Soil depositions of ^{137}Cs determined from in-situ measurements are related to depositions determined from laboratory analyses (Figure 5) by the equation,

$$^{137}Cs_{lab} = 1.2 \cdot (^{137}Cs_{fld})^{0.9} . \qquad (3)$$

Although there is considerable variability in slope estimates as evinced by the r-squared parameter (51.3%), Equation 3 provides evidence that soil depositions of ^{137}Cs determined from in-situ and laboratory analyses are statistically comparable. The Pearson correlation coefficient describing the degree of linear relationship between the two sets of measurements in the linear scale is 0.59 (p-value < 0.001).

Soil depositions of ^{241}Am estimated from in-situ measurements are related to depositions estimated from laboratory analyses (Figure 6) by Equation 4,

$$^{241}Am_{lab} = 0.87 \cdot (^{241}Am_{fld})^{0.97} . \qquad (4)$$

Soil deposition estimates for all locations were plotted when making the initial comparison; however, regression analysis was limited to soil depositions that were above decision levels for both in-situ measurements and laboratory analyses. As was the case for ^{137}Cs, soil deposition estimates for ^{241}Am made using in-situ measurements and laboratory analyses are statistically comparable. The r-squared coefficient for the linear fit to log-transformed data is 76%. The Pearson correlation coefficient for the comparison in the linear scale is 0.92 (p-value<0.001).

Comparison of ^{137}Cs Deposition Estimates Along Transects

The Mann Whitney test provided insufficient evidence for significant differences between median soil depositions of ^{137}Cs estimated from in-situ measurements and laboratory analyses for the 60 °, 90 ° and 120 ° transects (Figure 7) at the 0.05 level of significance (Table 1). Pearson correlation coefficients tabulated in Table 1 indicate there are differences in the degree of linear relationship between estimates, however. Insufficient data were available to test for differences between soil depositions of ^{137}Cs estimated from in-situ measurements and laboratory analyses along the 150 ° transect.

Whicker and Schultz (1982) have reported that ^{137}Cs deposition levels are related to precipitation, which decreases with distance from the Rocky Mountains. A slight easterly decrease in soil deposition of ^{137}Cs was therefore expected because transects radiate in an easterly direction from Colorado's Front Range as well as the 903 pad. No evidence for regional trends in soil depositions was observed; however. There is significant variability between deposition estimates for individual samples as indicated by analysis of variance components for ^{137}Cs soil depositions estimated from laboratory analyses of microplot samples within macroplots (coefficient of variation = 0.21) and between macroplots (coefficient of variation = 0.24). Comparability of coefficients of variation for ^{137}Cs is an indication that variability in deposition estimates made from laboratory analyses is probably unrelated to measurement or sample location. This is consistent with our belief that ^{137}Cs from global fallout was deposited uniformly around Rocky Flats. We have speculated that most of the variability in soil depositions of ^{137}Cs is related to local factors like soil erosion and sedimentation rather than regional factors like weather patterns, where in-situ measurements were made and samples taken around Rocky Flats.

Comparison of ^{241}Am Deposition Estimates Along Transects

Soil depositions of ^{241}Am, D(X, θ) (kBq m^{-2}), which decrease with distance, X (km), from the 903 pad (Figure 8) along transects bearing θ degrees from true North at a rate, B(θ), (kBq m^{-2} km^{-1}), can be described by Equation 5,

$$D(X,\theta) = B(\theta) \cdot X^{M(\theta)}. \tag{5}$$

Exponents, M(θ), and constants, B(θ), are listed in Table 2 along with p-values for the estimates. Depositions estimated from in-situ measurements decreased at a slower rate with distance from the 903 pad than depositions estimated from laboratory analyses; however, differences in rates lack statistical significance when uncertainties are considered. Pearson correlation coefficients for the two sets of ^{241}Am deposition estimates along each transect provide evidence that in-situ and laboratory analyses are correlated except along the 60 ° transect, where soil deposition estimates made from laboratory analyses are below the LLD.

Variations in soil depositions of ^{241}Am between macroplots (coefficient of variation = 3.4) were significantly greater than variations between microplots within macroplots (coefficient of variation = 0.18). This difference is consistent with the idea that variability in ^{241}Am soil deposition levels increases with distance from the 903 pad at Rocky Flats. The variation of ^{241}Am soil depositions between microplots within macroplots is about the same as variations of ^{137}Cs depositions both within and between macroplots, which should be expected where local processes like erosion are the principal factors that modified deposition.

Comparison of ^{239,240}Pu and ^{241}Am Soil Depositions

The relationship between soil depositions of ^{239,240}Pu determined by Webb et al. (1994) for on-site locations from radiochemical analyses and depositions of ^{241}Am estimated from in-situ gamma spectroscopy measurements at the same locations where samples analyzed by Webb were taken (Figure 9) is described by Equation 6,

$$^{239,240}Pu_{lab} = 3.2 \cdot (^{241}Am_{fld})^{1.20} . \qquad (6)$$

If regression analysis used to estimate the slope and exponent for this equation was limited to soil deposition estimates at on-site locations along the 90 ° and 120 ° transects, the equation becomes,

$$^{239,240}Pu_{lab} = 5.4 \cdot [^{241}Am_{lab}]^{1.1} . \qquad (7)$$

Hulse et al. (1999) reported that a nearly identical equation could be used to describe the ratio between ^{239,240}Pu determined by Webb et al.. (1994) with radiochemical analyses of splits from the same samples used to estimate soil concentrations of ^{241}Am in the top 3 cm determined from laboratory gamma spectroscopy. The ratio of 5.5 predicted by Equation 7 is consistent with similar estimates by Ibrahim et al. (1996), Litaor and Allen (1996), Mongan et al. (1996a) and Litaor et al. (1998) as well as theoretical estimates made by adding temporal releases of plutonium reported by ChemRisk (1994) while subtracting losses due to radioactive decay.

It is apparent from the difference between Equation 6 and Equation 7 that soil depositions of ^{241}Am determined with in-situ gamma spectroscopy measurements during this study under-predict soil depositions of ^{239,240}Pu by about 30%. The cause of this bias is uncertain. It is unlikely that bias was introduced during radiochemical analyses because a known activity of chemically equivalent tracer was used by Webb et al. (1994) to verify analyte recovery. Use of a tracer significantly lowered uncertainties in ^{239,240}Pu soil concentration and deposition estimates. In addition, Webb et al. reported that alpha counting standards used during his analyses were traceable to NIST.

CONCLUSIONS

Judicious use of in-situ measurements could provide a cost-effective alternative to laboratory analyses for estimating soil concentrations of ^{241}Am. Depending on the purpose and objectives of a study, and requirements for accuracy in soil deposition levels it may be reasonable to use in-situ measurements of ^{241}Am in lieu of soil sampling in some instances to predict the distribution of ^{239,240}Pu. In-situ measurements will be particularly useful when rapid screening is necessary to characterize the spatial distribution and rates at which soil contamination change following accidental radionuclide releases from nuclear facilities.

The following conclusions have been drawn from this analysis:

- Soil deposition estimates for ^{137}Cs made using in-situ and laboratory gamma spectroscopy measurements were statistically comparable. There is no evidence for spatial variations of ^{137}Cs. The coefficients of variation between microplots within macroplots (0.21) and between macroplots (0.24) for soil depositions of ^{137}Cs were nearly the same, which is consistent with deposition of ^{137}Cs from global fallout. Initial deposition at each macroplot was relatively uniform but deposition has since been modified by local processes; for example, erosion.

- Soil deposition estimates for ^{241}Am made from in-situ and laboratory gamma spectroscopy measurements were also statistically comparable. Soil depositions of ^{241}Am estimated from in-situ measurements decreased from 195 kBq m^{-2} to background levels of 0.06 ± 0.7 kBq m^{-2} at a rate that is nearly proportional to distance from the 903 pad at Rocky Flats. Soil deposition estimates made from laboratory analyses decreased from about 170 kBq m^{-2}, but at a more rapid rate that was nearly proportional to the inverse square of distance. The coefficient of variation between soil depositions of ^{241}Am between microplots within macroplots (0.18) was significantly lower than that between macroplots (3.4), which should be expected where the source for plutonium and ^{241}Am was the 903 pad at Rocky Flats.

- Differences in soil deposition estimates increase by about 0.5% per combined percent of soil mass attributable to soil moisture and rocks at 661.7 keV ($p<0.001$) and 0.6 to 0.7% at 59.5 keV ($p<0.001$).

- Differences in decision level estimates for soil depositions of ^{137}Cs measured in-situ (5.8 kBq m^{-2}) and estimates from laboratory analyses (8.4 kBq m^{-2}) are attributed to mean background levels and variability in soil deposition estimates.

- The decision level for ^{241}Am estimated using situ measurements (1.3 kBq m^{-2}) was higher than the estimate made using deposition estimates from laboratory analyses (0.35 kBq m^{-2}). The higher decision level for in-situ measurements of ^{241}Am was attributable to the higher background level and increased variability caused by decreased resolution in estimates for total absorption peaks areas.

- The ratio of 5.5 between soil depositions of ^{239,240}Pu and ^{241}Am determined from in-situ measurements at 9 on-site locations closest to the 903 pad along the 90 and 120 transects is similar to that for soil depositions determined from laboratory analyses of soil samples. The ratio decreases when measurements with higher uncertainties from more distant locations are included.

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Table 8. Median soil depositions of ^{137}Cs along transects east of Rocky Flats determined from in-situ measurements and laboratory analysis of soil samples and Pearson correlation coefficients describing the degree of linear relationship between the two sets of measurements.

Transect	In-Situ Deposition (kBq m^{-2})			21 cm Deposition From Soil Sample Analysis (kBq m^{-2})			Pearson Correlation Coefficient	p-value
	n	Median	95%C.I.	n	Median	95%C.I.		
60°	25	4.7	3.6-5.7	11	4.1	3.4-5.2	0.58	0.059
90°	26	4.9	4.3-5.4	12	4.7	3.4-6.5	0.72	0.009
120°	33	3.5	3.3-4.3	13	4.1	2.8-4.6	0.68	0.010
150°	15	2.6	1.8-3.5	6	2.2	1.3-3.5	0.95	0.003

Table 9. Constants, exponents and p-values of estimates for equations describing variations of ^{241}Am depositions (kBq m^{-2}) in soil with distance from the 903 pad at Rocky Flats and Pearson correlation coefficients describing the degree of linear relationship between the two sets of measurements.

Transect	In-Situ Deposition				21 cm Deposition From Soil Sample Analysis				Pearson Correlation Coefficient	p-value
	Exponent M(Θ)	p-value	Constant B(Θ) kBq m^{-2}	p-value	Exponent M(Θ)	p-value	Constant B(Θ) kBq m^{-2}	p-value		
60°	-0,17 ± 0,11	0,02	1,50 ± 1,20	0,14	-0,69 ± 0,19	0,005	0,98 ± 0,04	<0,001	0,49	0,12
90°	-1,10 ± 0,20	<0,001	7,69 ± 1,28	<0,001	-1,45 ± 0,24	<0,001	6,60 ± 0,27	<0,001	0,98	<0,001
120°	-1,07 ± 0,16	<0,001	3,07 ± 1,22	<0,001	-1,48 ± 0,20	<0,001	4,47 ± 0,16	<0,001	0,99	<0,001
150°	-0,92 ± 0,27	0,06	2,70 ± 1,60	0,007	-0,46 ± 0,20	0,087	0,40 ± 0,03	0,007	0,91	0,034

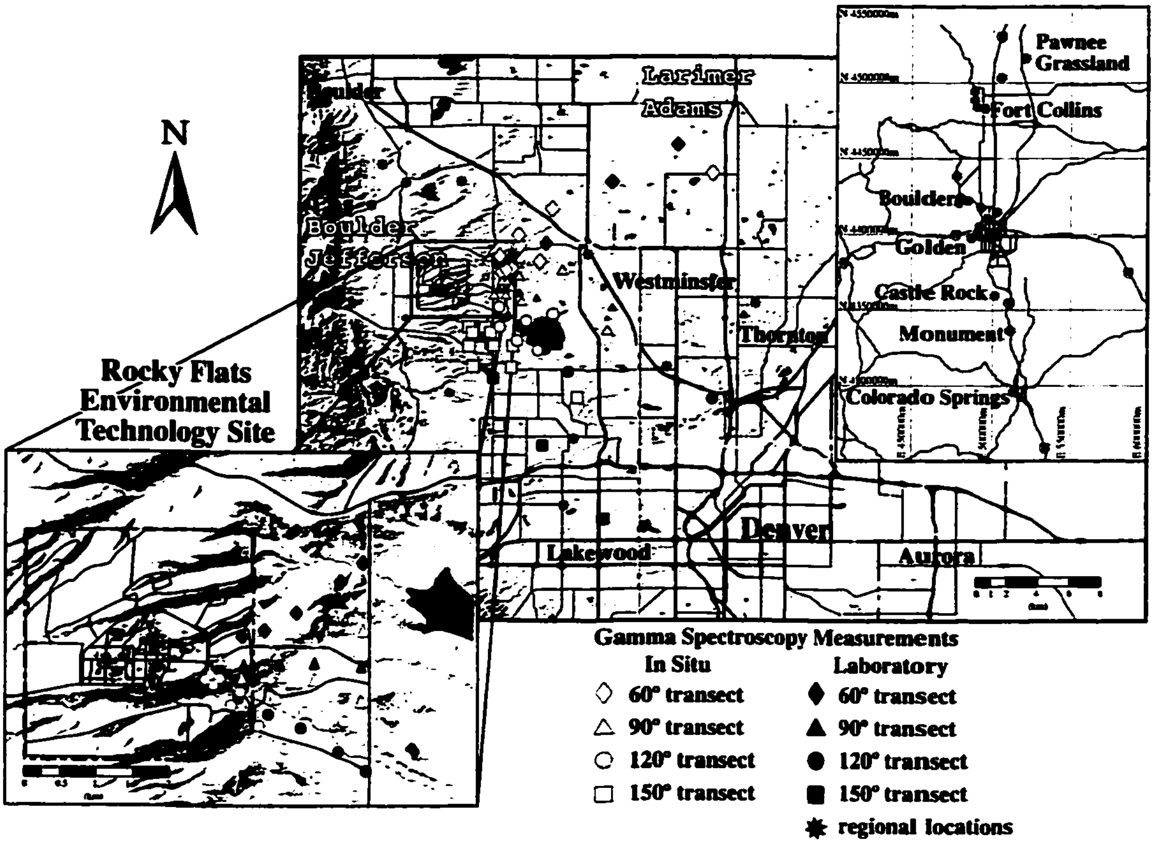

Figure 10. In-situ Gamma Spectroscopy and sample locations around Rocky Flats in Jefferson, Boulder and Adams Counties, Colorado and along Colorado's Front Range.

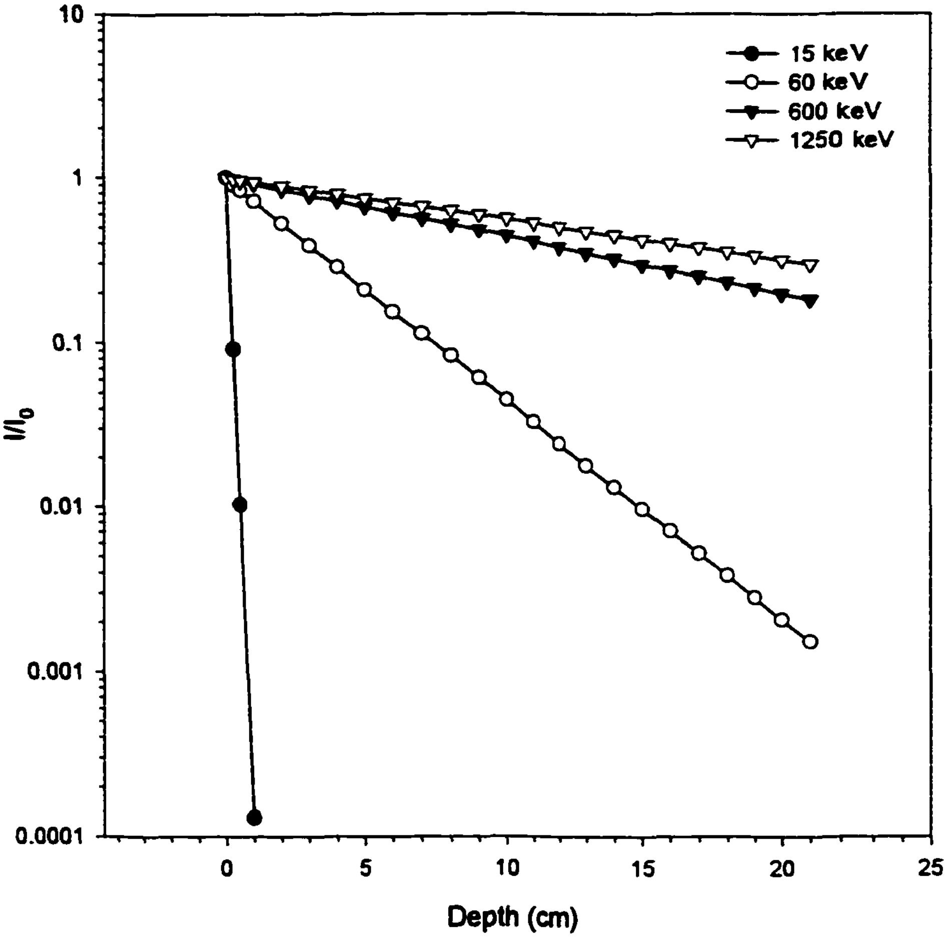

Figure 11. Attenuation of photons from a point source buried in Rocky Flats Soil Standard Number 1 (NIST 1980).

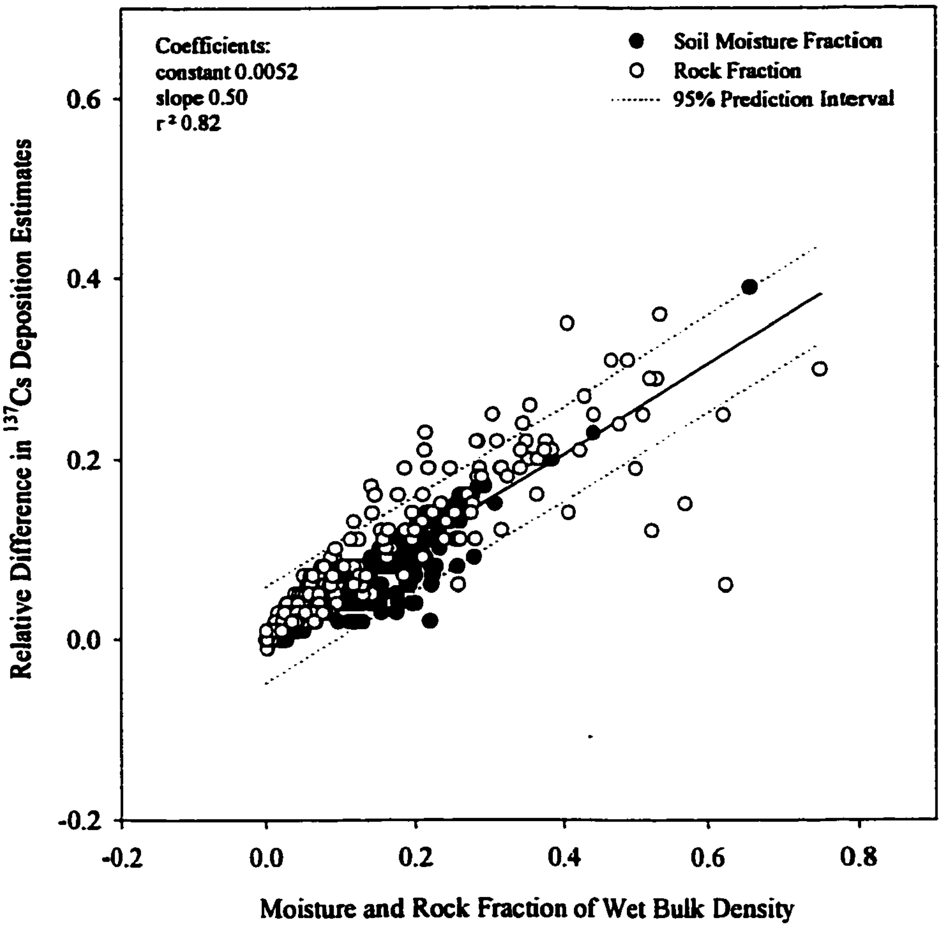

Figure 12. Fractional changes in soil deposition estimates of ^{137}Cs caused by moisture and rock mass.

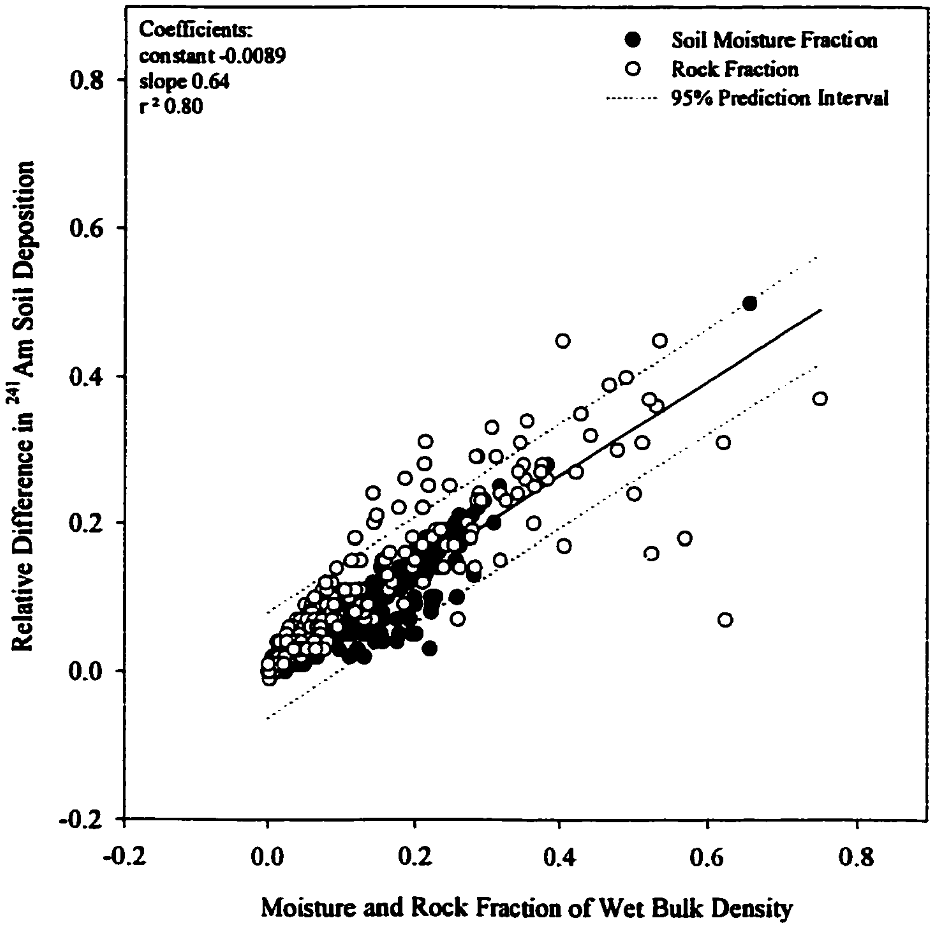

Figure 13. Fractional changes in soil deposition estimates of ^{241}Am caused by moisture and rock mass.

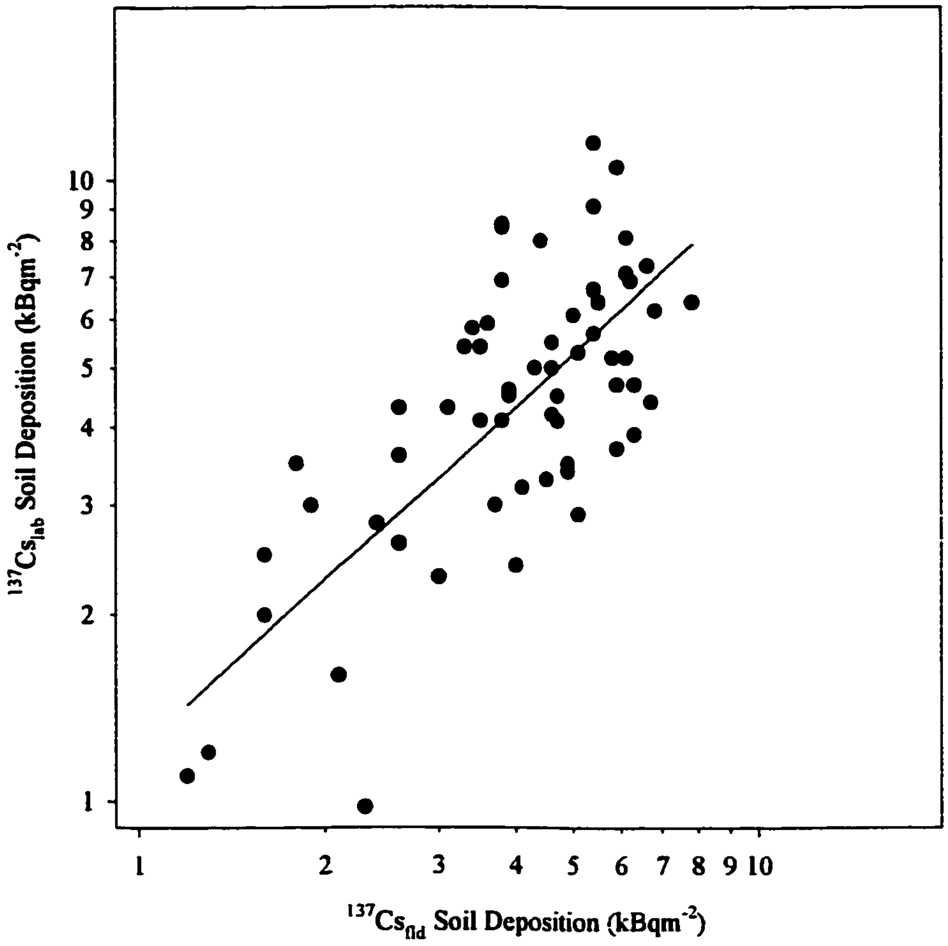

Figure 14. Comparison of ^{137}Cs soil depositions around Rocky Flats and along Colorado's Front Range determined using in-situ gamma spectroscopy measurements with depositions estimated from soil sample analysis with laboratory gamma spectroscopy.

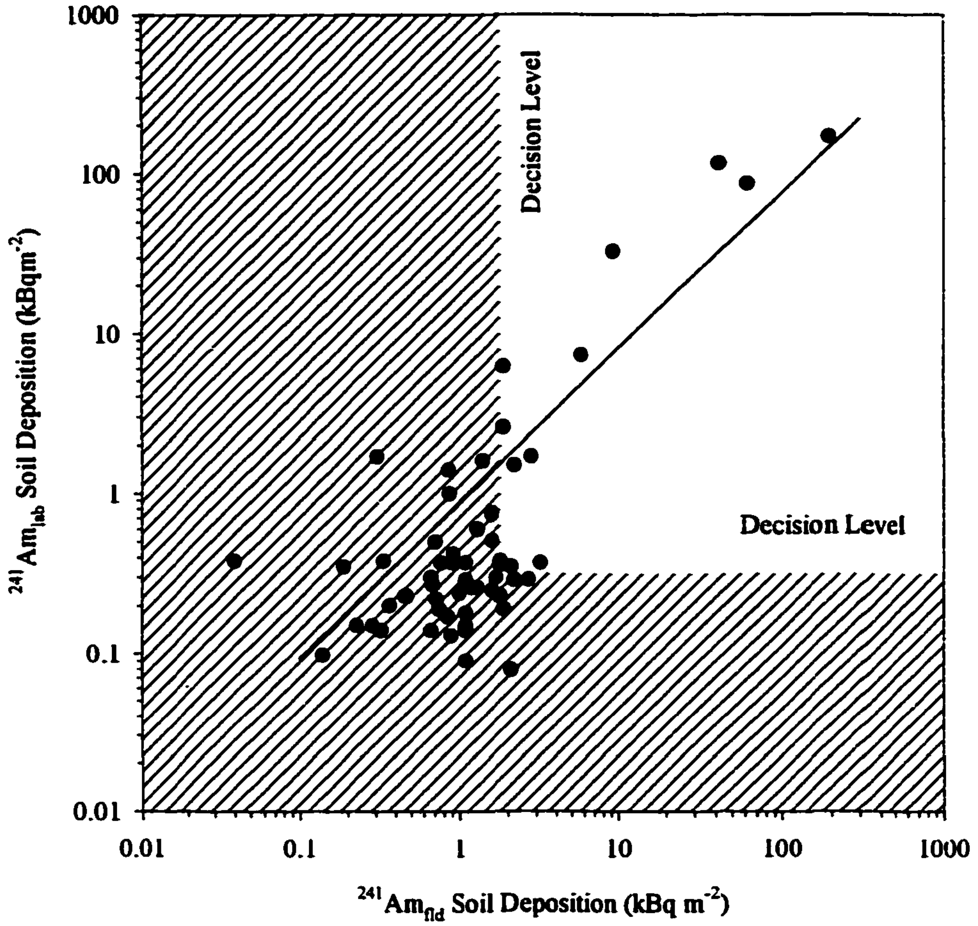

Figure 15. Comparison of ^{241}Am soil depositions around Rocky Flats and along Colorado's Front Range determined using in-situ gamma spectroscopy measurements with depositions estimated from soil sample analysis with laboratory gamma spectroscopy.

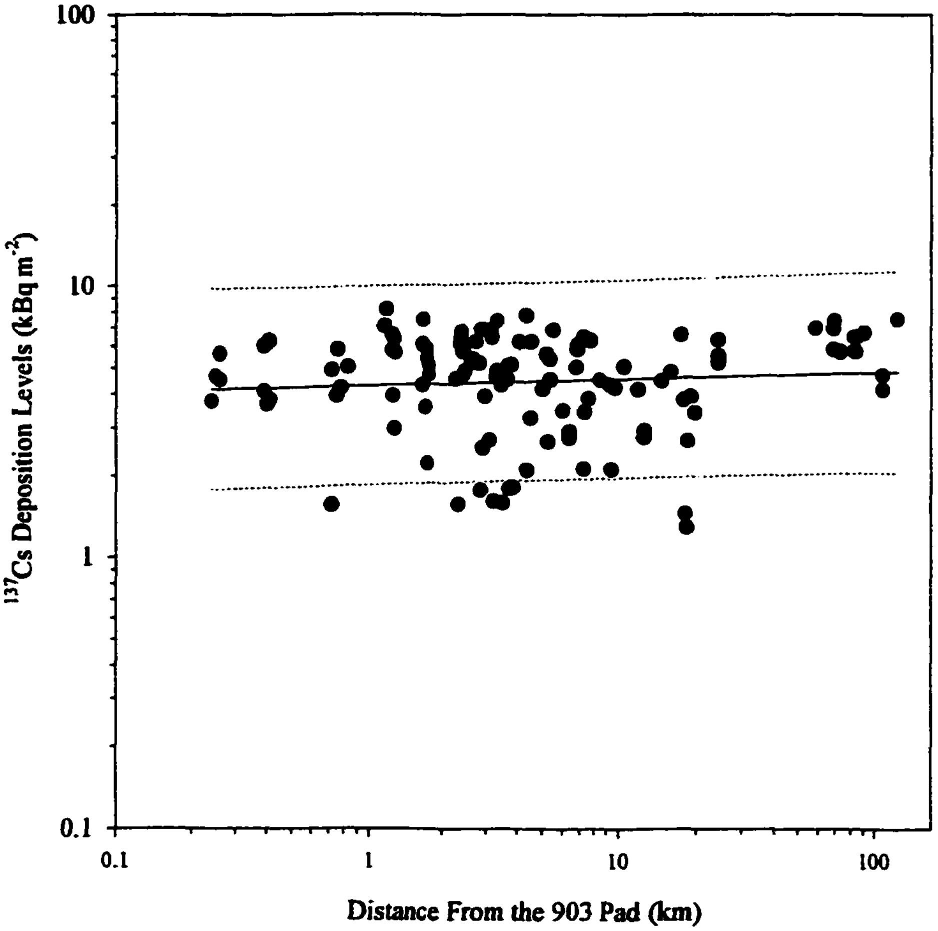

Figure 16. Variations of ^{137}Cs soil depositions around Rocky Flats and along Colorado's Front Range determined using in-situ gamma spectroscopy measurements with distance from the 903 pad.

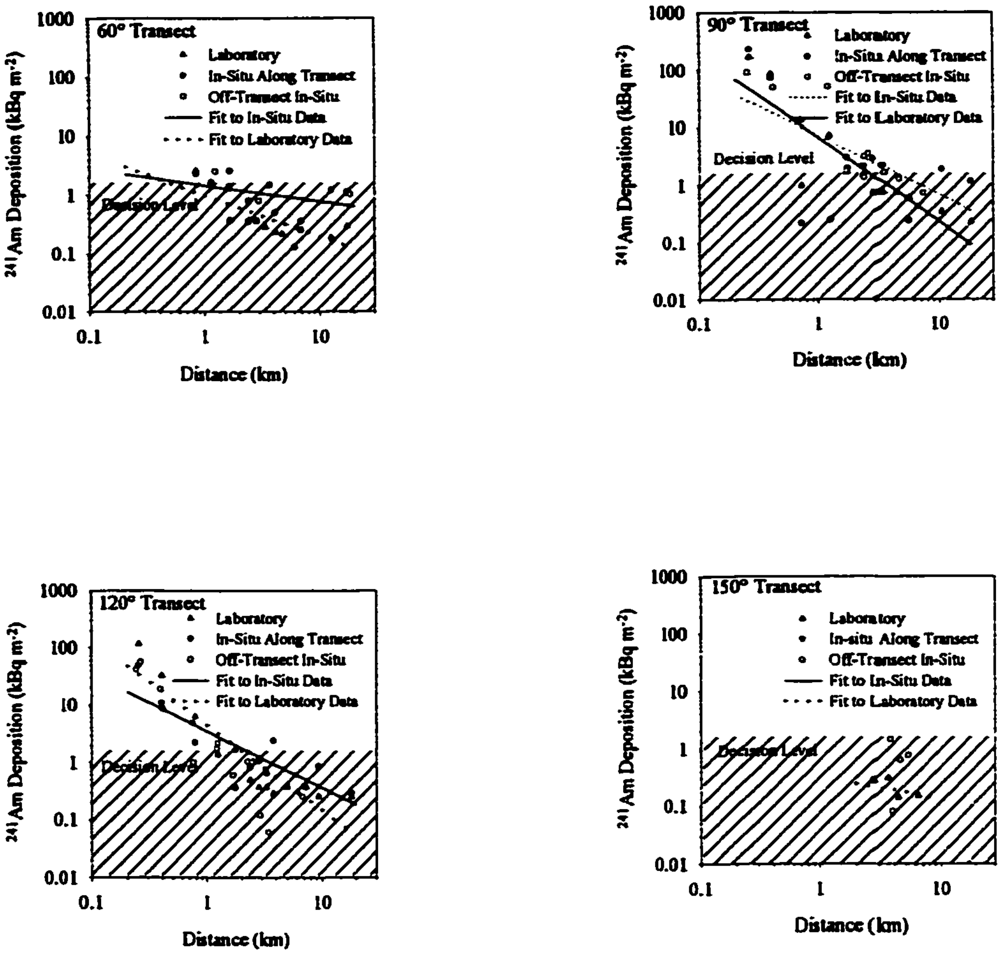

Figure 17. Variations in soil depositions of ^{241}Am estimated with in-situ gamma spectroscopy measurements and laboratory soil sample analysis with gamma spectroscopy along transects east of the 903 pad at the Rocky Flats Environmental Technology Site.

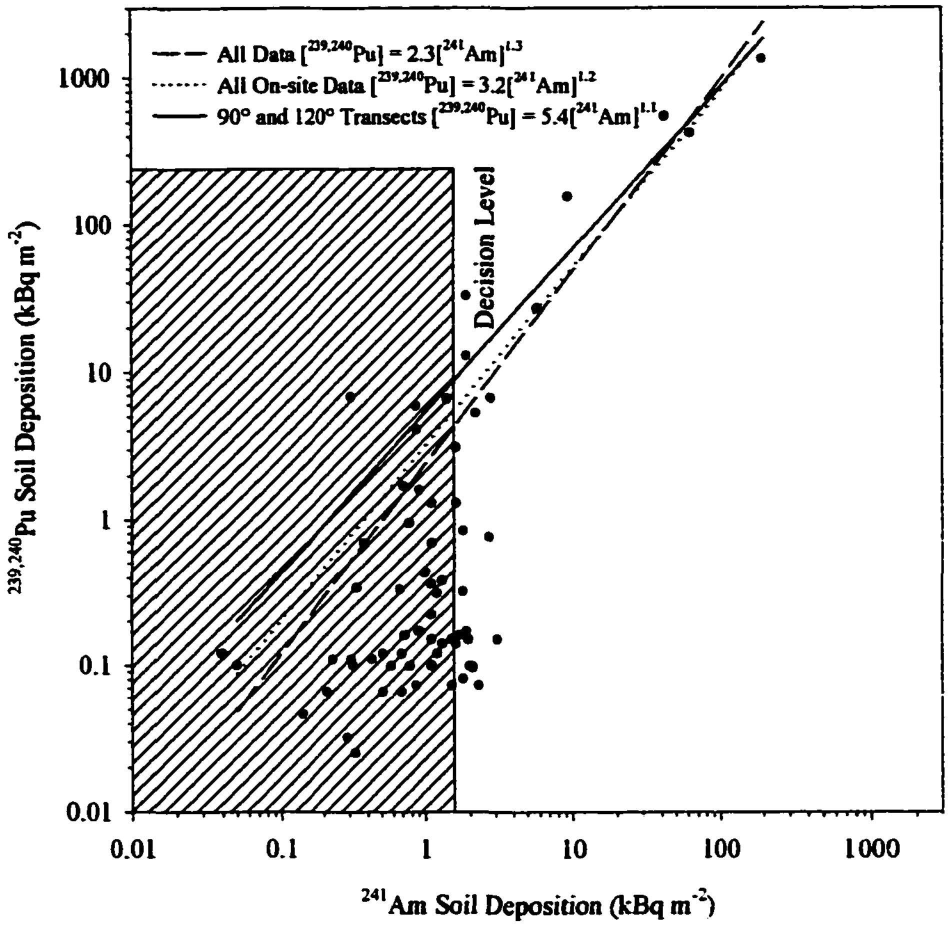

Figure 18. Comparison of ^{239,240}Pu soil depositions determined with radiochemical analyses with ^{241}Am soil deposition determined with in-situ gamma spectroscopy measurements.

SUMMARY, CONCLUSIONS AND RECOMMENDATIONS

The Radioecology Group at Colorado State University (CSU) has repeatedly sampled and analyzed soil as well as plants and animal tissues for americium and plutonium in an effort to understand transport processes as well as the environmental impact of these radionuclides on humans and other biota around the Rocky Flats Environmental Technology Site. In addition to developing a historical record of soil concentrations and depositions, the CSU effort has focused on the potential impact on human health and the environment.

Analytical models that may be developed in the future using information from the historical record that has been developed at CSU, of which this dissertation is now a part, provide a basis for understanding the fate and transport of americium and plutonium released from Rocky Flats that may be useful for predicting the future impact of these radionuclides on public health or on the environment. In addition, the historical record may provide a way to retrospectively evaluate any impact that might otherwise have gone unnoticed.

Research presented in this dissertation is the culmination of a desire to understand how ^{241}Am moved through the environment around Rocky Flats and the relationship between ^{241}Am and ^{239,240}Pu. That desire has grown to include an aspiration to comprehend the effects from mixed radiation fields, which is beyond the scope of this dissertation but an important component of public health studies involving radiation effects that should be addressed when analytic models are developed.

SUMMARY

Soil concentrations of naturally occurring ^{232}Th, ^{226}Ra and ^{40}K were estimated along with soil concentrations and depositions of anthropogenic ^{137}Cs from global fallout and ^{241}Am released from Rocky Flats using analyses of soil samples with gamma spectroscopy. Soil concentrations and depositions of ^{137}Cs and ^{241}Am were also estimated from in-situ gamma spectroscopy measurements. Two questions were answered by analyzing data from these measurements:

- Are there statistically significant differences between soil depositions of ^{137}Cs and ^{241}Am estimated from in-situ gamma spectroscopy measurements and laboratory analyses of soil samples?
- Are there statistically significant trends in background levels of ^{232}Th, ^{226}Ra and ^{40}K study in the study area?

Soil concentrations of ^{241}Am and ^{137}Cs in 21 cm deep soil profiles and soil depositions determined from laboratory gamma spectroscopy analyses were compared with corresponding concentrations and depositions of ^{239,240}Pu determined with radiochemical analyses to determine how soil distribution patterns for these radionuclides are related. Distributions of ^{137}Cs in soil profiles were also compared throughout the study area to determine how soil disturbance affected depth profiles. Two questions were answered by these comparisons.

- Are there significant differences between functions describing depth distributions of ^{137}Cs, ^{241}Am and ^{239,240}Pu in soil profiles where soil disturbance is absent?

- How does soil disturbance affect distributions of ^{137}Cs with depth?

Soil concentrations of ^{241}Am in the top 3 cm of soil determined from laboratory analyses with gamma spectroscopy and ^{239,240}Pu, also in the top 3 cm of soil, determined with radiochemical analyses were compared. This comparison was made to decide if plutonium concentrations can be predicted from ^{241}Am concentrations. A similar comparison was made between soil depositions determined from in-situ gamma spectroscopy measurements and soil depositions of ^{239,240}Pu. The questions that these comparisons addressed were:

- Is there a consistent, statistically significant relationship between soil concentrations of ^{241}Am in the top 3 cm of soil estimated from laboratory gamma spectroscopy measurements and ^{239,240}Pu concentrations, also in the top 3 cm of soil, determined with radiochemical analyses?

- Is there a consistent, statistically significant relationship between soil depositions of ^{241}Am estimated from in-situ gamma spectroscopy measurements and ^{239,240}Pu depositions determined with radiochemical analyses?

CONCLUSIONS

The following conclusions are drawn from this study:

- Soil deposition estimates for both ^{137}Cs and ^{241}Am made from in-situ gamma spectroscopy measurements and laboratory analyses are statistically comparable.

- The soil concentration of ^{232}Th, which has an overall mean of 96 Bq kg^{-1}, decreases in a northerly direction from Colorado Springs to Fort Collins at about

0.4 Bq kg^{-1} km^{-1}. The soil concentration of ^{40}K, which has a mean of 720 Bq kg^{-1}, also decreases in a northerly direction from Colorado Springs to Fort Collins at about 4 Bq kg^{-1} km^{-1}. The mean soil concentration of ^{226}Ra was 63 Bq kg^{-1}; however, there was no distinguishable spatial trend in soil concentrations of ^{226}Ra.

- There are no statistically significant differences between slopes of functions describing soil distributions of ^{137}Cs, ^{241}Am and ^{239,240}Pu with depth in 21 cm soil profiles at undisturbed sample locations.
- Soil concentrations of ^{137}Cs were uniform in 3 cm depth intervals to a total depth of 21 cm at sample locations where there was soil disturbance.
- The ratio between soil concentrations of ^{239,240}Pu and ^{241}Am in the top 3 cm of soil was determined to be 5.5 at four locations closest to the 903 pad on-site at Rocky Flats.
- The ratio between soil depositions of ^{239,240}Pu and ^{241}Am determined from in-situ gamma spectroscopy measurements was 5.4 for on-site measurement and soil sample locations along the 90° and 120° transects.

RECOMMENDATIONS

Soil action levels for ^{239,240}Pu around Rocky Flats are a current subject of public debate and comment. Once action levels have been specified, soil concentrations and depositions of ^{241}Am and ^{239,240}Pu described in the third and fourth chapters of this dissertation may complement similar data obtained by others and provide a basis for mixed field dosimetry that will be used to determine if and where remediation is necessary.

Research described in this dissertation also has implications for land use and the need to develop a vegetation management plan to minimize effects from soil resuspension in contaminated areas around Rocky Flats. More than 60% of ^{241}Am and ^{239,240}Pu soil depositions are retained in the top 3 cm of soil. There has been a decision to maintain the buffer zone at the site as a wildlife refuge to be administered by the U.S. Fish and Wildlife Service. Soil concentrations and depositions of ^{241}Am and ^{239,240}Pu that have been reported in this dissertation may again be useful for testing the basis for these land use decisions.

There are also other uses for results from this study. For example, future regional energy requirements may reduce public opposition to nuclear power generating facilities. This study provides preliminary information that can be used during development of environmental impact statements that will necessarily precede construction of a nuclear power plant. While this use is politically unacceptable at present, future requirements may dictate a change in attitudes toward nuclear power out of necessity.

Appendix A

Soil Concentrations and Depositions of ^{241}Am, ^{137}Cs and ^{239,240}Pu Estimated From Laboratory Gamma Spectroscopy Measurements

Location	UTM East Coordinate (m)	UTM North Coordinate (m)	Depth Interval (cm)	Number of Samples	^{241}Am Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{137}Cs Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{239,240}Pu Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	
AX1	483658	4415301	0.0 - 0.3	4	5300	390	4	81	2	2	10,600	11,900	
			0.0 - 3.0	6	4900	1300	6	81	21	3	33,100	8,200	
			3.0 - 6.0	4	1500	810	4	41	10	4	10,700	4,000	
			6.0 - 9.0	4	350	230	4	10	2	4	5,700	4,900	
			9.0 - 12.0	4	200	110	4	5	2	2	2,500	640	
			12.0 - 15.0	4	140	120	4	3	3	1	1,600	43	
			15.0 - 18.0	4	110	92	4	2	1	1	740	65	
			18.0 - 21.0	4	74	84	4	1	1	4	510	470	
		Deposition	0.0 - 21.0		170	60	kBq m^{-2}	3.6	0.8	kBq m^{-2}	1,330	610	kBq m^{-2}
AX2	483792	4415305	0.0 - 0.3	4	1500	190	4	86	7	4	6100	1700	
			0.0 - 3.0	4	2100	640	4	120	29	4	8000	1600	
			3.0 - 6.0	6	820	200	6	54	7	3	5300	790	
			6.0 - 9.0	4	190	66	4	18	9	4	870	170	
			9.0 - 12.0	4	59	43	4	6	3	4	460	340	
			12.0 - 15.0	4	43	16	4	2	3	4	280	190	
			15.0 - 18.0	4	37	22	4	2	2	4	210	180	
			18.0 - 21.0	4	21	12	4	1	2	4	110	50	
		Deposition	0.0 - 21.0		86	19	kBq m^{-2}	5.4	0.3	kBq m^{-2}	350	140	kBq m^{-2}
AX3	484124	4415316	0.0 - 0.3	5	9	4	5	12	5	6	120	130	
			0.0 - 3.0	5	14	4	5	20	4	6	79	68	
			3.0 - 6.0	5	24	18	5	8.5	2.7	5	60	58	
			6.0 - 9.0	4	3	2	4	2.0	2.1	5	22	23	
			9.0 - 12.0	4	1.6	1.2	4	0.4	0.2	4	7.6	5.5	
			12.0 - 15.0	4	0.9	0.3	4	0.3	0.2	2	6.9	6.5	
			15.0 - 18.0	4	0.9	0.7	4	0.5	0.3	4	1.1	0.7	
			18.0 - 21.0	4	0.2	0.2	4	0.4	0.1	3	0.6	0.1	
		Deposition	0.0 - 21.0		1.0	0.5	kBq m^{-2}	0.8	0.04	kBq m^{-2}	3.5	1.8	kBq m^{-2}
AX4	484642	4415326	0.0 - 0.3	5	120	9	5	94	20	6	710	170	
			0.0 - 3.0	4	150	51	4	130	53	4	590	180	
			3.0 - 6.0	4	54	25	4	55	33	5	140	89	
			6.0 - 9.0	4	14	14	4	14	15	5	56	30	
			9.0 - 12.0	4	5.8	2.3	4	5.5	2.8	6	39	33	
			12.0 - 15.0	6	3.3	2.7	6	2.8	2.0	7	16	26	
			15.0 - 18.0	4	2.4	2.1	4	1.3	0.8	4	10	5.4	
			18.0 - 21.0	4	3.0	0.9	4	1.2	0.7	4	5.5	3.0	
		Deposition	0.0 - 21.0		6.7	1.7	kBq m^{-2}	6.1	1.5	kBq m^{-2}	30	7.2	kBq m^{-2}

Location	UTM East Coordinate (m)	UTM North Coordinate (m)	Depth Interval (cm)	Number of Samples	^{241}Am Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{137}Cs Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{239,240}Pu Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	
AX5	485127	4415346	0.0 - 0.3	3	51	11	3	110	17	4	170	58	
			0.0 - 3.0	4	60	8	4	110	17	4	260	58	
			3.0 - 6.0	4	18	5	4	37	9	4	59	35	
			6.0 - 9.0	4	3.5	2.5	4	9.0	3.6	4	22	8.9	
			9.0 - 12.0	5	1.4	0.9	5	4.6	3.0	4	6.0	4.5	
			12.0 - 15.0	5	1.9	1.5	5	3.1	1.7	4	4.0	2.5	
			15.0 - 18.0	4	1.7	1.3	4	1.4	1.4	4	2.6	1.6	
			18.0 - 21.0	4	1.3	0.5	4	1.2	1.3	4	2.0	1.8	
		Deposition	0.0 - 21.0		1.7	0.2	kBq m^{-2}	3.3	0.4	kBq m^{-2}	6.7	1.7	kBq m^{-2}
AX6	485785	4415302	0.0 - 0.3	4	34	9	4	110	33	6	130	22	
			0.0 - 3.0	4	29	10	4	120	18	4	110	32	
			3.0 - 6.0	4	12	12	4	52	57	4	55	49	
			6.0 - 9.0	4	3.9	3.3	4	12	12	4	12	13	
			9.0 - 12.0	4	2.4	1.1	4	4.9	5.5	5	5.8	6.3	
			12.0 - 15.0	5	1.6	1.2	5	2.1	1.2	5	0.5	0.5	
			15.0 - 18.0	5	0.9	0.8	5	1.2	1.2	5	0.6	0.8	
			18.0 - 21.0	4	1.4	1.0	4	0.9	0.7	4	0.8	0.5	
		Deposition	0.0 - 21.0		1.5	0.6	kBq m^{-2}	5.3	2.9	kBq m^{-2}	5.3	2.6	kBq m^{-2}
BX1	484104	4415741	0.0 - 0.3	4	61	21	4	150	57	3	300	85	
			0.0 - 3.0	4	48	8	4	120	25	3	150	41	
			3.0 - 6.0	4	17	5	4	38	4	3	74	45	
			6.0 - 9.0	4	7.2	2.0	4	20	6	3	49	5	
			9.0 - 12.0	4	4.5	1.0	4	10	2	4	31	15	
			12.0 - 15.0	6	1.7	1.6	6	5.1	3.2	4	16	14	
			15.0 - 18.0	4	3.0	1.9	4	4.2	5.1	3	15	8	
			18.0 - 21.0	5	2.0	1.0	5	1.9	2.0	3	64	71	
		Deposition	0.0 - 21.0		2.6	0.2	kBq m^{-2}	6.1	0.7	kBq m^{-2}	12	3.3	kBq m^{-2}
BX2	484423	4415827	0.0 - 0.3	5	40	6	5	180	29	6	120	53	
			0.0 - 3.0	5	35	16	5	140	36	4	150	39	
			3.0 - 6.0	4	7.2	4.3	4	29	17	4	26	26	
			6.0 - 9.0	4	2.2	1.3	4	8.5	5.6	4	11	7	
			9.0 - 12.0	4	1.3	0.8	4	4.5	3.1	4	6.9	5.9	
			12.0 - 15.0	3	0.7	0.5	3	4.2	3.3	4	3.9	3.7	
			15.0 - 18.0	3	1.8	1.1	3	3.1	3.1	2	2.5	1.5	
			18.0 - 21.0	3	1.1	0.4	3	1.8	2.1	4	1.5	1.6	
		Deposition	0.0 - 21.0		1.7	0.7	kBq m^{-2}	6.4	2.0	kBq m^{-2}	6.6	2.1	kBq m^{-2}

Location	UTM East Coordinate (m)	UTM North Coordinate (m)	Depth Interval (cm)	Number of Samples	^{241}Am Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{137}Cs Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{239,240}Pu Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	
BX3	484877	4416074	0.0 - 0.3	4	9.2	7.3	4	89	60	5	100	140	
			0.0 - 3.0	4	8.7	3.9	4	93	43	4	25	16	
			3.0 - 6.0	4	4.8	1.8	4	71	29	4	12	6	
			6.0 - 9.0	5	2.1	1.0	5	17	9	3	6.8	4.6	
			9.0 - 12.0	5	1.9	1.0	5	4.9	0.9	4	1.6	0.8	
			12.0 - 15.0	4	1.1	1.5	4	3.6	2.7	4	0.7	0.3	
			15.0 - 18.0	4	1.3	1.2	4	2.2	1.8	3	0.4	0.2	
			18.0 - 21.0	4	1.5	0.9	4	1.0	0.4	2	0.9	0.9	
		Deposition	0.0 - 21.0		0.4	0.1	kBq m^{-2}	3.5	0.2	kBq m^{-2}	0.8	1.0	kBq m^{-2}
BX4	485488	4416513	0.0 - 0.3	4	3.1	1.2	4	99	8	5	17	8	
			0.0 - 3.0	4	5.3	3.9	4	98	21	4	23	21	
			3.0 - 6.0	4	2.2	1.1	4	26	9	3	3.4	1.8	
			6.0 - 9.0	4	0.4	0.6	4	9.3	7.0	3	1.5	1.4	
			9.0 - 12.0	4	0.9	0.6	4	4.6	3.9	4	4.9	8.6	
			12.0 - 15.0	4	1.2	0.5	4	2.7	2.3	3	0.5	0.4	
			15.0 - 18.0	4	1.0	1.3	4	1.2	0.6	4	0.3	0.2	
			18.0 - 21.0	4	1.0	0.6	4	0.9	0.9	3	0.1	0.0	
		Deposition	0.0 - 21.0		0.3	0.2	kBq m^{-2}	4.1	0.7	kBq m^{-2}	0.9	0.8	kBq m^{-2}
BX5	485808	4416774	0.0 - 0.3	5	2.3	0.8	5	100	17	7	6.4	2.3	
			0.0 - 3.0	4	3.4	1.6	4	91	29	4	7.1	4.6	
			3.0 - 6.0	4	1.4	0.5	4	33	6	4	2.4	0.6	
			6.0 - 9.0	4	0.8	0.4	4	9.8	2.0	4	0.8	0.2	
			9.0 - 12.0	4	1.5	0.4	4	3.8	1.0	4	0.3	0.1	
			12.0 - 15.0	4	1.8	1.1	4	0.9	0.3	4	0.1	0.1	
			15.0 - 18.0	4	0.9	0.3	4	0.9	0.4	5	0.0	0.1	
			18.0 - 21.0	5	1.5	0.6	5	0.6	0.4	5	0.2	0.2	
		Deposition	0.0 - 21.0		0.4	0.1	kBq m^{-2}	4.1	0.9	kBq m^{-2}	0.3	0.1	kBq m^{-2}
CX1	483618	4415182	0.0 - 0.3	5	2400	3200	5	41	11	2	4300	8	
			0.0 - 3.0	3	1500	440	3	60	6	4	5500	1200	
			3.0 - 6.0	6	690	110	6	35	4	5	3900	800	
			6.0 - 9.0	3	430	160	3	19	10	4	2300	850	
			9.0 - 12.0	4	210	100	4	8.7	5.3	4	1100	470	
			12.0 - 15.0	5	130	53	5	5.1	2.5	3	600	250	
			15.0 - 18.0	4	64	16	4	2.0	0.3	4	250	92	
			18.0 - 21.0	4	44	9	4	1.5	0.5	3	220	34	
		Deposition	0.0 - 21.0		100	19	kBq m^{-2}	4.3	0.5	kBq m^{-2}	540	53	kBq m^{-2}

Location	UTM East Coordinate (m)	UTM North Coordinate (m)	Depth Interval (cm)	Number of Samples	^{241}Am Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{137}Cs Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{239,240}Pu Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	
CX2	483751	4415127	0.0 - 0.3	4	240	52	4	37	6	4	1900	600	
			0.0 - 3.0	4	320	56	4	46	11	4	1880	670	
			3.0 - 6.0	6	310	35	6	45	7	2	1400	470	
			6.0 - 9.0	5	160	48	5	24	9	3	840	48	
			9.0 - 12.0	3	110	30	3	8.1	6.6	5	430	150	
			12.0 - 15.0	4	84	28	4	9.1	5.2	4	290	100	
			15.0 - 18.0	4	41	19	4	3.8	2.0	4	170	72	
			18.0 - 21.0	4	13	6	4	1.9	0.8	4	64	29	
		Deposition	0.0 - 21.0		33	2	kBq m^{-2}	4.5	0.8	kBq m^{-2}	170	110	kBq m^{-2}
CX3	484048	4414882	0.0 - 0.3	4	73	41	4	60	34	4	480	220	
			0.0 - 3.0	4	72	36	4	66	32	4	450	370	
			3.0 - 6.0	6	60	28	6	55	20	5	260	110	
			6.0 - 9.0	4	35	26	4	32	20	5	180	68	
			9.0 - 12.0	4	13	7	4	14	7	4	75	69	
			12.0 - 15.0	4	7.7	9.6	4	7.1	9.5	5	39	48	
			15.0 - 18.0	4	3.7	1.1	4	3.3	2.2	5	23	22	
			18.0 - 21.0	4	3.0	1.6	4	2.2	2.6	3	10	10	
		Deposition	0.0 - 21.0		6.1	2.3	kBq m^{-2}	5.7	2.0	kBq m^{-2}	32	13	kBq m^{-2}
CX4	484477	4414651	0.0 - 0.3	4	12	1	4	33	4	4	59	9	
			0.0 - 3.0	5	14	4	5	40	10	4	54	7	
			3.0 - 6.0	5	15	5	5	37	5	4	61	17	
			6.0 - 9.0	4	6.7	4.0	4	19	10	5	41	25	
			9.0 - 12.0	4	2.6	1.6	4	6.0	2.7	4	13	5	
			12.0 - 15.0	4	1.3	1.4	4	5.3	5.1	4	10	14	
			15.0 - 18.0	3	3.1	2.7	4	5.1	4.6	4	5.8	7.6	
			18.0 - 21.0	4	1.5	1.1	4	1.6	0.9	3	1.5	0.1	
		Deposition	0.0 - 21.0		1.4	0.4	kBq m^{-2}	3.7	1.0	kBq m^{-2}	6.0	0.9	kBq m^{-2}
CX5	484939	4414461	0.0 - 0.3	5	19	6	5	70	13	5	86	25	
			0.0 - 3.0	4	29	10	4	75	14	1	100	7	
			3.0 - 6.0	4	7.9	4.9	4	29	10	3	26	13	
			6.0 - 9.0	5	4.0	1.3	5	9.5	2.6	5	15	9	
			9.0 - 12.0	4	2.4	1.0	4	4.1	2.1	4	6.8	3.1	
			12.0 - 15.0	4	1.3	0.5	4	3.2	2.5	4	3.4	1.4	
			15.0 - 18.0	4	1.5	1.2	4	1.5	1.0	2	3.1	0.9	
			18.0 - 21.0	4	2.0	0.9	4	2.2	1.3	2	9.6	7.2	
		Deposition	0.0 - 21.0		1.7	0.2	kBq m^{-2}	4.5	1.0	kBq m^{-2}	5.6	1.9	kBq m^{-2}

Location	UTM East Coordinate (m)	UTM North Coordinate (m)	Depth Interval (cm)	Number of Samples	^{241}Am Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{137}Cs Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{239,240}Pu Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	
CX6	485458	4414126	0.0 - 0.3	4	13	6	4	100	29	3	65	11	
			0.0 - 3.0	4	11	4	4	130	16	3	52	11	
			3.0 - 6.0	5	4.4	1.9	5	56	32	4	19	12	
			6.0 - 9.0	4	2.2	1.2	4	29	39	4	9.8	5.7	
			9.0 - 12.0	5	1.5	0.3	5	3.6	1.5	5	3.3	2.4	
			12.0 - 15.0	4	2.2	1.4	4	26	49	4	0.9	0.7	
			15.0 - 18.0	4	1.5	0.9	4	1.0	0.9	4	0.7	0.5	
			18.0 - 21.0	4	0.8	0.8	4	0.8	0.2	4	0.4	0.3	
		Deposition	0.0 - 21.0		0.5	0.1	kBq m^{-2}	5.0	2.5	kBq m^{-2}	1.8	0.4	kBq m^{-2}
CX7	485848	4413861	0.0 - 0.3	4	1.2	0.7	4	12	4	4	3.2	0.8	
			0.0 - 3.0	5	1.8	0.9	5	14	2	4	4.6	2.1	
			3.0 - 6.0	4	2.0	1.1	4	10	1	4	4.3	1.1	
			6.0 - 9.0	4	1.3	0.6	4	10	1	4	4.8	1.7	
			9.0 - 12.0	4	1.9	1.3	4	11	2	4	3.7	1.5	
			12.0 - 15.0	5	1.3	0.3	5	12	3	3	12	15	
			15.0 - 18.0	4	1.3	0.9	4	11	3	2	12	7	
			18.0 - 21.0	4	1.0	0.6	4	5.8	2.4	2	3.5	4.0	
		Deposition	0.0 - 21.0		0.3	0.1	kBq m^{-2}	2.5	0.2	kBq m^{-2}	1.2	0.6	kBq m^{-2}
DX1	486280	4415274	0.0 - 0.3	4	18	4	4	120	27	3	59	8	
			0.0 - 3.0	4	27	9	4	190	39	4	96	22	
			3.0 - 6.0	1	5.1	0.4	1	14	1	1	31	1	
			6.0 - 9.0	1	1.3	0.2	1	2.6	0.3	1	2.5	0.1	
			9.0 - 12.0	1	1.2	0.2	1	0.8	0.2	1	1.5	0.1	
			12.0 - 15.0	1	1.8	0.3	1	0.5	0.2	2	0.5	0.1	
			15.0 - 18.0	2	0.3	0.1	2	0.3	0.1	1	0.3	0.0	
			18.0 - 21.0	1	1.0	0.2	1	0.6	0.2	1	0.6	0.0	
		Deposition	0.0 - 21.0		0.7	0.3	kBq m^{-2}	6.1	1.4	kBq m^{-2}	2.8	1.2	kBq m^{-2}
DX2	486708	4415256	0.0 - 0.3	4	13	3	4	130	9	4	52	8	
			0.0 - 3.0	4	11	4	4	190	49	5	33	23	
			3.0 - 6.0	1	1.7	0.2	1	19	1	1	5.0	0.2	
			6.0 - 9.0	1	3.5	0.4	1	8.2	0.6	1	-0.1	0.0	
			9.0 - 12.0	1	2.6	0.3	1	0.2	0.1	1	0.4	0.0	
			12.0 - 15.0	1	2.1	0.3	1	0.7	0.2	0	*	*	
			15.0 - 18.0	1	3.3	0.4	1	0.3	0.1	1	0.3	0.0	
			18.0 - 21.0	1	2.7	0.3	1	0.3	0.1	1	0.8	0.1	
		Deposition	0.0 - 21.0		0.7	0.5	kBq m^{-2}	6.1	1.5	kBq m^{-2}	1.3	0.3	kBq m^{-2}

Location	UTM East Coordinate (m)	UTM North Coordinate (m)	Depth Interval (cm)	Number of Samples	^{241}Am Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{137}Cs Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{239,240}Pu Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	
DX3	487713	4415095	0.0 - 0.3	4	8.7	2.6	4	152.7	54.5	6	30.5	21.0	
			0.0 - 3.0	5	5.5	1.4	5	158.0	32.3	4	29.3	13.6	
			3.0 - 6.0	1	1.5	0.4	1	15.1	1.3	1	3.4	0.2	
			6.0 - 9.0	1	1.8	0.2	1	17.5	0.9	1	1.9	0.1	
			9.0 - 12.0	1	1.6	0.4	1	1.7	0.4	1	0.3	0.1	
			12.0 - 15.0	1	2.3	0.4	1	1.7	0.4	1	0.2	0.1	
			15.0 - 18.0	1	1.7	0.3	1	0.9	0.2	1	0.6	0.1	
			18.0 - 21.0	1	0.8	0.2	1	2.9	0.4	1	0.2	0.1	
		Deposition	0.0 - 21.0		7.4	0.5	kBq m^{-2}	5.7	2.8	kBq m^{-2}	0.9	0.3	kBq m^{-2}
DX4	488889	4414787	0.0 - 0.3	4	2.4	0.7	4	86	36	2	4.4	0.6	
			0.0 - 3.0	6	2.8	1.2	6	153	61	4	7.3	1.3	
			3.0 - 6.0	1	3.2	0.3	1	24	1	0	*	*	
			6.0 - 9.0	1	1.7	0.2	1	6.6	0.5	1	0.4	0.0	
			9.0 - 12.0	1	1.6	0.2	1	2.4	0.3	1	0.0	0.0	
			12.0 - 15.0	1	1.0	0.2	1	0.7	0.2	1	0.1	0.0	
			15.0 - 18.0	1	1.3	0.2	1	0.9	0.2	1	0.1	0.0	
			18.0 - 21.0	1	2.0	0.3	1	0.7	0.2	1	0.1	0.0	
		Deposition	0.0 - 21.0		0.6	0.3	kBq m^{-2}	6.1	1.7	kBq m^{-2}	0.4	0.3	kBq m^{-2}
DX5	493817	4414073	0.0 - 0.3	5	1.7	0.6	5	74	15	4	1.8	0.5	
			0.0 - 3.0	4	1.9	0.8	4	83	29	4	2.4	0.4	
			3.0 - 6.0	1	1.9	0.3	1	18	1	0	*	*	
			6.0 - 9.0	1	0.6	0.2	1	13	1	1	0.2	0.0	
			9.0 - 12.0	1	0.6	0.0	1	5.1	0.2	0	*	*	
			12.0 - 15.0	1	2.1	0.3	1	3.2	0.4	1	0.1	0.0	
			15.0 - 18.0	1	2.1	0.3	1	1.9	0.3	0	*	*	
			18.0 - 21.0	1	1.7	0.2	1	1.6	0.3	1	-0.2	0.0	
		Deposition	0.0 - 21.0		0.4	0.1	kBq m^{-2}	4.8	1.4	kBq m^{-2}	0.1	0.03	kBq m^{-2}
DX6	502328	4413641	0.0 - 0.3	5	1.4	0.5	5	62	26	5	1.5	0.1	
			0.0 - 3.0	5	1.4	0.6	5	50	27	4	1.7	0.5	
			3.0 - 6.0	1	1.5	0.2	1	22	1	0	*	*	
			6.0 - 9.0	1	0.4	0.1	1	5.2	0.4	1	0.1	0.0	
			9.0 - 12.0	1	1.4	0.2	1	1.4	0.2	0	*	*	
			12.0 - 15.0	1	1.7	0.2	1	1.3	0.2	1	0.0	0.0	
			15.0 - 18.0	1	0.3	0.0	1	1.0	0.2	0	*	*	
			18.0 - 21.0	1	1.4	0.2	1	1.1	0.2	0	*	*	
		Deposition	0.0 - 21.0		0.2	0.1	kBq m^{-2}	4.6	2.4	kBq m^{-2}	0.2	0.07	kBq m^{-2}

Location	UTM East Coordinate (m)	UTM North Coordinate (m)	Depth Interval (cm)	Number of Samples	^{241}Am Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{137}Cs Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{239,240}Pu Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	
EX1	486357	4416887	0.0 - 0.3	6	1.1	0.5	6	40	3.0	9	2.0	1.2	
			0.0 - 3.0	4	2.0	1.4	4	35	2.3	5	1.9	0.2	
			3.0 - 6.0	1	1.4	0.2	1	39	1.4	0	*	*	
			6.0 - 9.0	1	1.3	0.4	1	36	2.2	1	2.2	0.1	
			9.0 - 12.0	1	1.1	0.3	1	19	1.3	1	2.2	0.2	
			12.0 - 15.0	1	2.1	0.5	1	28	2.1	1	3.2	0.4	
			15.0 - 18.0	1	0.4	0.1	1	20	0.9	0	*	*	
			18.0 - 21.0	1	0.6	0.2	1	36	1.5	1	1.0	0.1	
		Deposition	0.0 - 21.0		0.3	0.2	kBq m^{-2}	5.1	2.7	kBq m^{-2}	0.3	0.2	kBq m^{-2}
EX2	487051	4417255	0.0 - 0.3	4	2.0	1.4	4	85	15	8	4.8	1.5	
			0.0 - 3.0	5	1.9	1.3	5	149	20	4	8.9	2.6	
			3.0 - 6.0	1	1.3	0.2	1	40	1.4	1	1.4	0.1	
			6.0 - 9.0	1	0.4	0.0	1	9	0.2	1	0.3	0.0	
			9.0 - 12.0	1	0.9	0.2	1	4.6	0.4	1	0.1	0.1	
			12.0 - 15.0	1	1.0	0.3	1	1.5	0.5	1	0.1	0.0	
			15.0 - 18.0	1	1.7	0.2	1	1.5	0.3	0	*	*	
			18.0 - 21.0	1	1.1	0.2	1	1.1	0.2	1	-0.0	0.0	
		Deposition	0.0 - 21.0		0.3	0.1	kBq m^{-2}	8.7	4.5	kBq m^{-2}	0.4	0.2	kBq m^{-2}
EX3	487415	4417761	0.0 - 0.3	3	1.0	0.9	3	100	9	8	4.0	1.2	
			0.0 - 3.0	4	3.1	2.5	4	190	55	2	9.8	7.8	
			3.0 - 6.0	1	1.7	0.3	1	39	1.4	0	*	*	
			6.0 - 9.0	1	1.0	0.2	1	5.7	0.6	1	-0.1	0.0	
			9.0 - 12.0	1	1.3	0.2	1	1.6	0.3	0	*	*	
			12.0 - 15.0	1	0.8	0.2	1	2.5	0.4	1	-0.1	0.0	
			15.0 - 18.0	1	0.0	0.0	1	1.6	0.4	0	*	*	
			18.0 - 21.0	1	1.2	0.4	1	3.2	0.7	1	-1.5	0.1	
		Deposition	0.0 - 21.0		0.2	0.1	kBq m^{-2}	6.1	2.4	kBq m^{-2}	0.2	0.03	kBq m^{-2}
EX4	489541	4418414	0.0 - 0.3	4	2.3	1.1	4	170	30	7	8.2	3.3	
			0.0 - 3.0	5	1.8	1.2	5	93	52	5	3.0	2.9	
			3.0 - 6.0	1	2.2	0.5	1	2.6	0.6	1	0.1	0.1	
			6.0 - 9.0	2	3.1	1.4	2	120	4	1	3.5	0.1	
			9.0 - 12.0	1	0.7	0.3	1	1.3	0.4	1	-0.0	0.0	
			12.0 - 15.0	1	0.9	0.2	1	9.6	0.7	1	-0.1	0.0	
			15.0 - 18.0	1	1.5	0.2	1	1.1	0.2	1	-0.1	0.0	
			18.0 - 21.0	1	0.7	0.1	1	1.1	0.2	1	0.0	0.0	
		Deposition	0.0 - 21.0		0.3	0.1	kBq m^{-2}	11.5	2.5	kBq m^{-2}	0.3	0.1	kBq m^{-2}

Location	UTM East Coordinate (m)	UTM North Coordinate (m)	Depth Interval (cm)	Number of Samples	^{241}Am Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{137}Cs Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{239,240}Pu Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	
EX5	493780	4422426	0.0 - 0.3	4	1.7	1.2	4	50	12	8	1.1	0.5	
			0.0 - 3.0	5	0.6	0.5	5	40	19	4	1.0	0.5	
			3.0 - 6.0	1	1.5	0.3	1	12	0.9	0	•	•	
			6.0 - 9.0	1	0.7	0.0	1	3.7	0.1	1	0.1	0.0	
			9.0 - 12.0	1	1.0	0.2	1	1.4	0.3	0	•	•	
			12.0 - 15.0	1	0.5	0.2	1	1.1	0.3	1	0.0	0.0	
			15.0 - 18.0	1	0.4	0.1	1	0.5	0.2	0	•	•	
			18.0 - 21.0	1	0.6	0.2	1	0.5	0.2	1	-0.1	0.1	
		Deposition	0.0 - 21.0		0.3	0.1	kBq m^{-2}	1.4	0.7	kBq m^{-2}	0.02	0.01	kBq m^{-2}
EX6	497942	4424872	0.0 - 0.3	4	1.2	0.7	4	164	15	6	2.5	0.5	
			0.0 - 3.0	4	1.8	0.7	4	201	29	4	3.5	0.9	
			3.0 - 6.0	1	0.6	0.1	1	46	2	0	•	•	
			6.0 - 9.0	1	0.8	0.1	1	2.9	0.3	1	0.3	0.0	
			9.0 - 12.0	1	0.9	0.1	1	0.9	0.2	0	•	•	
			12.0 - 15.0	0	•	•	0	•	•	1	0.0	0.0	
			15.0 - 18.0	0	•	•	1	0.5	0.2	1	-0.0	0.0	
			18.0 - 21.0	1	0.3	0.0	1	0.3	0.0	1	0.0	0.0	
		Deposition	0.0 - 21.0		0.3	0.2	kBq m^{-2}	4.9	3.6	kBq m^{-2}	0.2	0.0	kBq m^{-2}
FX1	486311	4413733	0.0 - 0.3	6	4.4	0.5	6	85	10	6	24	5	
			0.0 - 3.0	5	9.8	8.0	5	90	10	4	18	5	
			3.0 - 6.0	1	1.0	0.2	1	11	1	1	3.5	0.2	
			6.0 - 9.0	0	•	•	0	•	•	1	8.6	1.2	
			9.0 - 12.0	1	0.8	0.2	1	6.0	0.6	1	2.0	0.1	
			12.0 - 15.0	1	1.6	0.2	1	9.6	0.7	1	1.4	0.1	
			15.0 - 18.0	1	1.1	0.2	1	3.7	0.4	1	0.8	0.1	
			18.0 - 21.0	1	0.9	0.2	1	4.5	0.4	1	0.7	0.0	
		Deposition	0.0 - 21.0		0.2	0.2	kBq m^{-2}	2.9	1.6	kBq m^{-2}	1.3	0.9	kBq m^{-2}
FX2	486769	4413573	0.0 - 0.3	4	3.2	1.0	4	67	9	5	17	5	
			0.0 - 3.0	4	6.6	1.9	4	140	13	4	30	14	
			3.0 - 6.0	1	0.7	0.2	1	21	1	1	2.1	0.1	
			6.0 - 9.0	1	1.3	0.2	1	83	2	1	0.7	0.1	
			9.0 - 12.0	1	0.8	0.2	1	2.5	0.3	1	0.3	0.0	
			12.0 - 15.0	1	0.5	0.1	1	2.5	0.3	1	0.3	0.0	
			15.0 - 18.0	1	1.3	0.3	1	3.2	0.6	1	0.5	0.0	
			18.0 - 21.0	1	0.3	0.2	1	4.0	0.6	1	0.4	0.1	
		Deposition	0.0 - 21.0		0.3	0.1	kBq m^{-2}	5.1	2.5	kBq m^{-2}	0.7	0.2	kBq m^{-2}

Location	UTM East Coordinate (m)	UTM North Coordinate (m)	Depth Interval (cm)	Number of Samples	^{241}Am Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{137}Cs Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{239,240}Pu Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	
FX3	487813	4412962	0,0 - 0,3	4	1,6	0,8	4	72	11	6	10	4,3	
			0,0 - 3,0	4	5,8	3,3	4	100	41	4	7,7	5,4	
			3,0 - 6,0	1	2,7	0,3	1	99	2	0	*	*	
			6,0 - 9,0	1	1,2	0,4	1	21	2	1	0,9	0,1	
			9,0 - 12,0	2	0,5	0,4	2	6,2	1,6	1	0,8	0,0	
			12,0 - 15,0	1	0,8	0,2	1	22	1	1	0,4	0,0	
			15,0 - 18,0	1	0,6	0,1	1	5,7	0,5	0	*	*	
			18,0 - 21,0	1	0,0	0,0	1	1,0	0,2	1	0,1	0,0	
		Deposition	0,0 - 21,0		0,4	0,02	kBq m^{-2}	5,2	2,9	kBq m^{-2}	0,3	0,2	kBq m^{-2}
FX4	489490	4411397	0,0 - 0,3	5	1,2	0,3	5	29	6,4	8	1,1	0,4	
			0,0 - 3,0	4	0,6	0,3	4	28	3,1	5	1,0	0,5	
			3,0 - 6,0	1	1,2	0,2	1	25	1,1	1	0,5	0,1	
			6,0 - 9,0	1	2,7	0,6	1	30	2,3	1	0,5	0,1	
			9,0 - 12,0	1	2,7	0,5	1	10,0	1,1	1	0,2	0,1	
			12,0 - 15,0	1	1,3	0,2	1	3,8	0,4	1	0,1	0,0	
			15,0 - 18,0	1	0,1	0,0	1	2,5	0,3	1	0,0	0,0	
			18,0 - 21,0	1	4,7	0,8	1	3,6	0,8	1	0,5	0,0	
		Deposition	0,0 - 21,0		0,4	0,3	kBq m^{-2}	4,5	1,1	kBq m^{-2}	0,1	0,04	kBq m^{-2}
FX5	491032	4409904	0,0 - 0,3	4	1,1	0,8	4	47	46	7	12	29	
			0,0 - 3,0	4	0,7	0,2	4	40	17	5	1,3	0,6	
			3,0 - 6,0	1	2,2	0,5	1	14	0,1	1	2,8	0,1	
			6,0 - 9,0	1	0,6	0,2	1	9,8	0,7	1	0,6	0,1	
			9,0 - 12,0	1	1,2	0,2	1	7,1	0,6	0	-0,0	0,1	
			12,0 - 15,0	1	1,0	0,2	1	6,3	0,6	2	0,0	0,1	
			15,0 - 18,0	1	2,2	0,5	1	3,1	0,6	0	-0,0	0,4	
			18,0 - 21,0	1	1,8	0,4	1	1,5	0,4	2	-0,0	0,0	
		Deposition	0,0 - 21,0		0,3	0,1	kBq m^{-2}	4,3	3,1	kBq m^{-2}	0,1	0,05	kBq m^{-2}
FX6	500234	4408098	0,0 - 0,3	4	0,9	0,2	4	12	4,7	8	0,1	0,6	
			0,0 - 3,0	5	1,0	0,3	5	11	5,2	4	0,5	0,7	
			3,0 - 6,0	1	1,7	0,3	1	2,9	0,4	1	0,1	0,0	
			6,0 - 9,0	1	1,6	0,3	1	2,5	0,4	1	1,6	0,0	
			9,0 - 12,0	1	0,5	0,2	1	0,8	0,2	1	0,0	0,0	
			12,0 - 15,0	1	0,7	0,2	1	0,4	0,1	1	0,3	0,0	
			15,0 - 18,0	1	0,0	0,0	1	1,1	0,3	1	0,0	0,0	
			18,0 - 21,0	1	1,1	0,2	1	0,1	0,0	1	0,2	0,0	
		Deposition	0,0 - 21,0		0,3	0,1	kBq m^{-2}	1,1	0,4	kBq m^{-2}	0,2	0,2	kBq m^{-2}

Location	UTM East Coordinate (m)	UTM North Coordinate (m)	Depth Interval (cm)	Number of Samples	^{241}Am Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{137}Cs Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{239,240}Pu Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	
GX1	484670	4412884	0.0 - 0.3	4	1.4	0.5	4	120	24	6	5.3	1.5	
			0.0 - 3.0	4	2.1	1.6	4	110	14	5	3.6	1.0	
			3.0 - 6.0	1	0.8	0.3	1	20	0.2	2	0.6	0.3	
			6.0 - 9.0	1	0.5	0.1	1	4.6	0.5	1	0.2	0.0	
			9.0 - 12.0	1	0.5	0.1	1	3.1	0.4	0	•	•	
			12.0 - 15.0	1	0.7	0.1	1	3.2	0.4	1	-0.0	0.0	
			15.0 - 18.0	1	1.9	0.3	1	0.5	0.2	0	•	•	
			18.0 - 21.0	1	1.4	0.2	1	0.4	0.1	1	-0.1	0.0	
		Deposition	0.0 - 21.0		0.3	0.1	kBq m^{-2}	1.3	0.9	kBq m^{-2}	0.1	0.08	kBq m^{-2}
GX2	485076	4412190	0.0 - 0.3	5	0.8	0.6	5	28	5.0	9	0.7	0.3	
			0.0 - 3.0	5	1.5	0.4	5	42	17	5	1.1	0.7	
			3.0 - 6.0	1	0.3	0.0	1	52	1.5	0	•	•	
			6.0 - 9.0	1	0.8	0.1	1	5.5	0.5	1	0.1	0.0	
			9.0 - 12.0	1	1.1	0.2	1	0.7	0.2	0	•	•	
			12.0 - 15.0	1	0.5	0.1	1	0.8	0.2	1	-0.1	0.0	
			15.0 - 18.0	1	0.7	0.1	1	0.5	0.1	0	•	•	
			18.0 - 21.0	1	1.7	0.4	1	1.1	0.3	0	•	•	
		Deposition	0.0 - 21.0		0.3	0.1	kBq m^{-2}	3.0	0.6	kBq m^{-2}	0.08	0.07	kBq m^{-2}
GX3	485749	4411663	0.0 - 0.3	5	1.1	0.5	5	14	1.5	10	0.4	0.2	
			0.0 - 3.0	4	1.2	0.8	4	16	2.1	2	0.4	0.0	
			3.0 - 6.0	1	0.0	0.0	1	24	1.1	0	•	•	
			6.0 - 9.0	1	1.4	0.2	1	18	0.9	1	0.0	0.1	
			9.0 - 12.0	1	0.0	0.0	1	12	0.7	0	•	•	
			12.0 - 15.0	1	0.5	0.1	1	10	0.6	1	0.4	0.0	
			15.0 - 18.0	1	1.1	0.2	1	6.6	0.4	0	•	•	
			18.0 - 21.0	1	0.8	0.1	1	2.5	0.3	1	0.1	0.0	
		Deposition	0.0 - 21.0		0.1	0.0	kBq m^{-2}	2.0	0.1	kBq m^{-2}	0.04	0.10	kBq m^{-2}
GX4	485926	4409440	0.0 - 0.3	5	0.8	0.4	5	32	2.3	9	0.7	0.2	
			0.0 - 3.0	5	1.0	0.4	5	46	10	4	0.9	0.4	
			3.0 - 6.0	1	0.7	0.2	1	52	1.6	0	•	•	
			6.0 - 9.0	1	0.9	0.2	1	21	1.0	1	0.5	0.0	
			9.0 - 12.0	1	0.5	0.0	1	8.3	0.3	0	•	•	
			12.0 - 15.0	1	1.5	0.2	1	3.4	0.4	1	0.1	0.0	
			15.0 - 18.0	1	0.3	0.1	1	2.2	0.3	0	•	•	
			18.0 - 21.0	1	1.2	0.2	1	0.7	0.2	1	0.0	0.0	
		Deposition	0.0 - 21.0		0.2	0.0	kBq m^{-2}	1.6	0.2	kBq m^{-2}	0.03	0.01	kBq m^{-2}

Location	UTM East Coordinate (m)	UTM North Coordinate (m)	Depth Interval (cm)	Number of Samples	^{241}Am Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{137}Cs Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{239,240}Pu Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	
GX5	489345	4405011	0.0 - 0.3	4	1.1	0.5	4	51	12	2	1.4	0.7	
			0.0 - 3.0	4	1.4	0.3	4	66	17	4	1.3	0.4	
			3.0 - 6.0	1	0.7	0.2	1	50	1.7	0	•	•	
			6.0 - 9.0	2	1.2	0.9	2	11	1.0	1	0.2	0.0	
			9.0 - 12.0	1	0.3	0.1	1	0.7	0.2	0	•	•	
			12.0 - 15.0	1	0.7	0.2	1	1.7	0.3	1	0.1	0.0	
			15.0 - 18.0	1	1.5	0.3	1	0.9	0.2	0	•	•	
			18.0 - 21.0	1	0.0	0.0	1	0.7	0.2	2	0.0	0.0	
		Deposition	0.0 - 21.0		0.1	0.0	kBq m^{-2}	2.3	0.4	kBq m^{-2}	0.05	0.01	kBq m^{-2}
GX6	493335	4400160	0.0 - 0.3	4	0.3	0.4	4	4.4	0.5	2	0.1	0.0	
			0.0 - 3.0	4	0.9	0.6	4	8.6	2.1	3	0.1	0.0	
			3.0 - 6.0	1	0.4	0.0	1	5.5	0.2	0	•	•	
			6.0 - 9.0	1	0.7	0.1	1	3.0	0.3	1	0.1	0.0	
			9.0 - 12.0	1	1.3	0.2	1	3.8	0.4	0	•	•	
			12.0 - 15.0	1	0.0	0.0	1	2.5	0.3	1	0.0	0.0	
			15.0 - 18.0	1	0.0	0.0	1	2.7	0.3	0	•	•	
			18.0 - 21.0	1	1.8	0.2	1	12.7	0.7	1	0.3	0.0	
		Deposition	0.0 - 21.0		0.1	0.0	kBq m^{-2}	1.2	0.1	kBq m^{-2}	0.02	0.01	kBq m^{-2}
K01 Boulder	478656	4423727	0.0 - 0.3	•	•	•	•	•	•	•	•	•	
			0.0 - 3.0	1	0.4	0.1	1	60	1.5	1	2.0	0.1	
		Deposition	0.0 - 21.0		0.2	0.04	kBq m^{-2}	8.5	0.4	kBq m^{-2}	0.2		kBq m^{-2}
K02 Golden	486018	4397711	0.0 - 0.3	•	•	•	•	•	•	•	•	•	
			0.0 - 3.0	1	0.4	0.1	1	54	1.7	1	1.2	0.1	
		Deposition	0.0 - 21.0		0.3	0.05	kBq m^{-2}	5.4	0.3	kBq m^{-2}	0.1		kBq m^{-2}
K03 Marshall	480047	4422093	0.0 - 0.3	•	•	•	•	•	•	•	•	•	
			0.0 - 3.0	1	0.8	0.2	1	56	1.5	1	2.6	0.1	
		Deposition	0.0 - 21.0		0.1	0.02	kBq m^{-2}	5.4	0.2	kBq m^{-2}	0.2		kBq m^{-2}
K04 Eldorado Springs	477952	4420840	0.0 - 0.3	•	•	•	•	•	•	•	•	•	
			0.0 - 3.0	1	0.7	0.1	1	110	1.8	1	3.7	1.5	
		Deposition	0.0 - 21.0		•	•	kBq m^{-2}	•	•	kBq m^{-2}	•	•	kBq m^{-2}

Location	UTM East Coordinate (m)	UTM North Coordinate (m)	Depth Interval (cm)	Number of Samples	^{241}Am Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{137}Cs Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{239,240}Pu Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	
K05 Wheatridge	490677	4401214	0.0 - 0.3	*	*	*	*	*	*	*	*	*	
			0.0 - 3.0	1	1	0	1	59	2	1	1	1	
		Deposition	0.0 - 21.0		0.0	0.0	kBq m^{-2}	6.0	0.3	kBq m^{-2}	0.0	0.0	kBq m^{-2}
K06 Arvada	491362	4405615	0.0 - 0.3	*	*	*	*	*	*	*	*	*	
			0.0 - 3.0	1	1.7	0.3	2	61	2.0	1	1.4	0.4	
		Deposition			0.1	0.4	kBq m^{-2}	2.6	0.2	kBq m^{-2}	0.1		kBq m^{-2}
K07 Westminster	497184	4410443	0.0 - 0.3	*	*	*	*	*	*	*	*	*	
			0.0 - 3.0	1	1.8	0.3	1	86	2.3	1	1.9	0.1	
		Deposition			0.4	0.06	kBq m^{-2}	6.9	0.4	kBq m^{-2}	0.2		kBq m^{-2}
K08 Thornton/ Northglenn	503091	4414425	0.0 - 0.3	*	*	*	*	*	*	*	*	*	
			0.0 - 3.0	1	1.3	0.2	1	67	2.0	1	1.0	0.1	
		Deposition	0.0 - 21.0		0.2	0.06	kBq m^{-2}	4.3	0.3	kBq m^{-2}	0.1		kBq m^{-2}
K09 Broomfield	492293	4417808	0.0 - 0.3	*	*	*	*	*	*	*	*	*	
			0.0 - 3.0	1	1.5	0.2	1	95	2.2	2	3.7	0.1	
		Deposition	0.0 - 21.0		0.3	0.05	kBq m^{-2}	8.4	0.3	kBq m^{-2}	0.3		kBq m^{-2}
K10 Superior	484127	4422542	0.0 - 0.3	*	*	*	*	*	*	*	*	*	
			0.0 - 3.0	2	1.2	0.0	2	92	4.3	1	2.6	0.2	
		Deposition	0.0 - 21.0	0	0.1	0.03	kBq m^{-2}	7.1	0.2	kBq m^{-2}	0.2		kBq m^{-2}
Z01 Pawnee Grassland Weld Co	520524	4516126	0.0 - 0.3	1	4.8	0.8	1	69	3.7	2	1.5	0.9	
			0.0 - 3.0	1	0.5	0.3	1	80	4.2	1	1.2	0.1	
		Deposition	0.0 - 21.0		0.2	0.04	kBq m^{-2}	4.5	0.3	kBq m^{-2}	0.1		kBq m^{-2}
Z02 CSU Airport Weld Co	487734	4494329	0.0 - 0.3	1	1.1	0.2	1	71	1.8	2	0.5	1.4	
			0.0 - 3.0	1	4.8	0.7	1	110	3.7	1	2.1	0.1	
		Deposition	0.0 - 21.0		0.2	0.0	kBq m^{-2}	5.7	0.2	kBq m^{-2}	0.1		kBq m^{-2}

Location	UTM East Coordinate (m)	UTM North Coordinate (m)	Depth Interval (cm)	Number of Samples	^{241}Am Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{137}Cs Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{239,240}Pu Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	
Z03 Weld/Larimer Co Line	505381	4503233	0.0 - 0.3	1	0.7	0.2	1	130	2.7	2	2.1	0.7	
			0.0 - 3.0	1	6.0	0.9	1	120	4.5	1	1.5	0.1	
		Deposition	0.0 - 21.0		0.5	0.1	kBq m^{-2}	6.9	0.5	kBq m^{-2}	0.1		kBq m^{-2}
Z04 Dixon Reservoir Larimer Co	487947	4488793	0.0 - 0.3	1	1.1	0.1	1	86	1.2	2	1.5	0.3	
			0.0 - 3.0	1	0.9	0.2	1	120	2.7	1	2.1	0.1	
		Deposition	0.0 - 21.0		0.2	0.04	kBq m^{-2}	8.0	0.3	kBq m^{-2}	0.1		kBq m^{-2}
Z05 Southwest Fort Collins	489303	4484305	0.0 - 0.3	1	1.4	0.2	1	150	3.0	2	2.2	0.1	
			0.0 - 3.0	1	0.9	0.2	1	140	2.9	1	2.1	0.1	
		Deposition	0.0 - 21.0		0.4	0.06	kBq m^{-2}	6.4	0.3	kBq m^{-2}	0.1		kBq m^{-2}
Z06 Southeast Fort Collins	494866	4483200	0.0 - 0.3	1	0.6	0.2	1	76	2.2	2	1.0	0.1	
			0.0 - 3.0	1	1.8	0.3	1	85	2.3	1	1.1	0.1	
		Deposition	0.0 - 21.0		0.2	0.05	kBq m^{-2}	9.1	0.4	kBq m^{-2}	0.1		kBq m^{-2}
Z07 Colorado Springs	513609	4295761	0.0 - 0.3	1	1.7	0.3	1	120	2.8	0	*		
			0.0 - 3.0	1	2.2	0.3	1	190	3.4	1	3.3	0.1	
		Deposition	0.0 - 21.0		0.1	0.02	kBq m^{-2}	10.5	1.0	kBq m^{-2}	*		kBq m^{-2}
Z08 Castle Rock	501481	4359837	0.0 - 0.3	1	0.9	0.2	1	110	2.6	2	7.9	9.1	
			0.0 - 3.0	1	2.6	0.3	1	170	3.2	1	3.2	0.1	
		Deposition	0.0 - 21.0		0.3	0.06	kBq m^{-2}	8.1	0.4	kBq m^{-2}	0.2		kBq m^{-2}
Z09 Monument	511224	4336527	0.0 - 0.3	1	1.6	0.3	1	100	2.5	2	3.0	2.2	
			0.0 - 3.0	3	10.4	15.9	3	100	45	1	2.1	0.1	
		Deposition	0.0 - 21.0		0.4	0.1	kBq m^{-2}	5.2	1.2	kBq m^{-2}	*		kBq m^{-2}

Location	UTM East Coordinate (m)	UTM North Coordinate (m)	Depth Interval (cm)	Number of Samples	^{241}Am Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{137}Cs Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{239,240}Pu Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	
Z10	4516126	4438567	0.0 - 0.3	3	1.8	1.5	3	48	73	2	2.6	0.2	
Left-hand Valley Reservoir			0.0 - 3.0	2	6.9	9.6	2	7.6	8.2	1	2.7	0.1	
Boulder, Co		Deposition	0.0 - 21.0		0.2	0.04	kBq m^{-2}	2.9	0.1	kBq m^{-2}	0.7		kBq m^{-2}

Appendix B

Soil Concentrations of ^{226}Ra, ^{232}Th and ^{40}K at Locations Around Rocky Flats Environmental Technology Site and Along Colorado's Front Range Corridor

Location	UTM East Coordinate (m)	UTM North Coordinate (m)	Depth Interval (cm)	Number of Samples	^{226}Ra Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{232}Th Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{40}K Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})
AX1	483,658	4,415,301	0,0 - 0,3	4	44	6	4	61	7	4	600	76
			0,0 - 3,0	6	56	7	6	94	5	6	730	59
			3,0 - 6,0	4	68	6	4	100	7	4	760	37
			6,0 - 9,0	4	59	3	4	100	4	4	740	47
			9,0 - 12,0	4	61	8	4	99	8	4	700	98
			12,0 - 15,0	4	55	5	4	100	5	4	740	56
			15,0 - 18,0	4	61	7	4	100	7	4	750	27
			18,0 - 21,0	4	59	10	4	96	11	4	700	120
		Concentration	0,0 - 21,0	30	59	8	30	100	10	30	730	66
AX2	483,792	4,415,305	0,0 - 0,3	4	35	8	4	69	3	4	670	41
			0,0 - 3,0	4	53	8	4	90	8	4	790	25
			3,0 - 6,0	6	51	8	6	88	6	6	750	65
			6,0 - 9,0	4	54	7	4	95	6	4	790	63
			9,0 - 12,0	4	49	4	4	92	8	4	810	34
			12,0 - 15,0	4	55	9	4	99	6	4	810	59
			15,0 - 18,0	4	50	2	4	97	6	4	830	20
			18,0 - 21,0	4	54	9	4	98	7	4	850	32
		Mean Concentration	0,0 - 21,0	30	52	8	30	94	8	30	800	53
AX3	484,124	4,415,316	0,0 - 0,3	5	40	6	5	75	3	5	910	31
			0,0 - 3,0	5	45	5	5	79	6	5	920	44
			3,0 - 6,0	5	37	8	5	77	4	5	910	28
			6,0 - 9,0	4	30	4	4	75	4	4	780	120
			9,0 - 12,0	4	30	6	4	76	8	4	800	140
			12,0 - 15,0	4	35	10	4	80	10	4	840	91
			15,0 - 18,0	4	31	10	4	77	11	4	860	160
			18,0 - 21,0	4	23	7	4	74	13	4	810	170
		Mean Concentration	0,0 - 21,0	30	34	10	30	77	12	30	850	120

Location	UTM East Coordinate (m)	UTM North Coordinate (m)	Depth Interval (cm)	Number of Samples	^{226}Ra Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{232}Th Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{40}K Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})
AX4	484,642	4,415,326	0.0 - 0.3	5	55	8	5	86	5	5	640	26
			0.0 - 3.0	4	53	8	4	98	6	4	770	39
			3.0 - 6.0	4	58	9	4	100	8	4	750	40
			6.0 - 9.0	4	60	8	4	110	6	4	800	79
			9.0 - 12.0	4	55	7	4	100	9	4	780	160
			12.0 - 15.0	6	64	7	6	110	5	6	840	35
			15.0 - 18.0	4	62	7	4	110	6	4	820	41
			18.0 - 21.0	4	61	5	4	110	5	4	840	50
		Mean Concentration	0.0 - 21.0	30	59	7	30	110	8	30	800	72
AX5	485,127	4,415,346	0.0 - 0.3	3	52	7	3	88	8	3	590	160
			0.0 - 3.0	4	57	7	4	100	4	4	760	34
			3.0 - 6.0	4	61	7	4	120	5	4	830	64
			6.0 - 9.0	4	57	6	4	120	4	4	840	27
			9.0 - 12.0	5	56	7	5	120	7	5	860	57
			12.0 - 15.0	5	62	8	5	110	9	5	820	170
			15.0 - 18.0	4	62	11	4	110	3	4	860	48
			18.0 - 21.0	4	60	5	4	120	5	4	890	27
		Mean Concentration	0.0 - 21.0	30	59	7	30	110	9	30	840	82
AX6	485,785	4,415,302	0.0 - 0.3	4	56	9	4	94	6	4	740	90
			0.0 - 3.0	4	68	6	4	99	6	4	770	60
			3.0 - 6.0	4	63	4	4	100	4	4	840	32
			6.0 - 9.0	4	73	14	4	100	4	4	840	34
			9.0 - 12.0	4	76	12	4	100	5	4	880	20
			12.0 - 15.0	5	78	10	5	99	4	5	890	46
			15.0 - 18.0	5	81	12	5	100	6	5	890	33
			18.0 - 21.0	4	76	8	4	100	3	4	880	32
		Mean Concentration	0.0 - 21.0	30	74	10	30	100	5	30	860	53

Location	UTM East Coordinate (m)	UTM North Coordinate (m)	Depth Interval (cm)	Number of Samples	^{226}Ra Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{232}Th Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{40}K Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})
BX1	484,104	4,415,741	0.0 - 0.3	4	58	10	4	94	3	4	580	49
			0.0 - 3.0	4	56	3	4	95	5	4	700	13
			3.0 - 6.0	4	56	14	4	96	4	4	730	25
			6.0 - 9.0	4	56	4	4	93	5	4	710	27
			9.0 - 12.0	4	55	17	4	100	5	4	750	58
			12.0 - 15.0	6	58	10	6	97	6	6	700	28
			15.0 - 18.0	4	58	9	4	97	7	4	720	44
			18.0 - 21.0	5	58	11	5	98	5	5	700	34
		Mean Concentration	0.0 - 21.0	31	57	10	31	97	6	31	710	36
BX2	484,423	4,415,827	0.0 - 0.3	5	46	6	5	71	5	5	690	61
			0.0 - 3.0	5	46	7	5	73	6	5	830	38
			3.0 - 6.0	4	54	9	4	80	7	4	800	36
			6.0 - 9.0	4	47	7	4	76	6	4	760	25
			9.0 - 12.0	4	44	8	4	72	5	4	720	41
			12.0 - 15.0	3	45	11	3	69	6	3	750	120
			15.0 - 18.0	3	48	4	3	78	5	3	750	31
			18.0 - 21.0	3	44	7	3	72	5	3	720	66
		Mean Concentration	0.0 - 21.0	26	47	8	26	74	8	26	770	61
BX3	484,877	4,416,074	0.0 - 0.3	4	49	12	4	75	8	4	740	223
			0.0 - 3.0	4	46	4	4	72	4	4	750	42
			3.0 - 6.0	4	54	4	4	83	4	4	780	19
			6.0 - 9.0	5	55	6	5	83	6	5	780	15
			9.0 - 12.0	5	62	10	5	89	4	5	810	62
			12.0 - 15.0	4	56	9	4	86	4	4	830	76
			15.0 - 18.0	4	61	6	4	85	5	4	790	40
			18.0 - 21.0	4	60	6	4	80	5	4	730	98
		Mean Concentration	0.0 - 21.0	27	56	8	27	83	7	27	780	63

Location	UTM East Coordinate (m)	UTM North Coordinate (m)	Depth Interval (cm)	Number of Samples	^{226}Ra Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{232}Th Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{40}K Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})
BX4	485,488	4,416,513	0.0 - 0.3	4	53	8	4	73	3	4	610	53
			0.0 - 3.0	4	52	9	4	67	9	4	680	61
			3.0 - 6.0	4	63	10	4	78	6	4	750	42
			6.0 - 9.0	4	55	4	4	79	4	4	740	31
			9.0 - 12.0	4	57	6	4	77	5	4	750	46
			12.0 - 15.0	4	67	9	4	87	5	4	740	47
			15.0 - 18.0	4	60	8	4	83	6	4	730	74
			18.0 - 21.0	4	70	16	4	89	12	4	800	130
		Mean Concentration	0.0 - 21.0	28	61	11	28	80	12	28	740	69
BX5	485,808	4,416,774	0.0 - 0.3	5	55	4	5	70	4	5	570	25
			0.0 - 3.0	4	61	5	4	68	5	4	680	73
			3.0 - 6.0	4	64	7	4	63	4	4	650	31
			6.0 - 9.0	4	62	4	4	69	5	4	630	32
			9.0 - 12.0	4	58	6	4	72	6	4	660	13
			12.0 - 15.0	4	54	5	4	67	6	4	640	34
			15.0 - 18.0	4	66	6	4	75	5	4	660	51
			18.0 - 21.0	5	65	8	5	73	5	5	640	54
		Mean Concentration	0.0 - 21.0	29	62	7	29	70	7	29	650	43
CX1	483,618	4,415,182	0.0 - 0.3	5	63	6	5	97	17	5	770	76
			0.0 - 3.0	3	68	9	3	100	20	3	880	45
			3.0 - 6.0	6	58	11	6	110	7	6	810	110
			6.0 - 9.0	3	60	5	3	120	8	3	850	52
			9.0 - 12.0	4	68	7	4	100	19	4	850	70
			12.0 - 15.0	5	74	5	5	120	5	5	880	29
			15.0 - 18.0	4	61	9	4	110	7	4	780	110
			18.0 - 21.0	4	64	11	4	120	9	4	830	68
		Mean Concentration	0.0 - 21.0	29	65	9	28	110	11	29	840	77

Location	UTM East Coordinate (m)	UTM North Coordinate (m)	Depth Interval (cm)	Number of Samples	^{226}Ra Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{232}Th Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{40}K Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})
CX2	483,751	4,415,127	0.0 - 0.3	4	44	15	4	94	4	4	690	60
			0.0 - 3.0	4	54	8	4	100	6	4	730	45
			3.0 - 6.0	6	55	4	6	100	15	6	750	52
			6.0 - 9.0	4.5	69	8	5	110	17	4	780	35
			9.0 - 12.0	3	56	5	3	96	22	3	760	52
			12.0 - 15.0	4	65	3	4	110	5	4	800	52
			15.0 - 18.0	4	59	4	4	110	5	4	810	61
			18.0 - 21.0	4	63	10	4	110	9	4	790	58
		Mean Concentration	0.0 - 21.0	29	60	8	28	110	9	29	770	52
CX3	484,048	4,414,882	0.0 - 0.3	4	51	18	4	63	6	4	590	57
			0.0 - 3.0	4	64	4	4	75	6	4	730	18
			3.0 - 6.0	6	55	8	6	76	6	6	710	69
			6.0 - 9.0	4	62	12	4	85	4	4	760	27
			9.0 - 12.0	4	62	5	4	80	4	4	810	41
			12.0 - 15.0	4	46	21	4	60	23	4	590	380
			15.0 - 18.0	4	64	8	4	81	2	4	780	25
			18.0 - 21.0	4	62	6	3	80	5	3	800	24
		Mean Concentration	0.0 - 21.0	28	61	8	28	79	6	29	740	150
CX4	484,477	4,414,651	0.0 - 0.3	4	58	10	4	92	5	4	710	64
			0.0 - 3.0	5	71	11	5	96	6	5	740	37
			3.0 - 6.0	5	67	7	5	100	7	5	770	32
			6.0 - 9.0	4	58	9	4	94	4	4	720	48
			9.0 - 12.0	4	68	10	4	100	7	4	730	35
			12.0 - 15.0	4	58	5	4	100	5	4	740	38
			15.0 - 18.0	4	70	5	4	100	6	4	770	57
			18.0 - 21.0	4	75	17	4	100	6	4	700	56
		Mean Concentration	0.0 - 21.0	30	67	9	30	110	6	30	740	45

Location	UTM East Coordinate (m)	UTM North Coordinate (m)	Depth Interval (cm)	Number of Samples	^{226}Ra Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{232}Th Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{40}K Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})
CX5	484,939	4,414,461	0.0 - 0.3	5	54	9	5	87	6	5	680	37
			0.0 - 3.0	4	67	11	4	100	3	4	790	20
			3.0 - 6.0	4	64	4	4	100	6	4	820	50
			6.0 - 9.0	5	67	6	5	100	5	5	810	34
			9.0 - 12.0	4	73	11	4	100	4	4	850	56
			12.0 - 15.0	4	70	5	4	100	5	4	840	51
			15.0 - 18.0	4	65	7	4	100	7	4	830	49
			18.0 - 21.0	4	63	8	4	96	4	4	790	72
		Mean Concentration	0.0 - 21.0	29	67	8	29	100	6	29	820	48
CX6	485,458	4,414,126	0.0 - 0.3	4	46	16	4	73	17	4	490	190
			0.0 - 3.0	4	53	9	4	83	9	4	630	63
			3.0 - 6.0	5	61	7	5	110	5	5	750	29
			6.0 - 9.0	4	61	6	4	110	6	4	790	36
			9.0 - 12.0	5	63	7	5	110	7	5	800	17
			12.0 - 15.0	4	57	6	4	110	4	4	820	44
			15.0 - 18.0	4	61	8	4	110	4	4	780	33
			18.0 - 21.0	4	64	7	4	100	5	4	780	27
		Mean Concentration	0.0 - 21.0	30	60	8	30	100	11	30	770	66
CX7	485,848	4,413,861	0.0 - 0.3	4	59	4	4	84	8	4	670	97
			0.0 - 3.0	5	69	10	5	96	5	5	780	74
			3.0 - 6.0	4	95	32	4	97	5	4	740	94
			6.0 - 9.0	4	75	10	4	98	5	4	760	12
			9.0 - 12.0	4	73	13	4	101	7	4	780	16
			12.0 - 15.0	5	68	12	5	97	5	5	770	22
			15.0 - 18.0	4	85	19	4	100	6	4	750	70
			18.0 - 21.0	4	69	13	4	91	4	4	750	63
		Mean Concentration	0.0 - 21.0	29	76	19	29	97	7	29	760	55

Location	UTM East Coordinate (m)	UTM North Coordinate (m)	Depth Interval (cm)	Number of Samples	^{226}Ra Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{232}Th Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{40}K Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})
DX1	486,280	4,415,274	0,0 - 0,3	4	50	11	4	67	10	4	460	96
			0,0 - 3,0	4	58	8	4	110	9	4	760	49
			3,0 - 6,0	1	68	6	1	130	4	1	790	20
			6,0 - 9,0	1	73	6	1	130	4	1	830	20
			9,0 - 12,0	1	75	6	1	130	4	1	800	20
			12,0 - 15,0	1	57	5	1	120	4	1	830	21
			15,0 - 18,0	2	74	4	2	110	3	2	740	6
			18,0 - 21,0	1	57	5	1	110	3	1	670	18
		Mean Concentration	0,0 - 21,0	4	69	7	4	120	11	4	760	52
DX2	486,708	4,415,256	0,0 - 0,3	4	43	3	4	78	8	4	470	68
			0,0 - 3,0	4	64	5	4	110	7	4	720	27
			3,0 - 6,0	1	72	6	1	130	10	1	790	20
			6,0 - 9,0	1	73	6	1	140	10	1	850	21
			9,0 - 12,0	1	70	6	1	130	10	1	790	20
			12,0 - 15,0	1	76	6	1	120	9	1	870	21
			15,0 - 18,0	1	82	6	1	140	10	1	870	21
			18,0 - 21,0	1	72	6	1	130	9	1	820	20
		Mean Concentration	0,0 - 21,0	4	71	7	4	120	7	4	780	55
DX3	487,713	4,415,095	0,0 - 0,3	4	55	11	4	92	11	4	590	64
			0,0 - 3,0	5	64	6	5	130	12	5	740	66
			3,0 - 6,0	1	72	10	1	140	17	1	700	20
			6,0 - 9,0	1	81	7	1	130	10	1	840	21
			9,0 - 12,0	1	74	9	1	150	18	1	760	31
			12,0 - 15,0	1	73	10	1	120	16	1	690	29
			15,0 - 18,0	1	72	6	1	140	10	1	810	20
			18,0 - 21,0	1	86	7	1	130	10	1	840	20
		Mean Concentration	0,0 - 21,0	4	75	7	4	130	14	4	780	50

Location	UTM East Coordinate (m)	UTM North Coordinate (m)	Depth Interval (cm)	Number of Samples	^{226}Ra Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{232}Th Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{40}K Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})
DX4	488,889	4,414,787	0.0 - 0.3	4	45	6	4	82	6	4	600	82
			0.0 - 3.0	6	76	5	6	110	6	6	780	28
			3.0 - 6.0	1	66	6	1	130	10	1	780	20
			6.0 - 9.0	1	73	6	1	130	10	1	860	21
			9.0 - 12.0	1	71	6	1	130	10	1	870	21
			12.0 - 15.0	1	74	6	1	130	10	1	880	21
			15.0 - 18.0	1	76	6	1	130	10	1	850	21
			18.0 - 21.0	1	71	6	1	120	9	1	860	21
		Mean Concentration	0.0 - 21.0	4	73	8	4	130	10	4	820	45
DX5	493,817	4,414,073	0.0 - 0.3	5	45	4	5	69	4	5	520	58
			0.0 - 3.0	4	53	7	4	85	5	4	600	32
			3.0 - 6.0	1	67	6	1	94	8	1	670	18
			6.0 - 9.0	1	46	4	1	91	8	1	620	18
			9.0 - 12.0	1	55	2	1	79	2	1	590	5
			12.0 - 15.0	1	54	5	1	93	8	1	570	17
			15.0 - 18.0	1	66	6	1	87	8	1	620	18
			18.0 - 21.0	1	60	5	1	85	8	1	550	17
		Mean Concentration	0.0 - 21.0	4	50	6	4	82	6	4	590	29
DX6	502,328	4,413,641	0.0 - 0.3	5	60	3	5	82	8	5	730	61
			0.0 - 3.0	5	64	7	5	87	7	5	750	62
			3.0 - 6.0	1	77	5	1	100	7	1	760	17
			6.0 - 9.0	1	72	5	1	99	7	1	740	16
			9.0 - 12.0	1	75	6	1	100	9	1	750	20
			12.0 - 15.0	1	74	6	1	100	9	1	700	19
			15.0 - 18.0	1	69	6	1	88	8	1	680	19
			18.0 - 21.0	1	65	6	1	93	8	1	640	18
		Mean Concentration	0.0 - 21.0	6	65	4	6	87	8	6	770	58

Location	UTM East Coordinate (m)	UTM North Coordinate (m)	Depth Interval (cm)	Number of Samples	^{226}Ra Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{232}Th Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{40}K Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})
EX1	486,357	4,416,887	0,0 - 0,3	6	60	4	6	92	3	6	670	48
			0,0 - 3,0	4	69	9	4	100	8	4	660	54
			3,0 - 6,0	1	62	5	1	100	9	1	730	19
			6,0 - 9,0	1	61	8	1	110	15	1	650	28
			9,0 - 12,0	1	92	12	1	120	16	1	750	30
			12,0 - 15,0	1	74	10	1	120	16	1	630	28
			15,0 - 18,0	1	66	5	1	90	8	1	670	18
			18,0 - 21,0	1	79	6	1	110	9	1	660	18
		Mean Concentration	0,0 - 21,0	4	67	3	4	100	9	4	660	13
EX2	487,051	4,417,255	0,0 - 0,3	4	37	8	4	65	14	4	480	37
			0,0 - 3,0	5	58	12	5	87	10	5	550	44
			3,0 - 6,0	1	65	6	1	90	8	1	610	18
			6,0 - 9,0	1	62	2	1	100	3	1	610	6
			9,0 - 12,0	1	76	6	1	110	9	1	620	18
			12,0 - 15,0	1	70	10	1	120	16	1	620	28
			15,0 - 18,0	1	67	6	1	100	9	1	670	18
			18,0 - 21,0	1	63	5	1	94	8	1	590	17
		Mean Concentration	0,0 - 21,0	4	66	7	4	110	15	4	610	46
EX3	487,415	4,417,761	0,0 - 0,3	3	46	14	3	65	16	3	530	130
			0,0 - 3,0	4	54	3	4	68	12	4	520	38
			3,0 - 6,0	1	60	5	1	98	8	1	650	18
			6,0 - 9,0	1	76	7	1	90	8	1	620	18
			9,0 - 12,0	1	57	5	1	90	8	1	700	19
			12,0 - 15,0	1	64	6	1	93	8	1	640	18
			15,0 - 18,0	1	50	8	1	83	14	1	690	33
			18,0 - 21,0	1	64	8	1	110	16	1	690	29
		Mean Concentration	0,0 - 21,0	4	61	8	4	88	12	4	630	31

Location	UTM East Coordinate (m)	UTM North Coordinate (m)	Depth Interval (cm)	Number of Samples	^{226}Ra Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{232}Th Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{40}K Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})
EX4	489,541	4,418,414	0.0 - 0.3	4	44	12	4	64	4	4	450	24
			0.0 - 3.0	5	59	13	5	76	9	5	550	37
			3.0 - 6.0	1	49	8	1	100	15	1	620	28
			6.0 - 9.0	2	46	7	2	92	11	2	520	56
			9.0 - 12.0	1	78	11	1	100	15	1	570	27
			12.0 - 15.0	1	73	6	1	77	7	1	540	17
			15.0 - 18.0	1	64	6	1	82	8	1	570	17
			18.0 - 21.0	1	66	6	1	81	8	1	550	17
		Mean Concentration	0.0 - 21.0	4	54	6	4	82	5	4	530	11
EX5	493,780	4,422,426	0.0 - 0.3	4	31	4	4	35	3	4	340	33
			0.0 - 3.0	5	38	5	5	46	3	5	360	43
			3.0 - 6.0	1	43	5	1	49	6	1	390	14
			6.0 - 9.0	1	46	1	1	47	2	1	410	4
			9.0 - 12.0	1	36	4	1	41	6	1	360	14
			12.0 - 15.0	1	40	4	1	43	6	1	370	14
			15.0 - 18.0	1	43	5	1	40	5	1	360	14
			18.0 - 21.0	1	46	5	1	45	6	1	400	14
		Mean Concentration	0.0 - 21.0	4	33	5	4	41	5	4	360	29
EX6	497,942	4,424,872	0.0 - 0.3	4	63	7	4	87	8	4	640	62
			0.0 - 3.0	4	72	9	4	100	5	4	740	48
			3.0 - 6.0	1	76	6	1	130	9	1	790	20
			6.0 - 9.0	1	89	7	1	120	9	1	810	20
			9.0 - 12.0	1	82	6	1	120	9	1	880	21
			12.0 - 15.0	0	*	*	0	*	*	0	*	*
			15.0 - 18.0	1	60	5	1	120	10	1	900	21
			18.0 - 21.0	1	74	6	1	110	9	1	870	21
		Mean Concentration	0.0 - 21.0	4	67	8	4	100	17	4	720	140

Location	UTM East Coordinate (m)	UTM North Coordinate (m)	Depth Interval (cm)	Number of Samples	^{226}Ra Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{232}Th Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{40}K Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})
FX1	486,311	4,413,733	0.0 - 0.3	6	72	17	6	100	5	6	680	38
			0.0 - 3.0	5	88	14	5	110	3	5	720	30
			3.0 - 6.0	1	80	6	1	100	9	1	630	18
			6.0 - 9.0	0	*	*	0	*	*	0	*	*
			9.0 - 12.0	1	92	7	1	140	10	1	830	21
			12.0 - 15.0	1	82	7	1	120	9	1	860	21
			15.0 - 18.0	1	82	7	1	120	9	1	810	20
			18.0 - 21.0	1	72	6	1	130	10	1	820	20
		Mean Concentration	0.0 - 21.0	4	72	6	4	110	9	4	760	40
FX2	486,769	4,413,573	0.0 - 0.3	4	68	24	4	76	8	4	570	51
			0.0 - 3.0	4	68	9	4	90	5	4	700	25
			3.0 - 6.0	1	73	6	1	97	8	1	760	20
			6.0 - 9.0	1	68	6	1	88	8	1	770	20
			9.0 - 12.0	1	95	7	1	94	8	1	800	21
			12.0 - 15.0	1	98	7	1	120	9	1	720	19
			15.0 - 18.0	1	84	11	1	130	16	1	650	28
			18.0 - 21.0	1	110	13	1	140	17	1	690	29
		Mean Concentration	0.0 - 21.0	4	86	26	4	98	24	4	730	46
FX3	487,813	4,412,962	0.0 - 0.3	4	90	44	4	84	11	4	550	75
			0.0 - 3.0	4	81	17	4	100	15	4	650	60
			3.0 - 6.0	1	79	6	1	99	8	1	680	18
			6.0 - 9.0	1	98	11	1	130	16	1	720	29
			9.0 - 12.0	2	49	5	2	67	8	2	420	77
			12.0 - 15.0	1	68	5	1	95	7	1	590	15
			15.0 - 18.0	1	58	4	1	86	7	1	550	14
			18.0 - 21.0	1	63	5	1	92	7	1	570	15
		Mean Concentration	0.0 - 21.0	4	74	12	4	110	3	4	700	40

Location	UTM East Coordinate (m)	UTM North Coordinate (m)	Depth Interval (cm)	Number of Samples	^{226}Ra Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{232}Th Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{40}K Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})
FX4	489,490	4,411,397	0.0 - 0.3	5	63	13	5	68	13	5	580	28
			0.0 - 3.0	4	75	19	4	79	13	4	590	16
			3.0 - 6.0	1	42	4	1	71	7	1	630	18
			6.0 - 9.0	1	76	10	1	96	14	1	580	27
			9.0 - 12.0	1	78	11	1	96	14	1	620	28
			12.0 - 15.0	1	61	6	1	67	7	1	510	16
			15.0 - 18.0	1	54	5	1	66	7	1	610	18
			18.0 - 21.0	1	79	11	1	97	14	1	610	28
		Mean Concentration	0.0 - 21.0	4	59	12	4	82	18	4	600	28
FX5	491,032	4,409,904	0.0 - 0.3	4	68	14	4	84	11	4	540	78
			0.0 - 3.0	4	65	13	4	100	18	4	670	92
			3.0 - 6.0	1	77	10	1	120	16	1	670	29
			6.0 - 9.0	1	69	6	1	110	9	1	730	19
			9.0 - 12.0	1	75	6	1	100	8	1	780	20
			12.0 - 15.0	1	85	6	1	100	9	1	760	20
			15.0 - 18.0	1	92	11	1	130	16	1	710	30
			18.0 - 21.0	1	73	10	1	140	17	1	710	30
		Mean Concentration	0.0 - 21.0	4	82	14	4	120	18	4	670	12
FX6	500,234	4,408,098	0.0 - 0.3	4	64	9	4	81	6	4	670	72
			0.0 - 3.0	5	74	17	5	83	9	5	700	33
			3.0 - 6.0	1	60	6	1	80	8	1	620	18
			6.0 - 9.0	1	56	5	1	79	8	1	590	17
			9.0 - 12.0	1	57	5	1	73	7	1	500	16
			12.0 - 15.0	1	68	6	1	61	7	1	540	17
			15.0 - 18.0	1	39	4	1	58	6	1	490	16
			18.0 - 21.0	1	59	6	1	56	6	1	510	16
		Mean Concentration	0.0 - 21.0	4	58	6	4	74	18	4	630	83

Location	UTM East Coordinate (m)	UTM North Coordinate (m)	Depth Interval (cm)	Number of Samples	^{226}Ra Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{232}Th Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{40}K Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})
GX1	484,670	4,412,884	0.0 - 0.3	4	41	1	4	62	4	4	430	33
			0.0 - 3.0	4	63	7	4	94	3	4	610	33
			3.0 - 6.0	1	71	10	1	120	16	1	610	28
			6.0 - 9.0	1	50	5	1	86	8	1	630	18
			9.0 - 12.0	1	71	6	1	92	8	1	700	19
			12.0 - 15.0	1	66	6	1	90	8	1	600	17
			15.0 - 18.0	1	60	5	1	94	8	1	560	16
			18.0 - 21.0	1	54	5	1	91	8	1	580	17
		Mean Concentration	0.0 - 21.0	4	64	5	4	91	4	4	610	34
GX2	485,076	4,412,190	0.0 - 0.3	5	61	6	5	76	7	5	540	58
			0.0 - 3.0	5	76	10	5	91	12	5	650	47
			3.0 - 6.0	1	87	7	1	85	8	1	660	18
			6.0 - 9.0	1	77	6	1	80	8	1	640	18
			9.0 - 12.0	1	86	7	1	81	8	1	650	18
			12.0 - 15.0	1	82	6	1	81	8	1	660	18
			15.0 - 18.0	1	69	6	1	91	8	1	640	18
			18.0 - 21.0	1	76	10	1	110	16	1	630	28
		Mean Concentration	0.0 - 21.0	4	81	6	4	88	6	4	640	34
GX3	485,749	4,411,663	0.0 - 0.3	5	73	11	5	86	4	5	640	38
			0.0 - 3.0	4	70	9	4	94	5	4	670	9
			3.0 - 6.0	1	84	6	1	97	8	1	680	19
			6.0 - 9.0	1	62	5	1	94	8	1	650	18
			9.0 - 12.0	1	82	6	1	110	9	1	640	18
			12.0 - 15.0	1	78	6	1	95	8	1	650	18
			15.0 - 18.0	1	79	6	1	98	8	1	660	18
			18.0 - 21.0	1	90	7	1	100	9	1	650	18
		Mean Concentration	0.0 - 21.0	4	75	7	4	96	2	4	640	31

Location	UTM East Coordinate (m)	UTM North Coordinate (m)	Depth Interval (cm)	Number of Samples	^{226}Ra Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{232}Th Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{40}K Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})
GX4	485,926	4,409,440	0.0 - 0.3	5	49	4	5	66	7	5	550	54
			0.0 - 3.0	5	64	6	5	77	4	5	640	29
			3.0 - 6.0	1	70	6	1	81	8	1	670	18
			6.0 - 9.0	1	62	6	1	87	8	1	670	18
			9.0 - 12.0	1	60	3	1	73	4	1	670	10
			12.0 - 15.0	1	65	6	1	81	8	1	680	19
			15.0 - 18.0	1	58	5	1	85	8	1	690	19
			18.0 - 21.0	1	61	5	1	87	8	1	650	18
		Mean Concentration	0.0 - 21.0	4	62	3	4	78	2	4	650	22
GX5	489,345	4,405,011	0.0 - 0.3	4	45	8	4	63	5	4	540	40
			0.0 - 3.0	4	53	2	4	79	8	4	650	42
			3.0 - 6.0	1	53	5	1	81	8	1	660	18
			6.0 - 9.0	2	54	8	2	83	3	2	730	5
			9.0 - 12.0	1	55	5	1	86	8	1	760	20
			12.0 - 15.0	1	50	5	1	97	8	1	780	20
			15.0 - 18.0	1	52	5	1	82	8	1	790	20
			18.0 - 21.0	1	40	4	1	78	8	1	800	20
		Mean Concentration	0.0 - 21.0	4	48	6	4	84	13	4	720	49
GX6	493,335	4,400,160	0.0 - 0.3	4	40	3	4	60	4	4	540	60
			0.0 - 3.0	4	56	5	4	81	5	4	740	44
			3.0 - 6.0	1	64	3	1	82	4	1	750	10
			6.0 - 9.0	1	66	6	1	94	8	1	810	20
			9.0 - 12.0	1	66	6	1	78	7	1	700	19
			12.0 - 15.0	1	58	5	1	84	8	1	720	19
			15.0 - 18.0	1	67	6	1	96	8	1	720	19
			18.0 - 21.0	1	66	6	1	100	9	1	790	20
		Mean Concentration	0.0 - 21.0	4	59	3	4	85	6	4	740	45

Location	UTM East Coordinate (m)	UTM North Coordinate (m)	Depth Interval (cm)	Number of Samples	^{226}Ra Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{232}Th Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{40}K Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})
K01	478,656	4,423,727	0.0 - 0.3	0	*	*	0	*	*	0	*	*
Boulder			0.0 - 3.0	1	57	6	1	130	9	1	1,300	26
		Mean Concentration	0.0 - 21.0	2	68	6	2	140	9	2	1,200	400
K02	486,018	4,397,711	0.0 - 0.3	0	*	*	0	*	*	0	*	*
Golden			0.0 - 3.0	1	52	5	1	80	8	1	780	20
		Mean Concentration	0.0 - 21.0	1	82	6	1	81	8	1	840	21
K03	480,047	4,422,093	0.0 - 0.3	0	*	*	0	*	*	0	*	*
Marshall			0.0 - 3.0	1	68	6	1	110	9	1	770	20
		Mean Concentration	0.0 - 21.0	1	102	8	1	120	9	1	810	20
K04	477,952	4,420,840	0.0 - 0.3	0	*	*	0	*	*	0	*	*
Eldorado Springs			0.0 - 3.0	1	89	7	1	130	10	1	900	21
		Mean Concentration	0.0 - 21.0	0	*	*	0	*	*	0	*	*
K05	490,677	4,401,214	0.0 - 0.3	0	*	*	2	*	*	2	*	*
Wheatridge			0.0 - 3.0	1	84	7	1	110	9	1	810	20
		Mean Concentration	0.0 - 21.0	1	99	7	1	120	9	1	950	22
K06	491,362	4,405,615	0.0 - 0.3	0	*	*	0	*	*	0	*	*
Arvada			0.0 - 3.0	2	85	35	2	87	15	2	730	13
		Mean Concentration	0.0 - 21.0	1	60	5	1	78	8	1	670	19

Location	UTM East Coordinate (m)	UTM North Coordinate (m)	Depth Interval (cm)	Number of Samples	^{226}Ra Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{232}Th Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{40}K Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})
K07	497,184	4,410,443	0.0 - 0.3	0	*	*	0	*	*	0	*	*
Westminster			0.0 - 3.0	1	62	5	1	76	7	1	700	19
		Mean Concentration	0.0 - 21.0	1	64	6	1	72	7	1	610	18
K08	503,091	4,414,425	0.0 - 0.3	0	*	*	0	*	*	0	*	*
Thornton/ Northglenn			0.0 - 3.0	1	42	4	1	54	6	1	510	16
		Mean Concentration	0.0 - 21.0	1	74	6	1	70	7	1	510	16
K09	492,293	4,417,808	0.0 - 0.3	0	*	*	0	*	*	0	*	*
Broomfield			0.0 - 3.0	1	57	5	1	98	8	1	840	21
		Mean Concentration	0.0 - 21.0	1	66	6	1	100	9	1	940	22
K10	484,127	4,422,542	0.0 - 0.3	0	*	*	0	*	*	0	*	*
Superior			0.0 - 3.0	2	71	16	2	110	28	2	690	46
		Mean Concentration	0.0 - 21.0	1	59	6	1	110	9	1	680	26
Z01	520,524	4,516,126	0.0 - 0.3	1	80	10	1	84	13	1	340	13
Pawnee Grassland, Weld Co			0.0 - 3.0	1	92	13	1	97	16	1	360	16
		Mean Concentration	0.0 - 21.0	1	42	5	1	61	7	1	830	20

Location	UTM East Coordinate (m)	UTM North Coordinate (m)	Depth Interval (cm)	Number of Samples	^{226}Ra Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{232}Th Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})	Number of Samples	^{40}K Mean (Bq kg^{-1})	Std Dev (Bq kg^{-1})
Z02	487,734	4,494,329	0.0 - 0.3	1	63	6	1	50	6	1	480	16
CSU Airport, Weld Co			0.0 - 3.0	1	120	15	1	110	13	1	260	12
		Mean Concentration	0.0 - 21.0	1	90	7	1	78	8	1	650	18
Z03	505,381	4,503,233	0.0 - 0.3	1	64	6	1	56	7	1	640	18
Weld/Larimer Co Line			0.0 - 3.0	1	120	12	1	86	14	1	280	12
		Mean Concentration	0.0 - 21.0	1	85	11	1	92	14	1	290	12
Z04	487,947	4,488,793	0.0 - 0.3	1	51	3	1	48	3	1	460	8
Dixon Reservoir, Larimer Co			0.0 - 3.0	1	72	6	1	58	7	1	480	16
		Mean Concentration	0.0 - 21.0	1	72	6	1	60	7	1	500	16
Z05	489,303	4,484,305	0.0 - 0.3	1	100	7	1	60	7	1	530	16
Southwest Fort Collins			0.0 - 3.0	1	130	8	1	64	7	1	530	16
		Mean Concentration	0.0 - 21.0	1	110	7	1	69	7	1	550	17
Z06	494,866	4,483,200	0.0 - 0.3	1	42	5	1	51	6	1	550	17
Southeast Fort Collins			0.0 - 3.0	1	62	6	1	58	7	1	650	18
		Mean Concentration	0.0 - 21.0	1	74	6	1	74	7	1	710	19

Location	UTM East Coordinate (m)	UTM North Coordinate (m)	Depth Interval (cm)	Number of Samples	^{226}Ra Mean ($Bq\ kg^{-1}$)	Std Dev ($Bq\ kg^{-1}$)	Number of Samples	^{232}Th Mean ($Bq\ kg^{-1}$)	Std Dev ($Bq\ kg^{-1}$)	Number of Samples	^{40}K Mean ($Bq\ kg^{-1}$)	Std Dev ($Bq\ kg^{-1}$)
Z07	513,609	4,295,761	0.0 - 0.3	1	72	6	1	95	8	1	690	19
Colorado Springs			0.0 - 3.0	1	110	7	1	180	11	1	920	22
		Mean Concentration	0.0 - 21.0	1	82	2	1	160	3	1	890	6
Z08	501,481	4,359,837	0.0 - 0.3	1	81	6	1	110	9	1	1,200	25
Castle Rock			0.0 - 3.0	1	98	7	1	150	11	1	1,500	28
		Mean Concentration	0.0 - 21.0	1	110	7	1	160	11	1	1,600	29
Z09	511,224	4,336,527	0.0 - 0.3	1	63	6	1	59	7	1	1,700	29
Monument			0.0 - 3.0	3	69	25	3	86	32	3	1,400	830
		Mean Concentration	0.0 - 21.0	1	68	6	1	93	8	1	2,000	32
Z10	476,416	4,438,567	0.0 - 0.3	3	66	29	3	75	30	3	540	27
Left-hand Valley Reservoir Boulder, Co			0.0 - 3.0	2	5	3	2	5	1	2	40	13
		Mean Concentration	0.0 - 21.0	2	52	70	2	40	48	2	340	430

Appendix C

Soil Deposition Estimates From In-situ Measurements for ^{241}Am and ^{137}Cs

With Soil Property Data

Location	Distance To 903 Pad km	UTM East Coordinate	UTM North Coordinate	Wet Bulk Density g cm^{-3}	Soil Moisture Fraction	Rock Fraction	^{241}Am Deposition KBq m^{-2}	Std Dev KBq m^{-2}	^{137}Cs Deposition KBq m^{-2}	Std Dev KBq m^{-2}
AN2	0.41	483,799	4,415,396	1.73	0.12	0.12	38	1.9	5.5	0.35
AN4	1.21	484,602	4,415,474	1.65	0.11	0.11	46	7.4	*	*
AN5	1.76	485,128	4,415,649	1.53	0.15	0.13	2.1	0.57	4.2	0.19
AN6	2.45	485,804	4,415,798	1.58	0.21	0.18	2.1	0.57	5.1	0.24
AS1	0.25	483,646	4,415,249	*	*	*	66	2.3	3.9	0.22
AS3	0.72	484,098	4,415,143	1.46	0.11	0.16	9.4	1.0	4.3	0.22
AS4	1.27	484,665	4,415,161	1.62	0.06	0.09	2.1	0.60	5.4	0.26
AS5	1.73	485,123	4,415,108	1.54	0.04	0.05	2.0	0.52	4.5	0.21
AS6	2.43	485,775	4,414,813	1.41	0.10	0.07	1.9	0.52	4.9	0.27
AX1	0.26	483,658	4,415,301	1.60	0.13	0.11	195	3.7	4.9	0.20
AX2	0.39	483,792	4,415,305	1.47	0.09	0.11	62	1.7	4.6	0.25
AX3	0.72	484,124	4,415,316	1.12	0.11	0.10	0.87	0.33	1.3	0.44
AX4	1.24	484,642	4,415,326	1.42	0.12	0.11	5.8	0.82	5.4	0.31
AX5	1.73	485,127	4,415,346	1.64	0.18	0.15	2.8	0.55	4.5	0.26
AX6	2.39	485,785	4,415,302	1.43	0.15	0.16	2.2	0.47	5.1	0.26
BN2	1.24	484,333	4,416,126	1.32	0.08	0.11	2.3	0.54	3.3	0.18
BN3	1.65	484,645	4,416,390	1.26	0.09	0.08	0.51	0.25	3.6	0.15
BN4	2.47	485,271	4,416,927	1.35	0.09	0.09	0.88	0.36	4.2	0.19
BN5	2.96	485,633	4,417,252	1.67	0.12	0.10	2.3	0.65	4.1	0.18
BS2	1.17	484,511	4,415,683	1.81	0.18	0.15	1.2	0.41	6.5	0.20
BS3	1.66	484,968	4,415,866	2.00	0.14	0.16	2.3	0.66	7.5	0.34
BS4	2.40	485,637	4,416,191	1.90	0.12	0.13	1.4	0.46	5.3	0.25
BX1	0.83	484,104	4,415,741	1.67	0.18	0.15	1.9	0.65	5.0	0.25
BX2	1.15	484,423	4,415,827	1.84	0.19	0.19	1.4	0.61	6.8	0.39
BX3	1.66	484,877	4,416,074	1.67	0.20	0.20	1.8	0.50	4.9	0.27
BX4	2.41	485,488	4,416,513	1.25	0.06	0.07	0.77	0.30	3.5	0.17
BX5	2.82	485,808	4,416,774	1.25	0.04	0.05	1.8	0.50	4.7	0.20
CN1	0.26	483,640	4,415,205	1.36	0.15	0.15	34	1.6	3.5	0.19
CN2	0.41	483,783	4,415,162	1.44	0.12	0.14	7.7	0.94	3.4	0.21
CN3	0.75	484,072	4,414,968	1.54	0.12	0.12	5.4	0.68	3.4	0.17
CN4	1.26	484,578	4,414,870	1.47	0.08	0.10	2.5	0.53	5.1	0.22
CN5	1.70	484,988	4,414,691	1.50	0.12	0.10	1.6	0.49	3.2	0.20
CN6	2.27	485,537	4,414,557	1.65	0.13	0.13	2.2	0.55	4.2	0.24

Location	Distance To 903 Pad km	UTM East Coordinate	UTM North Coordinate	Wet Bulk Density g cm^{-3}	Soil Moisture Fraction	Rock Fraction	^{241}Am Deposition KBq m^{-2}	Std Dev KBq m^{-2}	^{137}Cs Deposition KBq m^{-2}	Std Dev KBq m^{-2}
CS1	0.24	483,575	4,415,144	1.78	0.15	0.14	33	1.8	3.4	0.17
CS2	0.39	483,706	4,415,067	1.95	0.17	0.16	16	1.3	3.6	0.20
CS3	0.76	483,948	4,414,787	1.77	0.09	0.13	1.8	0.45	4.7	0.20
CS4	1.24	484,339	4,414,493	1.41	0.06	0.07	1.2	0.38	5.0	0.29
CS5	1.73	484,707	4,414,181	1.64	0.15	0.10	0.88	0.40	2.2	0.56
CS6	2.33	485,159	4,413,787	1.76	0.20	0.17	1.4	0.44	1.3	0.13
CS7	2.90	485,686	4,413,527	1.48	0.18	0.19	0.49	0.26	2.0	0.16
CX1	0.25	483,618	4,415,182	1.76	0.12	0.15	42	2.1	4.3	0.25
CX2	0.40	483,751	4,415,127	1.75	0.08	0.10	9.3	0.93	3.1	0.17
CX3	0.78	484,048	4,414,882	1.41	0.09	0.08	1.9	0.51	3.4	0.16
CX4	1.26	484,477	4,414,651	1.38	0.12	0.10	0.86	0.36	2.6	0.16
CX5	1.76	484,939	4,414,461	1.67	0.15	0.14	0.31	0.48	4.7	0.27
CX6	2.37	485,458	4,414,126	1.70	0.22	0.19	0.71	0.32	4.6	0.25
CX7	2.84	485,848	4,413,861	1.50	0.26	0.24	0.9	0.49	1.6	0.28
DE2	3.79	487,184	4,415,453	1.53	0.12	0.19	*	*	*	*
DN1	3.17	486,535	4,415,812	1.38	0.06	0.09	0.53	0.24	5.0	0.23
DN2	3.51	486,876	4,415,823	1.29	0.09	0.08	*	*	1.4	0.29
DN3	4.37	487,691	4,416,154	1.40	0.05	0.07	*	*	*	*
DN4	7.32	490,610	4,416,564	1.48	0.09	0.07	0.90	0.34	2.7	0.16
DS1	2.62	485,985	4,414,881	1.22	0.11	0.12	1.8	0.37	3.4	0.17
DS2	3.41	486,780	4,414,896	*	*	*	1.6	0.50	6.2	0.26
DS3	4.53	487,855	4,414,498	1.50	0.07	0.09	1.5	0.48	5.4	0.26
DS5	10.49	493,523	4,412,557	1.57	0.03	0.05	*	*	*	*
DX1	2.88	486,280	4,415,274	1.43	0.07	0.05	1.6	0.50	6.1	0.32
DX2	3.31	486,708	4,415,256	1.55	0.08	0.07	1.6	0.48	6.3	0.30
DX3	4.32	487,713	4,415,095	1.56	0.03	0.05	*	*	6.7	0.33
DX4	5.51	488,889	4,414,787	1.71	0.03	0.03	1.3	0.42	5.9	0.27
DX5	10.49	493,817	4,414,073	1.54	0.02	0.03	2.1	0.41	4.1	0.20
DX6	19.00	502,328	4,413,641	1.67	0.19	0.16	1.8	0.58	3.9	0.20
EE2	5.23	487,743	4,418,232	1.81	0.12	0.16	*	*	*	*
EN1	3.67	486,344	4,417,508	1.70	0.13	0.12	2.6	0.73	5.6	0.23
EN2	5.06	487,552	4,418,198	*	*	*	*	*	*	*
EN3	5.57	487,638	4,418,917	*	*	*	*	*	*	*
EN4	8.42	489,931	4,420,631	1.65	0.10	0.12	-0.09	0.08	3.9	0.19

Location	Distance To 903 Pad km	UTM East Coordinate	UTM North Coordinate	Wet Bulk Density g cm^{-3}	Soil Moisture Fraction	Rock Fraction	^{241}Am Deposition KBq m^{-2}	Std Dev KBq m^{-2}	^{137}Cs Deposition KBq m^{-2}	Std Dev KBq m^{-2}
ES1	3.28	486,478	4,416,444	1.58	0.11	0.11	-0.26	0.27	3.6	0.17
ES2	3.66	486,820	4,416,600	1.43	0.08	0.09	-0.29	0.30	3.8	0.18
ES3	4.09	487,234	4,416,734	1.72	0.11	0.09	0.89	0.40	6.4	0.31
ES4	6.01	489,107	4,417,196	1.79	0.14	0.13	0.70	0.30	3.0	0.16
ES6	18.48	500,216	4,422,975	1.36	0.14	0.14	0.83	0.27	1.7	0.33
EX1	3.35	486,357	4,416,887	*	*	*	1.1	0.38	2.4	0.15
EX2	4.14	487,051	4,417,255	*	*	*	1.0	0.39	6.6	0.25
EX3	4.70	487,415	4,417,761	*	*	*	0.72	0.33	7.8	0.31
EX4	6.88	489,541	4,418,414	1.63	0.15	0.15	1.2	0.45	5.4	0.25
EX5	12.58	493,780	4,422,426	1.53	0.10	0.13	0.75	0.31	2.3	0.12
EX6	17.40	497,942	4,424,872	1.76	0.04	0.07	1.7	0.54	6.3	0.31
FN1	3.13	486,296	4,414,131	2.33	0.04	0.04	0.90	0.41	6.3	0.31
FN2	3.44	486,570	4,413,969	2.11	0.11	0.07	1.5	0.46	3.5	0.19
FN3	5.20	488,165	4,413,219	1.54	0.10	0.10	0.09	0.06	5.1	0.26
FN4	6.79	489,993	4,413,707	1.50	0.05	0.07	0.84	0.34	4.1	0.19
FS1	3.84	486,333	4,412,830	1.72	0.11	0.08	0.85	0.39	1.7	0.42
FS3	5.34	487,541	4,411,939	1.81	0.12	0.12	0.21	0.13	4.9	0.22
FS4	6.86	488,985	4,411,331	1.76	0.15	0.14	0.84	0.36	5.0	0.24
FX1	3.31	486,311	4,413,733	1.59	0.15	0.15	1.1	0.40	4.0	0.22
FX2	3.79	486,769	4,413,573	1.57	0.15	0.15	2.7	0.59	4.6	0.24
FX3	5.00	487,813	4,412,962	1.71	0.21	0.18	0.34	0.19	3.8	0.20
FX4	7.24	489,490	4,411,397	1.69	0.20	0.21	0.04	0.04	1.9	0.12
FX5	9.35	491,032	4,409,904	1.66	0.18	0.19	1.3	0.42	1.8	0.13
FX6	18.31	500,234	4,408,098	1.63	0.13	0.15	1.6	0.49	1.2	0.40
GE3	4.51	485,940	4,411,585	1.76	0.11	0.12	1.4	0.48	3.0	0.16
GN1	3.21	485,391	4,412,798	1.88	0.12	0.12	-0.63	0.78	1.4	0.32
GN2	3.71	485,760	4,412,442	1.52	0.12	0.12	1.3	0.42	1.4	0.40
GN3	5.36	487,271	4,411,607	1.43	0.14	0.13	0.27	0.18	3.8	0.19
GN4	6.38	487,190	4,410,174	1.83	0.16	0.15	*	*	2.5	0.18
GN5	10.91	491,646	4,408,168	1.65	0.10	0.13		*		
GS1	2.66	484,349	4,412,821	1.42	0.09	0.09	0.84	0.36	4.7	0.27
GS2	3.09	484,565	4,412,443	1.64	0.11	0.10	0.73	0.37	2.6	0.16
GS3	5.25	484,599	4,410,198	1.48	0.14	0.11	0.89	0.37	2.4	0.14
GS5	9.51	486,312	4,406,260	1.11	0.11	0.13	*	*	*	*

Location	Distance To 903 Pad km	UTM East Coordinate	UTM North Coordinate	Wet Bulk Density g cm^{-3}	Soil Moisture Fraction	Rock Fraction	^{241}Am Deposition KBq m^{-2}	Std Dev KBq m^{-2}	^{137}Cs Deposition KBq m^{-2}	Std Dev KBq m^{-2}
GS6	*	*	*	1,45	0,07	0,09	*	*	*	*
GW3	3.87	484,344	4,411,562	*	*	*	1,0	0,36	3,1	0,15
GX1	2.74	484,670	4,412,884	1,74	0,13	0,10	1,1	0,45	5,9	0,22
GX2	3.54	485,076	4,412,190	1,67	0,17	0,15	*	*	3,7	0,17
GX3	4.34	485,749	4,411,663	1,41	0,12	0,15	0,67	0,28	1,6	0,38
GX4	6.39	485,926	4,409,440	1,16	0,13	0,13	0,29	0,16	2,1	0,11
GX5	11.89	489,345	4,405,011	1,01	0,11	0,12	0,14	0,09	3,0	0,15
GX6	18.12	493,335	4,400,160	1,30	0,12	0,12	0,33	0,20	1,3	0,44
K01	9.66	478,656	4,423,727	1,74	0,17	0,14	1,9	0,54	3,8	0,24
K02	17.79	486,018	4,397,711	1,57	0,20	0,18	0,69	0,32	3,3	0,20
K03	7.57	480,047	4,422,093	1,54	0,17	0,18	0,89	0,37	3,5	0,18
K04	7.76	477,952	4,420,840	2,00	0,13	0,15	3,06	0,68	6,5	0,36
K05	15.86	490,677	4,401,214	1,78	0,18	0,15	1,1	0,37	3,6	0,23
K06	12.55	491,362	4,405,615	1,46	0,25	0,22	2,1	0,56	2,6	0,16
K07	14.62	497,184	4,410,443	1,73	0,23	0,24	0,92	0,36	3,8	0,20
K08	19.71	503,091	4,414,425	1,47	0,22	0,23	0,23	0,17	2,6	0,16
K09	9.24	492,293	4,417,808	1,30	0,19	0,21	0,67	0,31	3,8	0,23
K10	7.27	484,127	4,422,542	1,73	0,14	0,17	1,1	0,45	6,1	0,34
Z01	107.43	520,524	4,516,126	1,65	.09	.09	0,47	0,25	3,9	0,28
Z02	79.14	487,734	4,494,329	*	*	*	1,1	0,65	5,4	1,5
Z03	90.63	505,381	4,503,233	1,52	0,11	0,11	1,6	0,74	6,2	0,14
Z04	73.62	487,947	4,488,793	1,17	0,09	0,08	0,37	0,09	4,4	0,21
Z05	69.25	489,303	4,484,305	1,12	0,10	0,10	0,19	0,19	5,5	0,68
Z06	68.85	494,866	4,483,200	1,45	0,11	0,11	0,85	0,48	5,4	0,17
Z07	123.31	513,609	4,295,761	1,39	0,09	0,08	1,1	0,37	5,9	0,27
Z08	58.35	501,481	4,359,837	1,57	0,14	0,12	2,2	0,82	6,1	0,29
Z09	83.55	511,224	4,336,527	1,84	0,16	0,16	3,2	3,13	5,8	0,43
Z10	24.28	476,416	4,438,567	1,70	0,16	0,17	0,46	0,65	5,1	0,44

Appendix D

Calibration Procedures for the In-situ Gamma Spectrometer

A procedure developed by Beck et al. (1972) and Helfer and Miller (1988) was used with modifications described in ICRU Report 53 (1994) to estimate soil depositions from in-situ gamma spectroscopy measurements. Using the notation in ICRU Report 53 for the full energy peak count rate at a particular energy, the detector calibration equation is,

$$\frac{\dot{N}}{A_x} = \frac{\dot{N}}{\dot{N}_0} \cdot \frac{\dot{N}_0}{\varphi} \cdot \frac{\varphi}{A_x}. \qquad (4)$$

- $\left(\frac{\dot{N}}{\dot{N}_0}\right)$ - angular correction factor,
- $\left(\frac{\dot{N}_0}{\varphi}\right)$ - full-energy peak count rate per unit fluence rate at a particular energy (cpm γ^{-1} m^2 s), and
- $\left(\frac{\varphi}{A_x}\right)$ - fluence rate per unit activity per unit area (γ s^{-1} kBq^{-1}).

The first two factors were determined experimentally and then compared with published data for similar detectors. The third, based on the relaxation mass determined by dividing soil density by the inverse relaxation length, which is a measure of the source distribution was determined from tables listed in ICRU Report 53 (1994). Soil depositions (kBq m^{-2}) were estimated from in-situ measurements by dividing time-adjusted counts (cpm) under the photopeak by the full energy peak count rate, $\left(\frac{\dot{N}}{A_x}\right)$ (cpm kBq^{-1} m^2), for a particular radionuclide.

Soil depositions (kBq m^{-2}) were estimated from laboratory analyses using soil densities to convert ^{137}Cs and ^{241}Am (Hulse et al. 1999), and $^{239,240}Pu$ soil concentrations (Webb 1996) (kBq kg^{-1}) for 3 cm depth intervals to incremental depositions. Incremental soil depositions for each microplot within macroplots were summed from the surface to a depth of 21 cm and then averaged to obtain the total deposition for each macroplot. Soil concentrations of ^{137}Cs and, ^{241}Am were at or below detection limits in the 18 to 21 cm sample interval at most locations where soil samples were taken. Webb (1994) reported that soil concentrations of $^{239,240}Pu$ were also close to or below the detection limit in the 18 to 21 cm sample interval.

Both the angular correction factor and the full-energy peak count rate per unit fluence rate depend on detector characteristics and response to photons at specific energy levels. These two factors were determined experimentally. The fluence rate per unit activity per unit area depends on photon energies, attenuation effects of soil and air (density), and source distributions including the rate at which soil concentrations of ^{137}Cs and ^{241}Am decrease with depth, and is therefore site dependent. This factor was calculated for each macroplot using measured wet bulk soil densities when available and average rates at which soil concentrations of ^{137}Cs and ^{241}Am decrease with depth that were previously determined by Hulse et al. (1999). The average of all measured wet bulk densities was used at locations where density measurements were unavailable.

Angular Correction Factor

Angular correction factors depend on photon energy and compensate for a detector's heterogeneous response to photons incident on the detector at varying angles relative to the detector's vertical axis. To estimate the angular correction factor, the detector was positioned one meter above a vertical, quarter-circle wooden frame while count rates for ^{137}Cs (661.7 keV) and ^{241}Am (59.5 keV) point sources were measured. Measurements were repeated as the point sources were rotated upward along the frame, around the detector from a point directly below the center of the detector (0°) to a horizontal position (90°) in 5° increments. The frame kept point sources one meter from the center of the detector's downward facing surface. Count rates at each 5° increment were normalized to the count rate when each source was directly below the detector at 0°. Measurements were repeated with the wooden frame oriented in three different azimuthal directions separated by 120° on a horizontal plain. Angular response factors for ^{137}Cs and ^{241}Am were determined by numerically integrating functions that were fit to the average normalized count rates for the three azimuthal directions, which have been plotted in Figure 2. A 127 MBq ^{137}Cs source, manufactured by the ICN Chemical & Radioisotope Division of International Chemical & Nuclear Corporation[23], and a 439 kBq ^{241}Am source manufactured by Amersham Buchler GmbH & Co KG[24] were used to estimate the angular response factors. Both sources are traceable to standards at the National Institute of Standards Technology (NIST).

Full-Energy Peak Count Rate Per Unit Fluence Rate

The full-energy peak count rate per unit fluence rate, which is a measure of the detectors response to a nearly plane-parallel beam of photons normal to the detector's downward facing face, was estimated by dividing the peak count rate measured when the source was directly below the detector at 0° by the fluence rate for each source determined using Equation 5,

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$$\phi = C(E) \cdot p(E) \cdot A \cdot (4\pi r^2)^{-1}. \qquad (5)$$

The energy-dependent factor, C(E), accounted for attenuation of photons between the source and detector. Photon yield per disintegration (s^{-1} Bq^{-1}) is described by p(E). The source, with activity A (Bq), was located a distance, r (m), from the detector's surface for photons with energies less than 100 keV and at a location within the detector determined by the photon cross section area of germanium for photons with energies between 100 keV and 1 MeV (ICRU 53 1994) as was the case for ^{137}Cs.

Fluence Rate Per Unit Activity Per Unit Area

The fluence rate of primary photons one meter above the ground per unit activity distributed in soil per unit area was estimated from values tabulated in ICRU 53 (1994) for relaxation masses that ranged from 0.0 (thin sheet) to 100 g cm^{-2} (approaching a uniform distribution). Relaxation masses for each location were estimated by dividing the soil density (g cm^{-3}), which includes effects from soil moisture, rocks, etc. by the mean rate at which soil concentrations decreased with depth (cm^{-1}). Hulse et al. (1999) found that while there are significant differences in soil densities between macroplots, rates at which soil concentrations of ^{137}Cs and ^{241}Am decreased with depth were relatively constant around Rocky Flats. Hulse et al. reported that the mean rates at which soil concentrations of ^{137}Cs decreased with depth were -0.25 ± 0.03 cm^{-1} for 18 on-site locations and -0.25 ± 0.06 cm^{-1} for 24 off-site locations. Hulse et al. (1999) also reported that the mean rate at which soil concentrations of ^{241}Am decreased with depth was -0.22 ± 0.04 cm^{-1} at four on-site locations where depth profiles were well developed.

Calibration Factors and In-situ Soil Deposition Estimates

The angular correction factor for 137Cs (661.7 keV) soil deposition estimates from measurements with the CSU detector, 0.95 □ı̃ for ^{137}Cs (661.7 keV), was determined using a 127 MBq ^{137}Cs source, manufactured by the ICN Chemical & Radioisotope Division of International Chemical & Nuclear Corporation[25]. The angular correction factor for ^{241}Am (59.5 keV) soil deposition estimates was determined to be 0.82 □ı̃ using a 439 kBq ^{241}Am source manufactured by Amersham Buchler GmbH & Co KG[26]. Both sources are traceable to standards at the National Institute of Standards Technology (NIST.

The same sources were used to determine the full-energy peak count rate per unit fluence rate for soil deposition estimates of 137Cs and 241Am using the CSU detector. The full-energy peak count rate for ^{137}Cs was estimated to be 0.093 ± 6.1cpm γ^{-1} m^2 s. The full-energy peak count rate for ^{241}Am was estimated to be 0.070 ± 5.1cpm γ^{-1} m^2 s.

These experimentally determined calibration factors for the CSU detector's response to low energy photons from ^{241}Am (59.5 keV) were compared with factors tabulated by Helfer and Miller (1988), which were generally for detectors with lower efficiencies than the CSU detector. It was therefore necessary to make projections beyond established data points, which is admittedly risky; however, acceptable in this instance since the objective was to validate order of magnitude of experimentally determined factors. The experimentally determined angular correction factor for ^{241}Am was approximately 10% lower than that estimated from published data. After accounting for

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the photon yield from, radioactive decay of ^{241}Am the full energy peak count rate per unit fluence rate was approximately 20% lower than that estimated from published data.

The fluence rate per unit activity per unit area is independent of detector characteristics. Averages have been estimated to enable comparisons; however, the fluence rate per unit activity per unit area was estimated separately for each measurement location using site-specific soil densities to determine relaxation masses that were used to estimate soil deposition levels. Soil densities for the 0 to 3 cm and 3 to 6 cm depth intervals at each measurement location were averaged when estimating the relaxation mass. The average fluence rate per unit activity per unit area for ^{137}Cs was estimated to be (check units) 510 γ s^{-1} kBq^{-1} using a relaxation mass of 5.6 g cm^{-3} (inverse relaxation length = -0.25 cm^{-1}, average soil density = 1.4 g cm^{-3}) photon yield per decay = ??. The average fluence rate per unit activity per unit area for ^{241}Am was estimated to be (check units) 84 γ s^{-1} kBq^{-1} using a relaxation mass of 6.36 g cm^{-2} (inverse relaxation length = -0.22 cm^{-1}, average soil density = 1.4 g cm^{-3}) photon yield per decay = 0.357.

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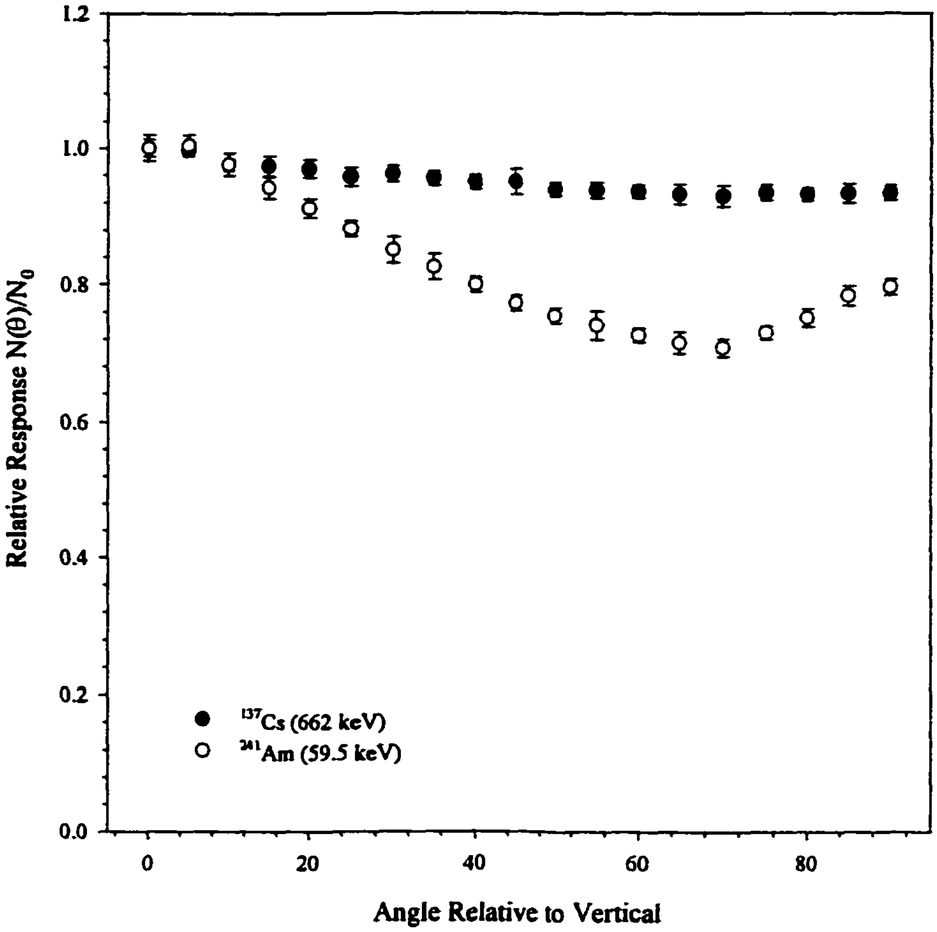
1.2
1.0
0.8
0.6
0.4
0.2
0.0
Relative Response N(θ)/N₀
0
20
40
60
80
Angle Relative to Vertical
● ¹³⁷Cs (662 keV)
○ ²⁴¹Am (59.5 keV)