Radionuclide Activity Concentrations in Conifer Trees at the Los Alamos National Laboratory

Internal Report

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Abstract

At a nuclear weapons complex we measured radionuclide concentrations in bark, pulp, and combined bark and pulp samples of conifer trees (*Pinus edulis* and *Pinus ponderosa*) to help make decisions on the disposition and use of large volumes of trees thinned as part of a wildfire management program. Ash samples were analyzed for one or more radioisotopes—^{137}Cs, ^{238}Pu, ^{239,240}Pu, ^{90}Sr, ^{241}Am, ^{234}U, ^{235}U, and/or ^{238}U—and ^{235}U/^{238}U ratios on a mass basis were evaluated to indicate the degree of intermixing of anthropogenic versus natural sources of uranium. Study areas at the Los Alamos National Laboratory (LANL) included sites where munitions have been routinely tested involving the use of depleted uranium-hardened targets over a period of approximately four decades. Most trees sampled at LANL contained depleted uranium and intermixing of natural uranium as well. At the firing sites, most of the individual samples had U isotope concentrations that were above the Regional Statistical Reference Level (RSRL) but comparisons of groups of data (i.e., means) resulted in statistically similar levels. Area G samples generally had statistically higher concentrations of ^{3}H, ^{234}U, ^{235}U, and ^{238}U, but similar levels of ^{241}Am, ^{137}Cs, ^{238}Pu, and ^{239,240}Pu. At DX sites, only ^{238}U levels in individual samples consistently exceeded the RSRL, but data group comparisons generally yielded non-significant differences. Additional information is needed on the distribution of U within particular groupings of tree rings (pre-munitions-testing era vs. post-munitions-testing era) and the analysis of ^{236}U, which is only found in anthropogenically- produced uranium.

Introduction

After the Cerro Grande Fire of 2000, conifer trees in Mortandad Canyon within the Los Alamos National Laboratory (LANL) were felled as a hazard reduction effort. The concentrations measured in those trees indicated a need to develop a procedure for the disposition of potentially contaminated trees felled during a subsequent extensive thinning program of conifer forests as a component of a Wildfire Hazard Reduction Program. This created large volumes of conifer wood for which several potential uses and disposal dispositions were identified. As a result, we developed a procedure for the identification, removal, and disposition of potentially contaminated trees and other vegetation from LANL (LANL 2001). An option in the *Procedure* that was sometimes exercised was to sample and analyze pulp and/or bark for radionuclide content. Specifically, the perception of risk to the health of citizens who considered using wood from LANL created the need to know whether radionuclide concentrations in thinned trees were elevated above levels in conifers from regional background areas, which are assumed to be outside of the influence of LANL. There were also cultural issues identified by neighboring Native Americans that related to the source (natural vs. anthropogenic) of uranium (U) isotopes in conifer trees.

We report the results of Lab-wide sampling and analysis of conifer trees for radioactivity concentrations and compare to levels in conifers from regional background areas. Decisions on the release of wood considered, among other factors, the results of our sampling. The data are also important to other environmental protection programs at LANL.

Background

In 2001 and 2002 samples of *Pinus edulis* (piñon pine) and *Pinus ponderosa* (ponderosa pine) were collected from areas of LANL that were planned for or in the process of being

thinned. Some of the areas were known to be relatively free of contiguous radionuclide contamination and others were suspect as based on radionuclide concentrations in sediment or soil. Developed shortly after the Cerro Grande Fire, the procedure for the identification, removal, and disposition of potentially contaminated trees from LANL disallowed the use of wood or other material from trees that were from areas known to have contamination; e.g. characterized "potential release sites (PRSs)", solid waste management units such as the Area G low-level radioactive waste material disposal area (MDA), or other areas of concern such as the modern floodplain area in Mortandad Canyon. Since conservative dose estimates from burning large volumes of contaminated wood had previously shown a negligible contribution compared with the annual radiation dose from natural factors and no health effects from the inhalation of radionuclides were expected (Gonzales and Bare 2001), the stimulus for the sampling described in this report was primarily perception of risk by the public, not actual health risk. The criteria used to gauge perception was whether or not levels of radionuclides in pulp and bark from onsite (LANL) trees were detectable and elevated statistically above background levels in trees from regional offsite areas (Areas that were upwind and upslope of LANL in the Jemez Mountains or that were in a different mountain range—the Sangre de Cristo Mountains—roughly 50 miles from LANL or in a direction [southeast] away from the predominant wind direction [northeast]. A three-step process (discussed in Methods) was used to determine whether a sample or group of samples should be considered "contaminated."

Members of San Ildefonso Pueblo, which shares a boundary with LANL, expressed their desire to avoid the use of natural resources containing any substance, such as isotopes of U that originated from Laboratory activities. One of the potential uses by the Pueblo was firewood to heat kilns used in curing pottery. Therefore, in addition to determining relative levels of U in

comparison to levels offsite, it was important to determine whether trees contained depleted uranium (DU) or enriched uranium, which would indicate anthropogenic sources of U. The depleted and enriched forms of uranium result from the processing of natural uranium to selectively increase or decrease the abundance of ^{235}U relative to ^{238}U (Gallaher et al. 1997). Enriched uranium is processed uranium containing more than 0.72 percent ^{235}U; DU contains less than 0.72 percent ^{235}U. The abundance of ^{235}U in highly enriched uranium may be greater than 90 percent, while the abundance of ^{235}U in highly depleted uranium may be about 0.25 percent (Gallaher et al. 1997). Ratios of $^{235}U/^{238}U$ on a mass basis were used to indicate whether trees were contaminated, at least in part, with DU. Sansone et al. (2001) recently applied this technique relating DU in tree bark to DU ammunition used during the 1999 war in Kosovo, and Edmands et al. (2001) recently applied the technique relating DU in *Quercus velutina* (black oak) to a longstanding U processing plant. In the processing plant study they found that most trees had DU, indicating its ability to be assimilated by plant roots. Researchers in both studies found that most trees had DU, indicating mobility of U in biological systems.

Methods

The procedures for determining an approach for sampling conifer tree materials are documented in an internal report (LANL 2001). Generally, we systematically divided (gridded) areas of the Laboratory planned for thinning into units and made mathematical estimates of optimal sampling sizes using one-tailed tests for binomial data and previous variance data on U in pine needles. We conducted cluster random sampling. Optimal sample sizes based on prior shoot tip data were lower than sample sizes based on prior wood data. Areas of relatively clean, low-contaminated, and moderately contaminated levels of radioactivity across LANL were

identified on the basis of historical soil sampling results and infrequent sampling of vegetation. Our knowledge of levels of radioactivity in vegetation consisted of sample results for overstory and understory from environmental surveillance sampling locations in 1998 and sampling of conifer pulp and bark from ponderosa pine trees in Mortandad Canyon in 2000. Figure 1 shows the areas of the Laboratory sampled and Table 1 identifies the analytes targeted for sampling at each area. This report addresses only radionuclides.

Sampling for radionuclide analysis consisted of cross-sectioning between three and five trees (subsamples) at breast height, sometimes partitioning bark from pulp, and then processing for radionuclide analysis. Tree selection was based on the lifetime of the Laboratory such that the trees were generally ≥60 years old. The three to five (depending on size of sample area) trees were treated as subsamples and were composited into one sample. Processing generally consisted of cubing samples in the laboratory and ashing approximately 1000 g of starting material through a 10-day step-wise temperature increase to 500 ^{o}C maximum. Fresh weights, stable dry weights, and stable ash weights were achieved.

The analytes targeted at each area varied depending on the results of historical soils sampling and sampling results for other media. Ash samples were analyzed for one or more radioisotopes—^{137}Cs, ^{238}Pu, ^{239,240}Pu, ^{90}Sr, ^{241}Am, ^{234}U, ^{235}U, and/or ^{238}U. Total U was sometimes also analyzed or calculated from the isotopic data. Strontium-90 analysis was by liquid scintillation; ^{238}Pu, ^{239}Pu, ^{234}U, ^{235}U, ^{238}U, and ^{241}Am were analyzed by alpha spectrometry; and total U was analyzed by kinetic phosphorescence analysis.

The following process from the *Procedure* was used to determine whether a sample or group of samples should be considered “contaminated,” or of perceived risk. Result values were qualified as nondetects if the result was less than or equal to three times the total propagated

uncertainty (99% confidence level). The Regional Statistical Reference Level (RSRL) for each radionuclide was computed as the background mean plus two standard deviations. The RSRL was used to evaluate the significance of radionuclide detections in onsite samples in comparison to the upper-level (95% confidence level) RSRL. Parametric statistical tests were used to assess differences in groups of radionuclide concentrations (represented by the mean) between onsite LANL samples versus regional background samples at the 0.05 probability level. Previous analyses of similar data have shown no differences in statistical outcome between parametric and nonparametric tests (Gonzales et al. 2000).

Results

Analytical results are summarized in Table 2 and detailed results are in Table 3. A scatter plot comparing ^{238}U results at all sites sampled is shown in Figure 2. Table 4 has uranium isotope masses (µg/g), mass ratios, and the tendency toward enriched, depleted, or natural uranium.

Firing Sites. The first area sampled was within the hazard zone of five of the Laboratory's oldest sites used for the open-air testing of explosives. Piñon tree bark from five firing sites operated by the Laboratory's Dynamic Experimentation (DX) Division were sampled–Ennie, Lower Slobovia, Minnie, Mo, and R306–and compared to three reference (background) samples. Each sample, one at each site, consisted of bark from five piñon trees (subsamples) roughly 300 to 400 ft from the shot centers. Samples were taken from the side of the tree facing the blast.

All three U isotopes (^{234}U, ^{235}U, ^{238}U) were detected in most of the onsite samples. Natural U is known to be elevated in soils within and around LANL. Uranium-234 and ^{238}U

were also detected in the reference samples and ^{235}U was not. Most of the individual samples at Ennie, Minnie, Mo, and R306 were "contaminated" as the U isotope concentrations exceeded the RSRL. Lower Slobovia individual samples were not "contaminated." It was of questionable value to group the data from the five different firing sites because of the variation in materials used from site to site. At the 95% confidence level, the onsite U-isotope values were not statistically different from the offsite values for all three isotopes, partially due to low sample sizes. The three highest levels of ^{238}U–5.68, 6.5, and 17.1 pCi/g-ash–compared with background bark samples that had a range of 0.17 to 0.42 pCi/g ash. Ponderosa pine bark samples previously taken from Mortandad Canyon had a mean concentration of 1.7 pCi ^{238}U/g-ash (Gonzales and Bare 2001), and the reference sample for that study had 0.38 pCi/g-ash of ^{238}U. Comparing the ^{235}U to ^{238}U mass, the ratios implied that the U type in the piñon bark at Ennie, Lower Slobovia, Mo, and R306 was predominantly "depleted" (Table 4), as would have been expected from sites where munitions testing has occurred since the 1960's.

DX West. Ten ponderosa pine samples (bark and pulp combined) were collected at DX West of Planning Area (PA) 3 and were compared to three offsite reference samples. As shown in Figure 1, the 10 onsite samples were taken in the northwest corner of PA 3 most likely away from the direct influence of any PRSs. The nearest PRSs were C-08-004 and -019 and the TA-08-002 inactive firing site (south and southwest of most of the samples) and 09-013. Each sample consisted of four subsamples, one from each corner of a grid square, that were composited. Ashed samples were analyzed for ^{137}Cs; ^{238}Pu and ^{239}Pu; ^{90}Sr; and ^{234}U, ^{235}U, and ^{238}U.

Most individual samples for most of the radionuclides did not exceed the RSRL. Samples exceeding the RSRL were as follows: 1/10 onsite samples for ^{137}Cs, 2/10 for ^{239}Pu, 9/10

for ^{238}U, and 1/10 for ^{238}Pu. Figure 2 shows the results for ^{238}U from this area as well as the background values and concentrations from other areas sampled during this study. No onsite results for ^{90}Sr and ^{234}U, nor ^{235}U exceeded the RSRL. When comparing the group of 10 onsite results to the group of three offsite results using student t-tests, there were no statistical differences at the 95% confidence level. The tendency was a predominance of DU with some evidence of natural uranium (Table 4).

DX North. Eight ponderosa pine samples collected at DX North, (shown in Fig. 1) were compared to the same three offsite reference samples as the DX West samples. Each sample consisted of four subsamples that were composited. Ashed samples were analyzed for ^{137}Cs; ^{238}Pu and ^{239}Pu; ^{90}Sr; and ^{234}U, ^{235}U, and ^{238}U. Samples exceeding the RSRL were as follows: 1/4 onsite samples for ^{137}Cs, 1/4 for ^{239}Pu, 2/4 for ^{234}U, 1/4 for ^{235}U, 3/4 for ^{238}U, and 2/4 for ^{238}Pu. No onsite results for ^{90}Sr exceeded the RSRL. The live tree ("6 Live," etc.) samples (as opposed to standing dead) generally had higher concentrations than sample IDs 21, 22, and 23. Comparing the group of eight onsite results to the offsite results using student t-tests, there were no statistical differences between means at the 95% confidence level. As opposed to the other DX sites, the mass ratio comparisons for the four samples yielded mixed results.

DX South. Eight ponderosa pine samples (bark and pulp combined) were taken at DX south (as shown) and were compared to the three offsite reference samples. Each sample consisted of four subsamples that were composited. Ashed samples were analyzed for ^{137}Cs; ^{238}Pu and ^{239}Pu; ^{90}Sr; and ^{234}U, ^{235}U, and ^{238}U. Most individual samples exceeded the RSRL. Samples exceeding the RSRL were as follows: 8/8 for ^{234}U; 8/8 for ^{238}U. None of the ^{137}Cs and ^{235}U samples exceeded RSRL. All eight samples typed as predominantly DU.

Area G. Nine piñon samples were collected around the "Area G" low-level radioactive waste MDA as shown in Figure 1. Five of the samples were collected from and elongated "shelf," "mesita," or "bench" in Cañada del Buey, north and east of the MDA. The other four samples were on the mesa at points south and east of Area G and east and north of Area J. The results were compared to five background samples taken from the Santa Fe National and Carson National Forests at points generally west and east of, but well removed from, LANL. The background sample locations (shown in Fig. 1) were around Saint Peters Dome and Bland Canyon in the Jemez Mountains; and Bland Canyon, Pajarito Ski Basin Road, and nearby Rancho Encantado Restaurant on Forest Road 102, all in the Sangre de Cristo Mountains. Each sample consisted of cross sections from three trees (subsamples). The analytes were ^{241}Am, ^{137}Cs, ^{3}H, ^{238}Pu, ^{239,240}Pu, ^{234}U, ^{235}U, and ^{238}U.

Figure 2 shows the distribution of detections in various sample groups. There was a 77% detection rate (56/72) in the onsite samples and 58% detection rate in the reference or background samples. Neither ^{137}Cs nor ^{3}H were detected in the pulp and bark samples east and north of Area J. Of the 56 detections total, 36 sample values (64%) exceeded the RSRL. None of the ^{137}Cs results exceeded the RSRL. Grouping all nine Area G-related samples and statistically comparing the means to the offsite sample mean, the onsite mean was statistically greater than the offsite mean for ^{3}H ($P = 0.014$), ^{234}U ($P = 0.0003$), ^{235}U ($P = 0.044$), and ^{238}U ($P = 0.0005$), showing the influence of an active disposal site. The Area G-related sample means were not statistically different than the offsite mean for ^{241}Am, ^{137}Cs, ^{238}Pu, and ^{239,240}Pu.

Discussion

Nearly 100 metric tons of DU have been released into the environment at LANL resulting in elevated concentrations in soil at isolated locations within LANL (Becker 1991). The mass ratio technique consistently indicated contamination of most conifer tree samples with DU. Intermixing of DU and natural U likely occurred in the onsite trees. When pulp and bark were separated and analyzed, the majority of measured ^{238}U was in pulp (not bark) indicating its movement into trees may be primarily the result of root uptake. Since ^{238}U is known to be a surrogate of nutrient S (Whicker and Shultz 1982), a macronutrient to plants, we believe that some of the ^{238}U measured in onsite samples is natural ^{238}U that was absorbed by the conifer tree roots. Plant/soil concentration ratios (uptake coefficients) have been estimated to range between 10^{-4} and values exceeding 10^{-1} (Whicker and Shultz 1982), depending on, among other things, plant species. Studies in other parts of the world have concluded that uptake of DU by plant roots does occur (Sansone et al. 2001, Edmands et al. 2001), and Edmands et al. (2001) have suggested that since the uptake of U isotopes is competitively depressed by Ca^{2+}, the uptake and translocation of certain U isotopes (e.g., UO_2^{2+}) may be similar to that of calcium. With a $^{235}U/^{238}U$ ratio of <0.0072 for DU and ~0.0072 for natural U, many of the onsite sample ratios indicated impure DU, likely intermixed with natural U. Studies on soils have shown that the immediate area in and around LANL has elevated levels of natural U as does the Pajarito Plateau compared with the broader regional area. A regional mean concentration of 6.5 ppm in soil (Nyhan et al. 2003) compares to 3–4 ppm in soil, a world-wide average concentration (Whicker and Shultz 1982, Sansone et al. 2001). Furthermore, studies elsewhere have concluded that U absorbed by tree roots and deposited in above-ground tree material translocates radially. Studying U in black oaks at a bog adjacent to a U processing plant, Edmands et al. (2001)

concluded that, because DU was detected in tree rings dating back to as many as 22 years before the introduction of DU to the area, after uptake DU moves radially within trees.

The comparison (Figs. 3 and 4) of activity concentrations to those measured in trees sampled at Mortandad Canyon shortly after the Cerro Grande Fire is important because one of the assumptions made when radiological doses were estimated for a burn scenario (Gonzales and Bare 2001) using the Mortandad data was that most of the maximum activity concentrations measured in the Mortandad samples were at the high end of the Lab-wide range of values that would be ultimately established by further sampling. This turned out to be true for the most part. As expected, the exception was for the U isotopes. Mortandad Canyon had the highest ^{137}Cs concentration and the seventh highest ^{238}U concentration in comparison to other sites across LANL. Depleted uranium is less radioactive than natural uranium.

The comparison of radionuclide concentrations in onsite vs. offsite samples in the munitions testing areas (DX sites and firing sites) was in general agreement with the same type of comparison that was made for radionuclide concentrations in samples of burnt ponderosa pine bark taken immediately after the Cerro Grande Fire in the same areas (Gonzales and Fresquez 2002).

In order to compare concentrations of U isotopes in trees as measured in this study to vegetation previously analyzed at LANL, we converted all reported values to activity concentrations per unit of oven-dry plant material. Our maximum concentration of total uranium–1.5[1] μg/g-dry in piñon tree bark at a firing site (R306)–compares to <u>maximum</u> concentrations of 320 μg total U per g standing dead vegetation (non-ashed), which was from a sample collected in 1975 at a location (E-F Site) where U concentrations in surface soils were 20 to 3,500 times higher than background levels (Miera et al. 1980); 0.65 μg/g-dry in the bark of

shrubs that were rooted in transuranic waste material (Wenzel et al. 1987); 0.073[2] μg/g-dry in understory vegetation collected at one of 12 LANL Environmental Surveillance Program onsite locations in 1998 (Gonzales et al. 2000); 0.066[3]μg/g-dry in overstory vegetation at one of the same 12 locations and same year; 0.05[3] μg/g-dry in pine needles from the TA-16 Weapons Engineering Tritium Facility in 1985 (Fresquez and Ennis 1995); and 0.72[3]μg/g-dry in overstory vegetation at Dual-Axis Radiographic Hydrodynamic Test facility in 2002 (Nyhan et al. 2003).

[1]Computed by converting radioisotopic data to U mass data and using ash/dry weight ratio of 0.029 for bark from Gonzales et al. (2001). [2]Computed using ash/dry weight ratio of 0.1 from Fresquez and Ferenbaugh (1999); [3]Computed using ash/dry weight ratio of 0.08 from Fresquez and Ferenbaugh (1999).

Conclusions

Trees from most areas sampled at LANL contained sufficient quantities of U and, occasionally ^{137}Cs, that thinned trees from the areas sampled either were not released or had limited use. Although ratios indicated that most of the trees in which U was measured have DU, a review of pertinent literature indicates that natural U likely exists in all trees sampled considering the elevated levels of natural uranium in Pajarito Plateau soils. Nevertheless, the presence of any amount of DU is associated with LANL and Native Americans have expressed their desire to not use natural resources that contain Laboratory-related substances. Additional information on the distribution of U within particular groupings of tree rings (pre-munitions-testing era vs. post-munitions-testing era) is needed to understand the dynamics of uptake and distribution and to further impact the wildfire management program including the disposition of thinned trees. Analysis of particular groupings of tree rings should include the analysis of ^{236}U, which is a uranium isotope that is formed through exposure of ^{235}U to a neutron source and does not exist in nature; thus its presence unambiguously indicates an anthropogenic component (Gallaher et al. 1997). Analyses of individual trees, rather than composites, will enable us to more clearly understand the relationship of anthropogenic sources of uranium to natural sources. The use of thermal ionization mass spectrometry would enable us to more accurately distinguish and quantify the contributions of the different sources of uranium in a single sample. This information will also complement the few studies that have been completed elsewhere relating to U released from other events around the world.

Literature Cited

Becker, N.M., "Influence of hydraulic and geomorphologic components of a semi-arid watershed on depleted uranium transport," Ph.D. Thesis, University of Wisconsin-Madison, Madison, WI, USA (1991).

Edmands, J.D., D.J. Brabander, and D.S. Coleman, "Uptake and mobility of uranium in black oaks: implications for biomonitoring depleted uranium-contamination groundwater," Chemosphere 44:789–795 (2001).

Fresquez, P.R. and M. Ennis, "Baseline radionuclide concentrations in soils and vegetation around the proposed Weapons Engineering Tritium Facility and the Weapons Subsystems Laboratory at TA-16," Los Alamos National Laboratory report LA-13028-MS (September 1995).

Fresquez, P. R., and J. K. Ferenbaugh, "Moisture conversion ratios for the foodstuffs and biota environmental surveillance programs at Los Alamos National Laboratory," Los Alamos National Laboratory report LA-UR-99-253 (1999).

Gallaher, B.M., D.W. Efurd, D.J. Rokop, T.M. Benjamin, and A.K. Stoker, "Survey of plutonium and uranium atom ratios and activity levels in Mortandad Canyon," Los Alamos National Laboratory report LA-13379-MS (1997).

Gonzales, G.J., P.R. Fresquez, M.A. Mullen, and L. Naranjo, Jr., "Radionuclide concentrations in vegetation at the Los Alamos National Laboratory," Los Alamos National Laboratory report LA-13704-PR (2000).

Gonzales, G.J., P.R. Fresquez, and C.M. Bare, "Contaminant concentrations in conifer tree bark and wood following the Cerro Grande Fire," Los Alamos National Laboratory report LA-UR-01-6157 (2001).

Gonzales, G.J. and P.R. Fresquez, "Contaminant concentrations in burned conifer tree bark collected within the Los Alamos National Laboratory," Los Alamos National Laboratory report LA-UR-02-705 (2002).

LANL (Los Alamos National Laboratory), "Identification, removal, and disposition of potentially contaminated trees and other vegetation from Los Alamos National Laboratory Technical Areas," Los Alamos National Laboratory report LA-UR-01-6855 (December 2001).

Miera, F.R., E.S. Gladney, W.C. Hanson, T.F. Gesell, and W.M. Lowder, "Mobility of elevated levels of uranium in the environment," Natural Radiation Environment III. Conference on Natural Radiation Environment III, 23 April 1978, Houston, TX. Department of Energy, Oak Ridge, TN; CONF-780422-Vol. 1 (1980).

Nyhan, J., P. Fresquez, and E. Taylor, "Concentrations of radionuclides and trace elements in soils and vegetation around the DARHT Facility during 2002," Los Alamos National Laboratory report LA-14056-PR (August 2003).

Sansone, U., P.R. Danesi, S. Barbizzi, M. Belli, M. Campbell, S. Gaudino, G. Jia, R. Ocone, A. Pati, S. Rosamilia, and L. Stellato, "Radioecological survey at selected sites hit by depleted uranium ammunitions during the 1999 Kosovo conflict," Sci. of the Total Environ., 281:23–35 (2001).

Wenzel, W.J., T.S. Foxx, A.F. Gallegos, G. Tierney, and J.C. Rodgers, “Cesium-137, plutonium-239/240, total uranium, and scandium in trees and shrubs growing in transuranic waste at Area B,” Los Alamos National Laboratory report LA-11126-MS, (1987).

Whicker, W. F. and V. Schultz, “Radioecology: Nuclear Energy and the Environment,” CRC Press, Inc., Boca Raton, FL (1982).

Table 1. Areas of conifer tree sampling and analytes.

	Tissue Type			Analytes													
	Bark	Wood	Bark & Wood Combined	Metals						Organics	Radionuclides						
Area				Be	Ba	Cu	Pb	Ni	Ag	PCBs	^{137}Cs	^{238}Pu	^{239}Pu	^{90}Sr	^{234}U	^{235}U	^{238}U
DX Firing Sites*	x			x	x		x								x	x	x
DARHT			x		x	x	x	x	x						x	x	x
ESA	x	x			x	x	x	x	x								
Area G*			x								x	x	x		x	x	x
DX West			x								x	x	x	x	x	x	x
DX South			x								x	x	x	x	x	x	x
DX North			x								x	x	x	x	x	x	x
Sandia			x							x							
Offsite Reference**	x	x															

*Trees sampled at Area G and the firing sites were piñon pine (*Pinus edulis*); all others were ponderosa pine (*Pinus ponderosa*).

**Reference samples were obtained for both piñon pine and ponderosa pine.

Table 2. Mean concentration (activity) (pCi/g-ash) of radionuclides in conifer tree samples collected in and around LANL.

Sample Type/ Location	^{137}Cs	^{238}Pu	^{239}Pu	^{90}Sr	^{234}U	^{235}U	^{238}U
Ponderosa							
DARHT					1.740	0.170	5.130
DX West	1.17	0.003	0.030	5.78	0.139	0.008	0.232
DX North	0.99	0.004	0.031	5.80	0.150	0.150	0.011
DX South	1.07	0.003	0.023	6.18	0.276	0.016	0.533
Offsite*	0.50	0.001	0.012	4.77	0.065	0.007	0.068
Piñon							
Area G	0.73	0.197	0.136		0.172	0.009	0.291
Offsite	0.71	0.003	0.117		0.073	0.005	0.077

*Includes reference sample collected as part of a study on radionuclides in ponderosa pine in Mortandad Canyon by Gonzales et al. (2001).

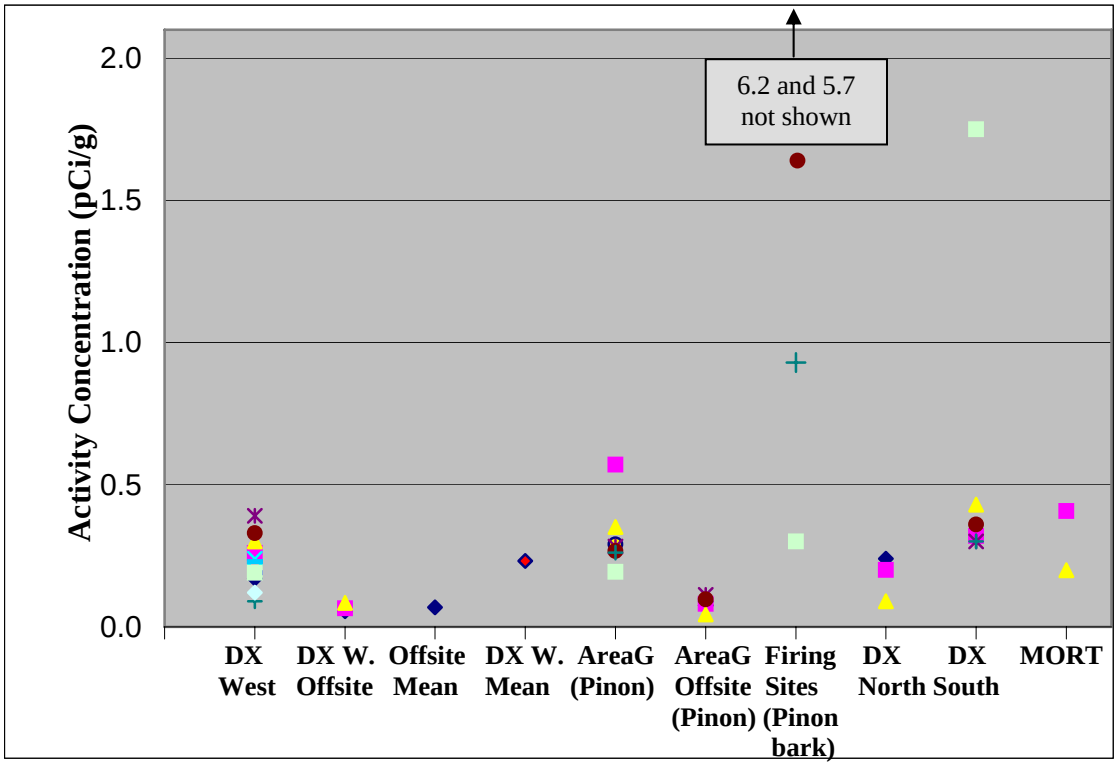

Figure 3. ^{238}U activity concentrations (activity) in ponderosa pine and piñon pulp from various locations within and around LANL.

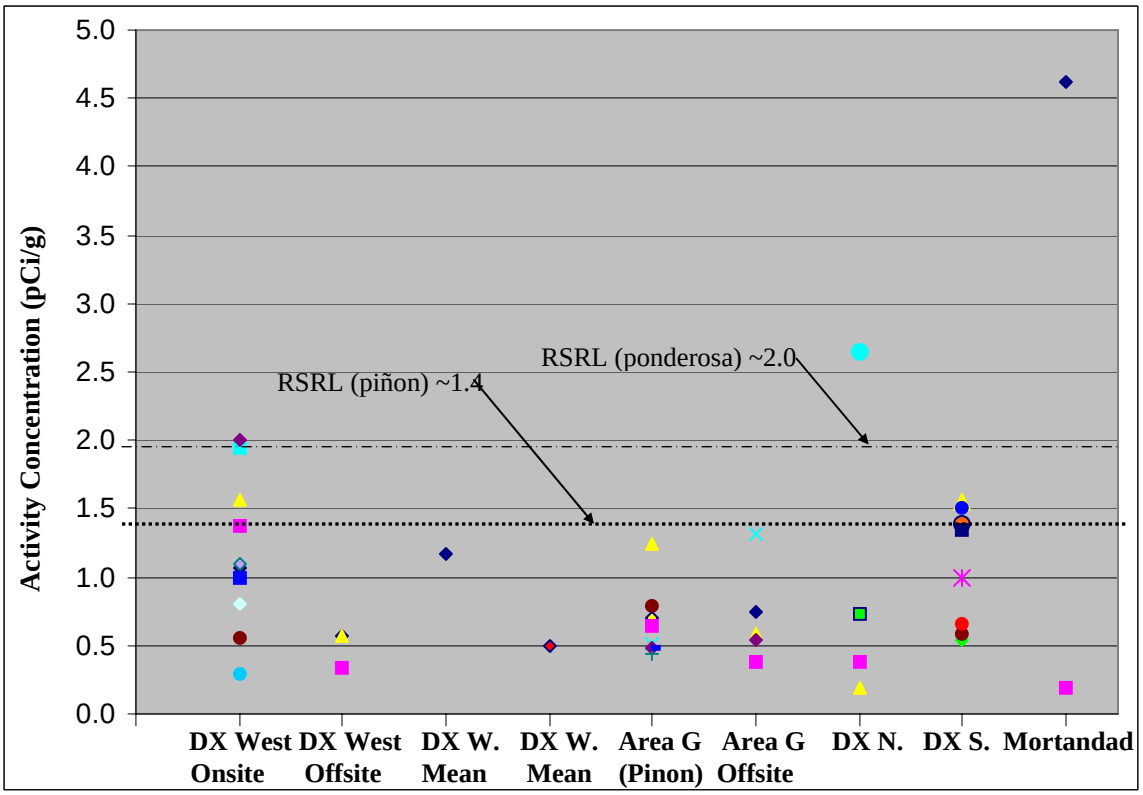

Figure 4. Plot of ^{137}Cs concentrations (activity) in ponderosa pine and piñon pulp wood and bark from various locations within and around LANL.

Table 3. Radionuclide activity concentrations (pCi/g-ash) in conifer trees at the Los Alamos National Laboratory.

		Analyte								
Area		^{241}Am	H^3	^{137}Cs	^{238}Pu	^{239}Pu	^{90}Sr	^{234}U	^{235}U	^{238}U
DX Firing Sites	7ENPB							1.16	0.215	5.68
	7LSPB							0.204	0.011	0.299
	7MNPB							0.49	0.076	1.64
	7MOPB							0.335	0.036	0.93
	7R306PB							6.5	0.51	17.1
DARHT	DX-DARHT-400							0.49	0.02	0.8
	DX-DARHT1-1200							1.9	0.12	3.9
	DX-DARHT2-1200							0.76	0.06	1.81
	DX-DARHT3-1200							0.39	0.04	1.26
	DX-DARHT4-1200							0.52	0.04	1.34
	DX-ENNIE1-250							0.48	0.06	3.27
	DX-ENNIE2-350							0.42	0.05	1.68
Area G	6 AreaJ-E	0.0203	0.15	0.7	0.0016	0.0261		0.139	0.0109	0.282
	7 AreaJ-N	0.0132	0.03	0.65	0.0088	0.0278		0.131	0.0073	0.261
	8 Soilloca #8	0.027	0.17	1.25	0.0084	0.05		0.238	0.0144	0.571
	9 TruPads-E	0.234	0.9	0.51	0.546	0.57		0.245	0.0156	0.351
	CdBueyS1	0.022	0.08	0.48	0.0177	0.034		0.159	0.0078	0.316
	CdBueyS2	0.015	0.4	0.79	0.005	0.03		0.106	0.0032	0.179
	CdBueyS3	0.043	0.41	0.44	0.0042	0.033		0.153	0.006	0.194
	CdBueyS4	0.057	0.9	0.48	0.049	0.105		0.169	0.0082	0.194
	CdBueyS5	0.18	1.64	1.28	1.13	0.344		0.207	0.0115	0.267
DX West	# 1 DX WEST			1.07	0.0027	0.02	3.97	0.13	0.0079	0.17
	# 2 DX WEST			1.56	0.0045	0.02	10	0.21	0.01	0.24
	# 3 DX WEST			1.94	0.004	0.03	7.5	0.05	0.01	0.39
	# 4 DX WEST			2.01	0.0033	0.02	6	0.12	0.01	0.33
	# 5 DX WEST			0.56	0.0008	0.03	5.5	0.15	0.0033	0.09
	# 7 DX WEST			1.1	0.0028	0.02	6	0.08	0.0033	0.19
	# 8 DX WEST			0.99	0.0023	0.03	4.59	0.16	0.008	0.23
	# 9 DX WEST			0.29	0.0036	0.02	5.9	0.13	0.0046	0.12
	# 11 DX WEST			0.8	0.0032	0.03	2.96	0.2	0.01	
	# 12 DX WEST			1.37	0.0042	0.03	5.33	0.2	0.0093	0.3
DX South	13			1.39	0.0035	0.03	5.30	0.21	0.01	0.40
	14			1.35	0.0027	0.02	8.20	0.20	0.01	0.32
	15			1.55	0.0039	0.04	6.20	0.24	0.01	0.43
	20			1.00	0.0026	0.02	5.00	0.23	0.01	0.41
	16-Live			0.54	0.0047	0.01	9.10	0.15	0.01	0.30
	17-Live			0.58	0.0032	0.01	4.62	0.18	0.01	0.36
	18-Live			0.66	0.0007	0.01	5.60	0.16	0.01	0.30
	19-Live			1.50	0.0027	0.04	5.40	0.84	0.06	1.75
DX North	21			0.73	0.0060	0.0200	4.28	0.06	0.0100	0.140
	22			0.38	0.0019	0.0100	5.70	0.13	0.0096	0.200
	23			0.19	0.0001	0.0036	5.60	0.08	0.0048	0.090
	6 Live			2.65	0.0089	0.0900	7.60	0.33	0.02	0.720

Table 4. Uranium isotope masses (µg/g), mass ratios, and source tendency. (Species were *Pinus ponderosa* unless otherwise noted.)

		Parameter			
Area	**Sample ID**	**^{235}U Mass**	**^{238}U Mass**	**^{235}U/^{238}U Ratio**	**U Source Tendency***
DX Firing Sites (*Pinus edulis)*	7ENPB	0.100467	17.005988	0.0059078	DU
	7LSPB	0.00514	0.8952096	0.0057419	DU
	7MNPB	0.035514	4.9101796	0.0072327	NAT
	7MOPB	0.016822	2.7844311	0.0060416	DU
	7R306PB	0.238318	51.197605	0.0046549	DU
DARHT	DX-DARHT-400	0.009346	2.3952096	0.0039019	DU
	DX-DARHT1-1200	0.056075	11.676647	0.0048023	DU
	DX-DARHT2-1200	0.028037	5.4191617	0.0051737	DU
	DX-DARHT3-1200	0.018692	3.7724551	0.0049548	DU
	DX-DARHT4-1200	0.018692	4.011976	0.0046589	DU
	DX-ENNIE1-250	0.028037	9.7904192	0.0028638	DU
	DX-ENNIE2-350	0.023364	5.0299401	0.0046451	DU
Area G (*Pinus edulis)*	6 AreaJ-E	0.005093	0.8443114	0.0060327	DU
	7 AreaJ-N	0.003411	0.7814371	0.0043653	DU
	8 Soilloca #8	0.006729	1.7095808	0.003936	DU
	9 TruPads-E	0.00729	1.0508982	0.0069367	NAT
	CdBueyS1	0.003645	0.9461078	0.0038525	DU
	CdBueyS2	0.001495	0.5359281	0.0027902	DU
	CdBueyS3	0.002804	0.5808383	0.0048271	DU
	CdBueyS4	0.003832	0.5808383	0.006597	NAT
	CdBueyS5	0.005374	0.7994012	0.0067223	NAT
DX West	# 1 DX WEST	0.003692	0.508982	0.0072529	NAT
	# 2 DX WEST	0.004673	0.7185629	0.0065031	NAT
	# 3 DX WEST	0.004673	1.1676647	0.0040019	DU
	# 4 DX WEST	0.004673	0.988024	0.0047295	DU
	# 5 DX WEST	0.001542	0.2694611	0.0057227	DU
	# 7 DX WEST	0.001542	0.5688623	0.0027108	DU
	# 8 DX WEST	0.003738	0.6886228	0.0054287	DU
	# 9 DX WEST	0.00215	0.3592814	0.0059829	DU
	# 11 DX WEST	0.004673	-	-	-
	# 12 DX WEST	0.004346	0.8982036	0.0048383	DU
DX South	13	0.004673	1.1976048	0.0039019	DU
	14	0.004673	0.9580838	0.0048773	DU
	15	0.004673	1.2874251	0.0036296	DU
	20	0.004673	1.2275449	0.0038067	DU
	16-Live	0.004673	0.8982036	0.0052025	DU
	17-Live	0.004673	1.0778443	0.0043354	DU
	18-Live	0.004673	0.8982036	0.0052025	DU
	19-Live	0.028037	5.239521	0.0053511	DU
DX North	21	0.004673	0.4191617	0.0111482	E
	22	0.004486	0.5988024	0.0074916	NAT
	23	0.002243	0.2694611	0.008324	E
	6 Live	0.009346	2.1556886	0.0043354	DU

*The tendency or dominance is <0.0065 = depleted uranium ("DU"), 0.0065–0.008 = natural uranium ("NAT"), and >0.008 = enriched uranium ("E").