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RADIONUCLIDES IN SOILS COLLECTED FROM WITHIN AND AROUND LOS ALAMOS NATIONAL LABORATORY: 1974–1996

Key Words: U, ^{241}Am, ^{137}Cs, ^{238}Pu, ^{239,240}Pu, ^{90}Sr, ^{3}H, contamination, exposure

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ABSTRACT

A soil sampling program is the most direct means of determining the types, concentration/activity, and distribution of radionuclides within and around nuclear facilities. Los Alamos National Laboratory (LANL), for example, has had a soil surveillance program since the early 1970s and the purpose of this paper was to 1) evaluate this 20+ year data set to determine if there are any statistical differences in radionuclides (^{3}H, ^{137}Cs, ^{238}Pu, ^{239,240}Pu, ^{241}Am, ^{90}Sr, ^{tot}U) and radioactivity (gross α, β, and γ), as a function of air emissions and fugitive dust, in surface soils (0–5 cm depth) collected from LANL, perimeter (PM) and background (BG) sites, and 2) determine if radionuclide concentrations are increasing or decreasing over time. Also, the total effective dose equivalent (TEDE) and the corresponding risk of excess cancer fatalities (RECF) to a PM

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community were estimated. Based on the long-term average, nine out of the ten radionuclide parameters measured in LANL soils (n=12) were significantly ($p<0.05$) higher in concentration than BG (n=6). Perimeter soils (n=10), on the other hand, showed less differences with only four out of the ten parameters being statistically higher in concentration than BG. Most radionuclides in LANL and PM areas, with the exception of ^{238}Pu in soils from PM, significantly decrease in concentration over time, so that by 1996 most radionuclides were approaching values similar to BG. The maximum net positive TEDE (i.e., the TEDE + two sigma for each radioisotope minus background and then only the positive doses summed) for a resident living around the PM of LANL, as modeled by the residual radioactive (RESRAD) code using a residential scenario for soils collected from 1974–1996, 1993–1996, and in 1996 was 2.9 mrem y^{-1} (29 μSv y^{-1}), 2.3 mrem y^{-1} (23 μSv y^{-1}), and 0.8 mrem y^{-1} (8 μSv y^{-1}), respectively. All upper bound TEDEs were far below the International Commission of Radiological Protection permissible dose limit of 100 mrem y^{-1} (1000 μSv y^{-1}) for all pathways, and the highest TEDE corresponds to a RECF of 1.5×10^{-6}—an estimate far below the Environmental Protection Agency guideline of 10^{-4}.

INTRODUCTION

A soil sampling and analysis program is the most direct means for determining the types, concentration/activity, and distribution of radionuclides in the environment within and around nuclear facilities (Mclendon, 1975; Nyhan et al., 1976; Litaor et al., 1994; Gallegos, 1995). Soil provides an integrating medium (reservoir) that can account for contaminants released to the atmosphere, either directly from gaseous effluents (air stack emissions) or indirectly from the resuspension of on-site contamination (fugitive dust) (Healy, 1977). Subsequently, the knowledge gained from a soil radiological surveillance program is critical for providing information about potential pathways (e.g., soil ingestion, food crops, resuspension into the air, and contamination of groundwater) that may result in a radiation dose to humans (Gilbert et al., 1988; Hakonson et al., 1981, Lee et al., 1985, Bunzl et al., 1994).

Soil surface samples have been collected from relatively undisturbed sites within and around Los Alamos National Laboratory (LANL), a nuclear weapons research, development, and testing facility, on an annual basis since the early 1970s and analyzed for a host of long-lived radiological constituents in an effort to determine the impact of Laboratory operations—as a function of air stack emissions and fugitive dust contamination—on the surrounding environment (Purtymun et al., 1980; Purtymun et al., 1987). The objectives of this study, therefore, were to (1) evaluate this 20-plus-year data set to determine if there were any statistical differences in the concentration of various radionuclides and radioactivity in soils collected from LANL, its perimeter (PM), and regional background (BG) areas over various time periods, and (2) determine if radioisotope concentrations are increasing or decreasing over time. Also, the total effective dose equivalent (TEDE) and risk of excess cancer fatalities (RECF) to a

resident(s) living around the PM of the Laboratory at the 95% confidence level were estimated.

MATERIALS AND METHODS

LANL is located in north-central New Mexico approximately 60 mi (97 km) north of Albuquerque (Figure 1). The Laboratory site, which ranges in elevation from 7800 ft (2371 m) on the west to 6200 ft (1885 m) on the east, encompasses about 43 mi^2 (111 km^2) and is divided into approximately 50 technical areas that are used for building sites, experimental areas, and waste disposal locations. Average precipitation around the LANL area is about 19 in. (483 mm) per year, and the major wind direction is southwesterly to northeasterly (Bowen, 1990).

Soil surface samples were collected from relatively level, open, and undisturbed areas from 12 LANL, 10 PM, and 6 regional BG locations. Again, sites sampled at LANL were not from areas where solid and/or liquid wastes have been released (e.g., firing sites, waste disposal sites, outfalls, etc.). Instead, the majority of LANL soil sampling sites were located downwind from major facilities and/or operations at LANL in an effort to assess radionuclides and radioactivity in soils that may be a result of air stack emissions (e.g., there were approximately 130 stacks at LANL in 1995 that could emit radionuclides) (ESP, 1996) and fugitive dust (e.g., the resuspension of dust from contaminated areas—firing sites, waste disposal sites, outfalls, etc.). Similarily, most PM stations were located on the downwind side of LANL (four sites mostly located on the

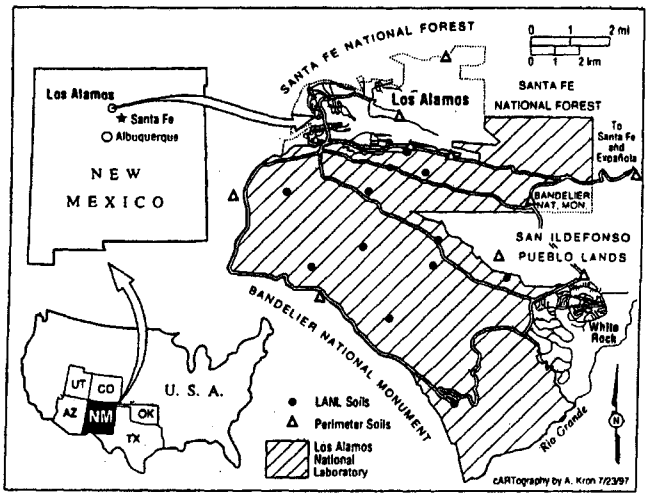

FIG. 1—Locations of Soil Sampling on LANL and Perimeter Areas

north/northeast side and four sites located on the east side) and one each on the west (U. S. Forest Service) and south (U. S. Park Service—Bandelier National Monument) side of the Laboratory to provide comprehensive coverage. All BG sites ranged from 20 mi (32 km) to 60 mi (96 km) away from the Laboratory on all sides and were beyond the likely range of significant impacts from LANL operations; and thus, radionuclides in soils from these sites were mostly a result of worldwide fallout and/or to naturally occurring radioactive materials in the earths crust.

At each site, soil surface samples were collected from the center and corners of a square area 33 ft (10 m) per side using a stainless steel soil ring 4 in. (101 mm) in diameter pushed 2 in. (51 mm) deep (ASTM 1990). The five subsamples were combined and mixed thoroughly in a large Ziploc bag to form a composite sample. All soil samples were submitted to an environmental chemistry group at LANL for the analysis of tritium (^{3}H), cesium (^{137}Cs), plutonium (^{238}Pu and $^{239,240}Pu$), strontium (^{90}Sr), americium (^{241}Am), and total uranium (^{tot}U) and gross alpha (α), beta (β), and gamma (γ) activity. These elements were selected on the basis of their history of use at the Laboratory, activity, and decay mode (half-life) (ESP, 1996). All methods of analysis have been previously reported (Purtymun et al., 1980; Purtymun et al., 1987); uranium, however, was analyzed by a fluorometric method from 1974 to 1976, by a delayed neutron activation method from 1977 to 1992, and by kinetic phosphorescence analysis from 1993 to 1996.

The long-term (1974 through 1996) and most recent (1993 through 1996 and 1996) average radionuclide concentrations at individual sites were estimated, and the concentrations at LANL, PM, and BG were compared to one another using a nonparametric Wilcoxon Rank Sum test at the 0.05 probability level (Gilbert, 1987). Also, a Mann-Kendall test at the 0.05 probability level was used to evaluate trends over the long term (1974 through 1996) (Gilbert, 1987). The general direction of trend was reported—a positive value for the statistic indicated an upward trend while a negative value for the statistic indicated a downward trend, even when the test result was not significant.

The TEDE (the effective dose equivalent from external radiation plus the committed effective dose equivalent from internal radiation), based on the average concentration of the seven soil radionuclides collected over the three assessment periods (1974–1996, 1993–1996, and 1996) from PM and BG sampling stations (Table 1), were calculated using version 5.61 of the residual radioactive (RESRAD) code (Yu et al., 1996) using a residential scenario (Fresquez et al., 1996). Soil ingestion (44 g^{-1}), inhalation of suspended dust (9×10^{-5} g^{-1} m^{3}), and ingestion of homegrown fruits, vegetables, and grains (a maximum ingestion rate of 352 lb. y^{-1} [160 kg y^{-1}] was employed) were considered to be the primary exposure pathways. Ingestion pathways for milk, meat and poultry, and fish/shellfish were not activated for this radiological assessment because these products are not raised in the principally urban Los Alamos area. Also, drinking and irrigation water pathways were not employed because the main aquifer is located beneath several hundred feet of volcanic tuff at a depth ranging from 1,150 to 1,200 ft (350 to 365 m) (ESP, 1996), and surface waters are intermittent and not used for irrigation. All LANL site-specific input parameters for the mesa-top environment around the Los Alamos area can be found in Fresquez et al. (1996). Finally, the risk of excess cancer fatalities (RECF) from whole-body radiation was determined by multiplying the dose (in mrem) by 5×10^{-7} excess cancer fatalities per person-mrem (NCRP, 1993).

RESULTS AND DISCUSSION

Based on the long-term average (1974 through 1996), most radionuclides

TABLE 1. Mean (+/- std dev) Radionuclide Concentration/Activity (dry weight basis) in Soils Collected from LANL, Perimeter, and Regional Background Locations from 1974–1996, 1993–1996, and 1996.

Year/Location	^{3}H pCi mL^{-1}	^{137}Cs pCi g^{-1}	^{238}Pu pCi g^{-1}	$^{239,240}Pu$ pCi g^{-1}	^{90}Sr pCi g^{-1}	^{238}U µg g^{-1}	^{241}Am pCi g^{-1}	Gross α pCi g^{-1}	Gross β pCi g^{-1}	Gross γ pCi g^{-1}
1974–1996										
LANL	7.32a[2] (10.04)	0.61a (0.32)	0.064ab (0.207)	0.069a (0.092)	0.52ab (0.27)	4.1a (0.9)	0.011ab (0.006)	9.5a (2.1)	9.6a (3.4)	4.3a (0.6)
Perimeter	2.11b (1.22)	0.59a (0.27)	0.003bc (0.003)	0.036a (0.046)	0.57bc (0.39)	3.7a (0.7)	0.011bc (0.009)	7.3b (1.3)	8.5a (2.7)	4.6a (1.1)
Background	1.31b (0.59)	0.42a (0.18)	0.002c (0.001)	0.009b (0.005)	0.29c (0.08)	2.4b (0.3)	0.006c (0.002)	7.6b (3.5)	6.2b (1.1)	2.9b (0.8)
1993–1996										
LANL	0.67a (0.83)	0.45a (0.28)	0.008ab (0.007)	0.077a (0.163)	0.42a (0.18)	3.0a (0.9)	0.009a (0.005)	6.5a (3.9)	6.6a (6.3)	3.5a (0.3)
Perimeter	0.24b (0.26)	0.38a (0.30)	0.007bc (0.011)	0.051a (0.081)	0.34a (0.18)	3.0a (0.7)	0.011a (0.013)	4.6a (2.0)	5.2a (1.5)	3.7a (0.4)
Background	-0.10c (0.23)	0.28a (0.13)	0.004c (0.002)	0.011b (0.004)	0.30a (0.07)	1.9b (0.4)	0.006a (0.001)	4.8a (1.2)	4.5a (0.7)	2.8b (0.4)
1996										
LANL	0.80ab (0.70)	0.39a (0.18)	0.002a (0.002)	0.027a (0.025)	0.30a (0.10)	3.3a (0.8)	0.008bc (0.005)	5.1a (1.5)	4.6a (1.5)	3.2a (0.7)
Perimeter	0.50bc (0.30)	0.37a (0.19)	0.002a (0.001)	0.046a (0.081)	0.30a (0.20)	3.4a (0.9)	0.012ab (0.013)	5.5a (2.6)	5.8a (1.5)	3.6a (0.6)
Background	0.40c (0.10)	0.27a (0.12)	0.002a (0.001)	0.010b (0.005)	0.20a (0.25)	2.3b (0.5)	0.006c (0.001)	4.4a (1.2)	5.0a (0.8)	2.7b (0.6)

[1]To convert pCi g^{-1} to Bq kg^{-1} multiply by 37.
[2]Means within the same column and year followed by the same letter are not significantly different at the 0.05 level using a nonparametric Wilcoxon Rank Sum Test.

and radioactivity, with the exception of ^{137}Cs, in LANL soils were significantly ($p<0.05$) higher in concentration than radionuclides and radioactivity in soils collected from regional BG locations (Table 1). Radionuclides in soils from BG areas compare well with radionuclide concentrations from other "background" locations in central Colorado (Hodge et al., 1996). Concentrations of ^{3}H, ^{238}Pu, $^{239,240}Pu$, and ^{60}U in soils collected from LANL areas were also generally higher than in soils collected from PM locations; these particular isotopes, then, were probably due to Laboratory operations.

Radionuclides and radioactivity in soils collected from PM soils showed less differences in concentrations than the LANL sites as only four out of the ten radionuclide parameters in PM soils were significantly ($p<0.05$) higher in concentration than in soils collected from BG in all three assessment periods. The higher radionuclide concentrations in soils collected from PM areas as compared to regional BG locations may be due, in part, to Laboratory operations, but probably more to worldwide fallout from nuclear weapons testing (Klement, 1965), the burn up of a satellite power source in the atmosphere (Perkins and Thomas, 1980), and reactor accidents (e.g., Chernobyl) (Andersson and Roed, 1994); and, in the case of uranium, to naturally occurring minerals in the earth's crust (Schulz, 1965). Radionuclides due to fallout vary from one area to another depending on wind patterns, elevation, and precipitation (Whicker and Schulz, 1982). Usually, higher amounts of radionuclides from fallout occur at higher elevations that receive greater amounts of precipitation than in areas at lower

Table 2. Results of the Mann-Kendall Nonparametric Test for Trend for Radionuclides in Soils Collected from LANL, PM, and BG from 1974–1996.

Study Site	^{3}H	^{137}Cs	^{238}Pu	^{239}Pu	^{90}Sr	^{234}U	^{241}Am	Gross α	Gross β	Gross γ
LANL	D**[1,2]	D*	D	D*	D	D**	U	D	D	D**
PM	D**	D**	U**	D	D*	D*	NT	D	D	D**
BG	D**	D**	U**	U**	D	D**	D	D	D*	D

[1]D=down, U=up, and NT=no trend.
[2]* and ** = significant at 0.05 and 0.01 probability level, respectively.

elevations that receive lower amounts of precipitation (Purtymun et al., 1990; Mitchell et al., 1990). Most of the regional BG areas in this study, for example, ranged from 5,600 ft (1,700 m) to 6,300 ft (1,920 m) above sea level and have an average rainfall of approximately 10 in. (25 cm) per year (USDOE 1976). By contrast, the PM areas were located above 7,200 ft (2,190 m) above sea level and receive about 19 in. (48 cm) of rainfall on average per year. The higher levels of uranium detected in the soils collected from the PM areas as compared with BG areas, on the other hand, may be a result of differences in the geology or mineralogy of the soils between the two sites. Soils in the LANL/PM area are derived from Bandelier (volcanic) tuff and have higher-than-average natural uranium contents, ranging from 3 to 11 $\mu g\ g^{-1}$ (Crowe et al., 1978; Longmire et al., 1995).

Although radionuclide and radioactivity levels were generally higher in PM and especially in LANL soils as compared with BG, most radionuclides, with the exception of ^{238}Pu in soils from PM areas, exhibited decreasing concentration

trends over time (Table 2). The statistically significant (but very small) increase of ^{238}Pu in PM soils over time may be related to the resuspension and redistribution of global fallout (Anspaugh et al., 1975) and/or to past LANL operations (Gallaher et al., 1997). ^{238}Pu and $^{239,240}Pu$ in soils from BG areas also exhibited statistically increasing trends over time, but in this case, the small increase in Pu levels in soils from BG areas over time were probably a reflection of the redistribution of fallout; the Pu levels in BG soils (Table 1), for example, were still well within worldwide fallout concentrations (Hodge et al., 1996). The decreasing concentrations of the other isotopes in soils collected from LANL and PM areas over time, on the other hand, may be a result of the cessation of above-ground nuclear weapons testing in the early 1960s (Klement, 1965), weathering (e.g., erosion, leaching, and wind) (Rogowski and Tamura, 1965), radioactive decay (half-life) (Whicker, 1982), and reductions in operations and/or better engineering controls employed by LANL (Fuehne, 1996). Tritium, in particular, which has a half-life of about 12 years exhibited the greatest decrease in activity over the 20-plus-year period of this study at all three sites, including BG. Indeed, by 1993–1996 and 1996, the majority of radionuclides and radioactivity in soils collected from both LANL and PM areas were statistically similar to values detected in regional BG locations (Table 1).

The maximum TEDE (the TEDE + two sigma of each radioisotope) and the maximum net positive (MNP) TEDE (the TEDE + two sigma of each radioisotope minus background and then only the positive doses summed) to a

Table 3. The Maximum TEDE, the MNP TEDE, and the RECF to a Member of the Public as estimated by RESRAD.

Year/Location	Maximum TEDE (mrem)[1]	MNP TEDE (mrem)	RECF MNP[2]
1974–1996			
Perimeter	8.7	2.9	1.5×10^{-6}
Reg. Background	5.9		
1993–1996			
Perimeter	5.5	2.3	1.2×10^{-6}
Reg. Background	3.2		
1996			
Perimeter	5.0	0.8	4.0×10^{-7}
Reg. Background	4.2		

[1]To convert mrem to μSv multiply by 10.
[2]MNP TEDE multiplied by 5×10^{-7} fatal cancers per mrem.

resident living around the PM of LANL may be found in Table 3. Based on the 1974–1996, 1993–1996, and 1996 assessment periods, the MNP TEDE—the dose that could potentially be attributed to Laboratory operations—was estimated at 2.9 mrem y^{-1} (29 μSv y^{-1}), 2.3 mrem y^{-1} (23 μSv y^{-1}), and 0.8 mrem y^{-1} (8 μSv y^{-1}), respectively. All upper bound doses were far below the International Commission on Radiological Protection (ICRP) permissible dose limit of 100 mrem y^{-1} (1000 μSv y^{-1}) set by the ICRP (1978) above that received from other man-made and/or natural sources in the Los Alamos area (e.g., 300 to 350 mrem y^{-1} [3000 to 3500 μS y^{-1}]) (ESP, 1996). The radionuclides that contributed the most to these doses (98%) measured at PM sites included ^{90}Sr, ^{137}Cs, and ^{60}U. All of these isotopes are by-products of fallout as well as radionuclides known to be released by the

Laboratory; uranium, however, is also from natural sources (Schulz, 1965). Finally, the RECF, based on the highest TEDE of 2.9 mrem y^{-1} equaled 1.5×10^{-6} (1.5 in a million) and was far below the Environmental Protection Agency upper bound guideline of 10^{-4} (100 in million) (USEPA, 1994).

CONCLUSIONS

The higher concentrations of radionuclides and radioactivity in soils collected from LANL sites as compared to BG sites were attributed mostly to Laboratory operations, especially during the early years, whereas the higher concentrations in soils collected from PM areas as compared to BG were attributed mostly to fallout and to naturally occurring sources. Although radionuclide concentrations and radioactivity in soils from PM and especially from LANL areas were higher than in soils collected from BG locations, most concentrations significantly decreased over time, so that by 1996 most radioisotopes/radioactivity were similar and/or approaching those values observed in soils from BG locations. Overall, based on the soil radionuclide source terms during 1974–1996, 1993–1996, and 1996, the corresponding dose and risk, at the 95% confidence level, indicate that there were no significant radionuclide contamination impacts from Laboratory operations, as a function of air emissions and fugitive dust, to the surrounding community.

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EMISSIONS OF VOLATILE ORGANIC COMPOUNDS FROM LARGE-SCALE INCINERATION PLANTS

Key Words: Emission, incineration, volatile organic compound (VOC).

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ABSTRACT

This paper gives results of the measurements and evaluation of emissions from seven Swedish incineration plants. The investigated incinerators ranged from 12 to 80 MW, and include Martin grate, Von Roll grate, Overthrust (W+E) grate, Vereinige Kesselwerke (V+K) grate, travelling grate, vibration grate and circulating fluidized bed (CFB) types. The analytical techniques used include on-line carbon monoxide (CO), nitrogen oxides (NO_x), sulfur dioxide (SO_2),