3-31-2006

Radioisotope Sampling in the Valles Caldera National Preserve

Pueblo of Jemez

Greg Kaufman

Sampling for Radioisotope Impacts from Los Alamos National Laboratory in the Valles Caldera National Preserve

A Final Report To the Citizens' Monitoring and Technical Assistance Fund

By Greg Kaufman
Pueblo of Jemez Department of Resource Protection

May 31, 2006

Table of Contents

1. Executive Summary 3
2. Historical Context of Project 6
3. Methodology 8
   - 3.1. Project Objectives 8
   - 3.2. Sample Collection 8
   - 3.3. Laboratory Analysis of Samples 9
4. Data Analysis 11
   - 4.1. Gross Alpha Radiation 12
   - 4.2. Gross Beta Radiation 14
   - 4.3. Composite Sample Results 15
5. Discussion and Conclusions 18
6. References 19

1. Executive Summary

This report describes the results of radioisotope measurement in air samples obtained in the Valles Caldera National Preserve (the Caldera) in northern New Mexico. The study measured Gross Alpha and Beta radiation as well as concentrations of Plutonium, Uranium and Americium isotopes. The Preserve is the ancestral domain of the Jemez (Hey'-mes) Pueblo Indians and is the location of countless Jemez cultural artifacts, shrines and holy places. The Caldera is also a sensitive and unique ecosystem that supplies the Jemez people with traditional plants and minerals as well as game for food. The head waters of the Jemez River form in the expansive Valles Grande (see cover photo) within the Preserve and eventually flow through the Pueblo itself, 40 miles to the south, providing irrigation and drinking water to the 2500 inhabitants of the Pueblo.

A mountain ridge separates the Caldera from the Pajarito Plateau to the east where the Los Alamos National Laboratory (LANL) is situated. For 63 years, LANL has been engaged in radioisotope research, high explosives testing, nuclear weapons design and myriad other heavy industrial activities. Robert Oppenheimer, Niels Bohr, Enrico Fermi and other icons of science developed the first atomic bomb there between 1943 and 1945. The bomb was detonated in central New Mexico at the Trinity Site on June 16, 1945. Today the lab employs 13,643 scientists, engineers, staff and contractors and is charged with protection of the United States by maintaining the functionality of the U.S. nuclear weapons arsenal. The Lab occupies 40 square miles and includes 537 occupied buildings spread across 49 technical areas.

LANL has several air emission sources that exhaust a wide variety of radionuclides including Plutonium-238, 239 and 240, Uranium-234, 235 and 238, Americium-241, and other fission materials. The Pueblo of Jemez Department of Resource Protection (DRP) has detected these and other radioisotopes in soil, stream water and stream sediment in the Caldera and on Pueblo Trust land at concentrations that, for many results, are twice the regional background concentration developed by LANL scientists. The DRP is currently studying, with the cooperation of LANL, whether the detected isotopes originated at LANL or from global fallout from nuclear weapons testing.

This study considered the hypothesis that radioisotopes emitted from LANL migrate to the west during winter months when the wind is frequently out of the north-northeast and are deposited on soils and water bodies in the Caldera. Over time, they are washed into the Jemez River and are ultimately deposited in river sediment. Since the Jemez River is heavily sediment laden, the concern is that sediments deposited on Jemez crops via irrigation flows could present a health issue for the Jemez People who grow and consume their own food.

The first step in investigating this hypothesis was to measure the concentration of radioisotopes in the air within the Caldera. The Jemez DRP obtained a grant from the Citizens' Monitoring and Technical Assistance Fund to install an air sampler in the Caldera. The DRP obtained a Radiological Air Sampling Network (AIRNET) sampling unit identical to those used by LANL to measure radioisotope concentrations in air around the LANL facility. With the assistance of

LANL's Air Quality Group (ESH-17), this unit was installed near the center of the Caldera. Samples were obtained throughout 2005 and were analyzed using the same sampling and analytical protocols used by LANL in their AIRNET program. The results indicate:

1. The concentration of Gross Alpha emitting radioisotopes is elevated compared to average regional concentrations measured by LANL. The Gross Alpha concentrations also exceed the national average calculated by the National Council on Radiation Protection and Measurements (NCRP) on which EPA regulatory standards for individual exposure are based.

2. Americium-241 and Plutonium-239/240, which are Alpha radiation-emitting isotopes, were measured in concentrations greatly exceeding the average regional concentrations measured by LANL but below the Department of Energy's *Derived Concentration Guidelines for Air Inhalation by the Public*.

3. The concentration of Gross Beta emitting radioisotopes is below average regional concentrations measured by LANL as well as the national average calculated by the NCRP.

The author would like to thank the Citizens' Monitoring and Technical Assistance Fund; Drs. Craig Eberhard and Jean Dewart of the LANL Air Quality Group for their technical assistance and generosity in loaning an AIRNET sampling unit to the Pueblo and Dr. Robert Parmenter of the Valles Caldera National Preserve for his assistance in siting the AIRNET sampler in the Caldera.


COLORADO
Valles Caldera
National Preserve
OK
Santa Fe
Albuquerque
ARIZONA
New Mexico
Las Cruces
TEXAS
MEXICO
New Mexico Department of Tourism Map

2. Historical Context of Project

The Pueblo of Jemez (Hey'-mes) is a federally recognized Indian Tribe inhabiting north-central New Mexico. Approximately 3,000 tribal members live in the Pueblo. The Pueblo is essentially a small municipality with an autonomous, sovereign government led by the Pueblo Governor, and 1st and 2nd Lieutenant Governors. They are assisted by a Tribal council that is comprised of former Pueblo Governors. In addition to this secular leadership, there is a system of religious leaders who also advise the Governors and Tribal Council. Approximately 240 individuals are employed by the Pueblo.

The Jemez People have occupied the river valley and surrounding mountains that bear the Jemez name for approximately 800 years. They are among the dozens of Pueblo cultures that were impacted by Spanish colonial occupation and settlement beginning in 1540. When the Mexican-American War ended in 1848, the Treaty of Guadalupe Hidalgo was executed by the Mexican and American Governments. This treaty brought into American control vast land holdings taken from the Pueblos by the Spanish Crown and given to Spanish settlers as grants. The Jemez, Cochiti, San Ildefonso, Santa Clara and other Pueblos lost ancestral lands held for centuries to the U.S. government this way including the land on which LANL was eventually constructed.[1] The Valles Caldera National Preserve to the immediate west of Los Alamos passed only recently from its original Spanish grantees, the Baca Family, to the federal government which purchased the 90,000 acre tract in 2000 for $101,000,000.[2]

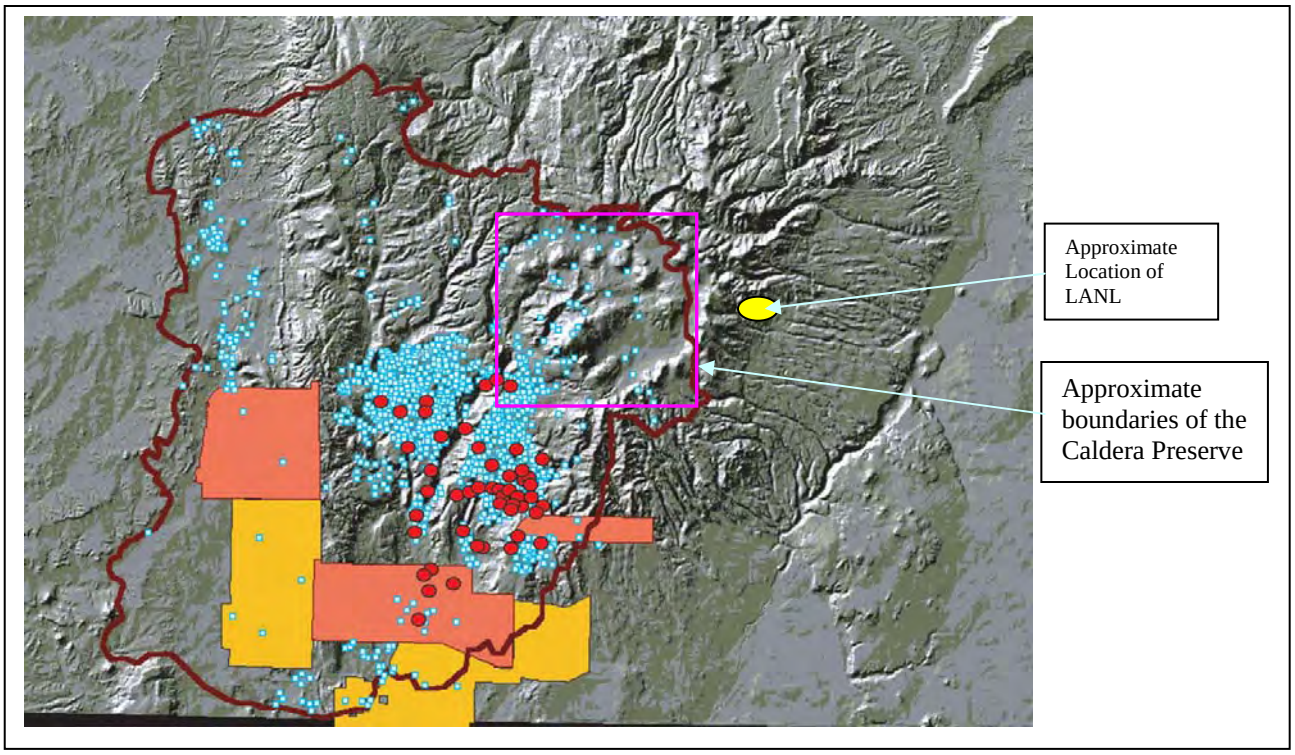
Currently, the Jemez People are custodians of 89,000 acres of land within a 40 mile radius of LANL. The Jemez ancestral domain includes the Caldera which contains Jemez cultural artifacts, shrines and holy places (see below). The Caldera is also a sensitive and unique ecosystem that supplies the Jemez people with traditional plants and minerals as well as game for food. The head waters of the Jemez River form in the expansive Valles Grande and eventually flow through the Pueblo itself, 40 miles to the south, providing irrigation and drinking water to the 2500 inhabitants of the Pueblo. Given the history of the Lab and confirmed environmental impacts at the LANL site, the Jemez people are understandably concerned about exposure to radioactive compounds and other toxins that may have been deposited on their ancestral and trust lands.

In 2001, the Jemez DRP began a soil study aimed at measuring the concentration of radionuclides in soil samples collected in the Valles Caldera National Preserve. Preliminary soil sample results reveal detectable amounts of Cesium-137, Plutonium-239/240, and Gross Beta Radiation, in concentrations greater than the regional background measure for soils.[3] When LANL scientists were presented with these preliminary findings they asserted that the measured concentrations were naturally occurring given the volcanic geology of the region. While Gross

[1] Land on which LANL was constructed passed from the Mexican government to the U.S. Government which sold parcels to private owners. A boy's school occupied the land until it was condemned by the U.S. government in 1943 for the construction of the then secret research lab.
[2] www.vallescaldera.gov
[3] LANL Regional Statistical Reference Levels (Fresquez, et al., 1996).

Alpha and Beta radiation could result from naturally occurring radioisotopes, Cesium-137 and Plutonium-239/240 are fission products; man-made nuclear materials.

The Pueblo of Jemez has been interested in confirming the soil sampling results by taking air samples in the vicinity of the soil sampling locations to see if the same radioisotopes are detected. This would give us an idea of what radioisotopes are currently being deposited on the soil and water bodies as opposed to those measured in soil samples which could be naturally occurring, or have been deposited anytime in the last 60 years.



3. Methodology

3.1. Project Objectives

The goal of this project was to assess the extent of radioisotope contamination on Jemez ancestral lands in the Valles Caldera National Preserve. This data collection effort was part of a larger, on-going project to measure radioisotope and heavy metal concentrations on both Jemez ancestral lands and Jemez trust lands. Once the on-going project is completed, appropriate steps can be taken to mitigate any health threat to the Jemez People.

For the study at hand, the analysis of air sampling results will help us understand:

- Whether airborne radioisotopes are reaching the upper Jemez River watershed.
- The relationship of the measured concentrations to regional background levels. If the concentrations are statistically greater than background, it will be reasonable to infer that LANL emissions are impacting the upper Jemez River watershed. If the concentrations are statistically lower than regional background, then the radioisotopes can be contributed to global fall-out.[4]
- If the concentrations are greater than regional background then the assertion that elevated radioisotope concentrations measured in soil are due to naturally occurring conditions can be refuted.

To reiterate, our hypothesis asserts that LANL's radioactive air emissions are:

a) above regional background measures (established by LANL)
b) are mobile in the upper atmosphere and are being blown toward the upper Jemez River watershed at least 50% of the time based on a combination of LANL and Jemez derived windrose data
c) being deposited on the soil in the upper Jemez River watershed.

3.2. Sample Collection

LANL collects radioisotope data from ambient air using a set of air sampling stations called the Radiological Air Sampling Network (AIRNET). The Pueblo of Jemez maintains one of these remote stations at the DRP office on the Pueblo. In order to create analytical results with the highest degree of comparability with those gathered by LANL, the Pueblo duplicated LANL's sampling and analytical protocols to the greatest degree possible.

[4] According to LANL publications on the calculation of background values, these values include global fallout and naturally occurring radioisotopes. Thus, any concentrations that exceed background can be attributed to LANL operations (McLin & Lyons, 2002).

An identical AIRNET station to those used by LANL was installed in the Valles Caldera by a team comprised of scientists from the Jemez DRP, the Caldera, and LANL. LANL supplied the AIRNET unit fitted with a Total Suspended Particulate pump and filter mechanism. Jemez DRP worked with Dr. Bob Parmenter of the Caldera staff to site the AIRNET station near a newly installed meteorological station. This will allow air sample results to eventually be correlated with wind speed and direction data at some future date. Power to the AIRNET station was connected by a commercial electrical contractor from a nearby powerline.

Once installed, the AIRNET station continuously pumped ambient air at a specific flow rate through 47mm Dynaweb polyester filters prepared by Paragon Analytics of Fort Collins, Colorado. Paragon is the same lab that analyzes LANL's AIRNET samples and was contracted by Jemez to do the same analyses on the filters used in this project. Suspended particles in the air were trapped on the filter creating a single sample. The samples were collected by Jemez DRP technicians on a two-week cycle during the eleven-month project period. Since the AIRNET location was at approximately 9,000 feet in elevation, access to the AIRNET station required skiing into the site during the winter months.

On a few occasions, the filter sampling time was extended by a few days due to weather. Since the sampling result is calculated as an average amount of radioactivity per cubic meter of air pumped through the sample, it is unlikely that minor variability in sampling times affected the validity of any one result.

3.3. Laboratory Analysis of Samples

As the samples were collected, they were sent to Paragon Analytics for radiochemical analysis. Jemez established a contract and scope of work with Paragon identical to those used by LANL. The scope of work was based on the Sampling and Analysis Plan and Data Quality Objectives used by LANL for their own AIRNET samples.

Instrumental gamma-emitting nuclide analysis was conducted on each sample prior to the determination of Gross Alpha and Beta activity. Each whole filter sample was individually front-face counted by proportional counting to determine Gross Alpha and Gross Beta activities. When the Alpha and Beta analyses were completed, each filter was split in half. The halves that accumulated over a calendar quarter were combined in a composite sample that was analyzed for Americium-241, Plutonium-238 and 239, Uranium-234, 235, and 238.

Quality Assurance/Quality Control samples developed by the lab included a laboratory control sample, a preparation blank, a matrix blank, and a matrix spike. Paragon additionally qualified any result that did not meet pre-determined quality control parameters defined in the contractual scope of work.

4. Data Analysis

Paragon Analytics transmitted the sampling results to Jemez DRP in electronic and hard copy formats. The sample results included analytical concentrations for each analyte in picocuries (pCi) per sample (filter) with analyte uncertainty, minimum detection limits and quality assurance qualifications (if any) listed for each result. Each result reported was the total radioactivity measured in each filter. In order to make the data comparable to LANL regional AIRNET data, it was necessary to calculate the average radioactivity for a given analyte per cubic meter of air and convert the result from picocuries (pCi) to femtocuries (fCi).[5] This was performed using the following calculation:

$$\text{Avg. fCi/m}^3 = \frac{C_{pCi}\ 1000}{[(F_1+F_2)/2]\ T_{min}\ .028316847}$$

where:

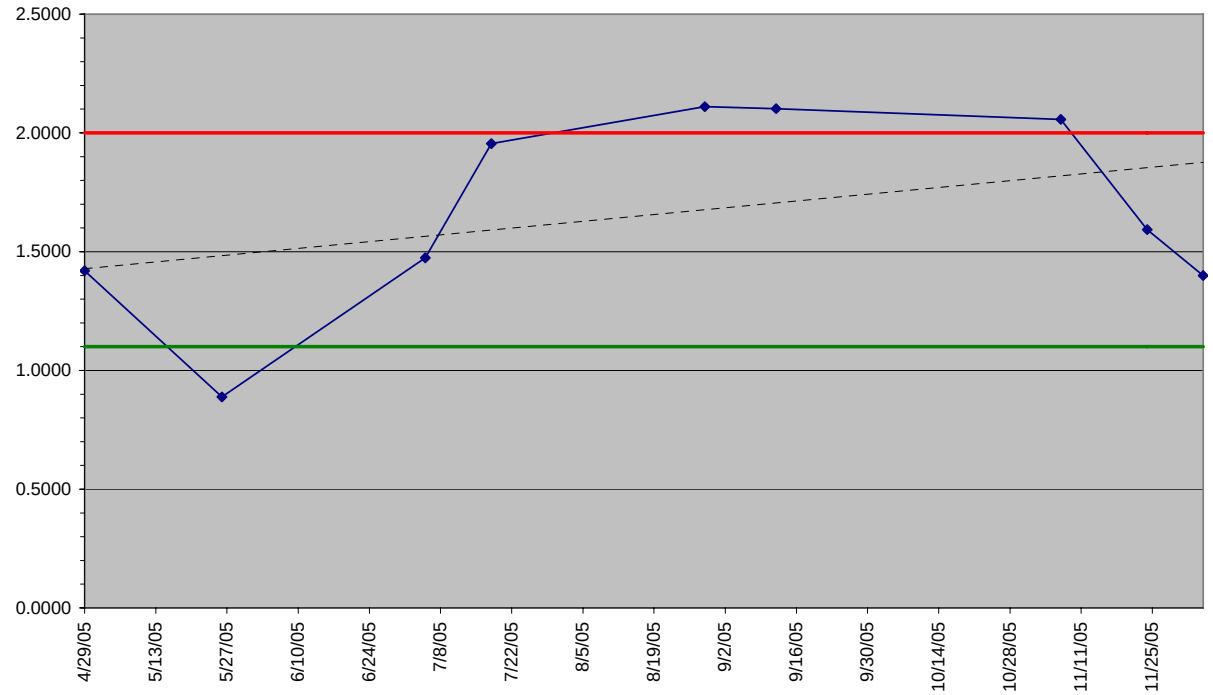
C_{pCi} = concentration of analyte measured in sample in picocuries
F_1 = starting air flow rate in cubic feet per minute
F_2 = ending air flow rate in cubic feet per minute
T_{min} = time in minutes the sample was exposed to ambient air (as measured by the AIRNET unit)

All data qualified by the Lab were culled from the data set as were duplicate sample results.[6] To maintain parity with LANL data, only the results that had a value greater than three times the standard deviation of the result's uncertainty were used creating a 95% confidence interval for the resulting data set.

[5] A picocurie equals 10^{-12} curies. A Femtocurie equals 10^{-15} curies.
[6] All qualifications associated with non-QA/QC samples were "U" since the measured result was below the minimum detectable concentration for the analytical method used. LANL includes U-qualified data in their data sets to gain a larger set of values from which to calculate the standard deviation. There are pros and cons associated with this approach. Jemez has elected not to use the U-qualified data.

4.1. Gross Alpha Radiation

Table 1. Average Gross Alpha Radiation in fCi/m^3 from Caldera AIRNET Station 2005



NCRP National Average for Gross Alpha Radiation[7]
LANL-measured Mean Regional Gross Alpha Radiation, 2004
Measured Gross Alpha Concentrations
95% Confidence Interval +/-0.3fCi/m^3

Table 1 documents the concentrations of Gross Alpha radiation measured by the AIRNET station in this study. The data included in the table represent only those measurements exceeding the 95% confidence interval. The dotted trend line (representing *method of least squares* calculation) indicates that the concentrations generally increased throughout the project period and peaked during the late Summer and early Fall at levels exceeding both the national average and LANL's Mean AIRNET-measured concentration for Gross Alpha radiation.

The prevailing opinion is that decay products of radon are the cause of measurable Gross Alpha and Beta radiation concentrations nationwide (NCRP 1975, NCRP, 1987, LANL, 2004). The graph above indicates a departure from the normal annual cycle measured by LANL where concentrations generally are higher in the winter due to weather inversions that trap radon closer

[7] National Council on Radiation Protection Measurements

to the ground.[8] Another cause could be the many alpha-emitting radioisotopes exhausted from LANL. In fact, LANL uses measurement of Gross Alpha and Beta radiation as a screening method for elevated levels of individual Alpha and Beta emitting radioisotopes (LANL, 2004). Possible Alpha emitting radioisotopes include:

- Americium-241
- Curium-244
- Plutonium-238, 239 and 240
- Uranium-238
- Thorium-232
- Radium-226

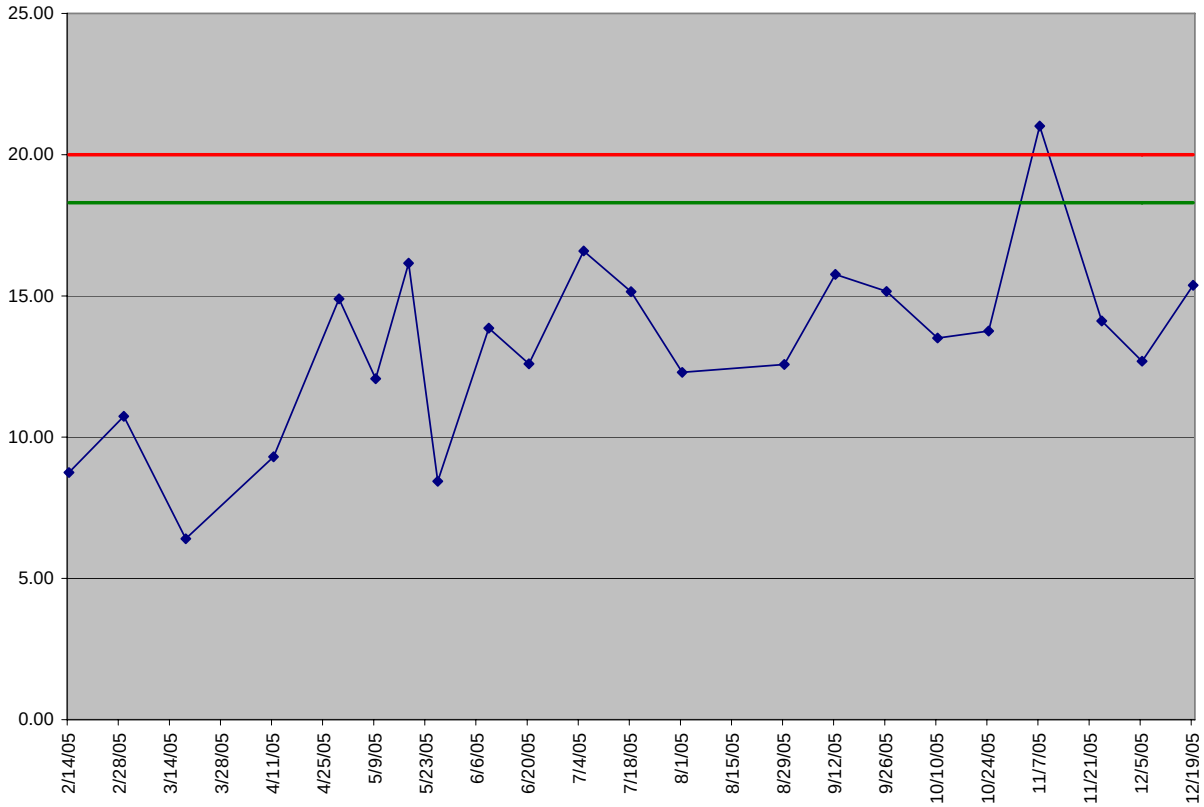
Part of the hypothesis formed at the outset of this project was that wind could be depositing radioactive particles from LANL on the Caldera. The graph above, however, indicates that the higher concentrations of alpha radiation were measured during the summer months when the prevailing winds are out of the southwest. These winds would tend to carry any radioactive particles away from Los Alamos and the Caldera toward the northeast. Thus, this component of the hypothesis is not proven by the data.

The Valles Caldera, and the corpus of the Jemez Mountains, are volcanic in nature. Uranium is naturally occurring in abundance in this area and it is possible that uranium and other radioisotopes found in regional soils are contributing to the elevated Gross Alpha radiation measurements.

[8] LANL, 2004

4.2. Gross Beta Radiation

Table 2. Average Gross Beta Radiation in fCi/m^3 from Caldera AIRNET Station 2005



_____ NCRP National Average for Gross Alpha Radiation[9]
_____ LANL-measured Mean Regional Gross Alpha Radiation, 2004
_____ Measured Gross Alpha Concentrations
95% Confidence Interval +/-1.0fCi/m^3

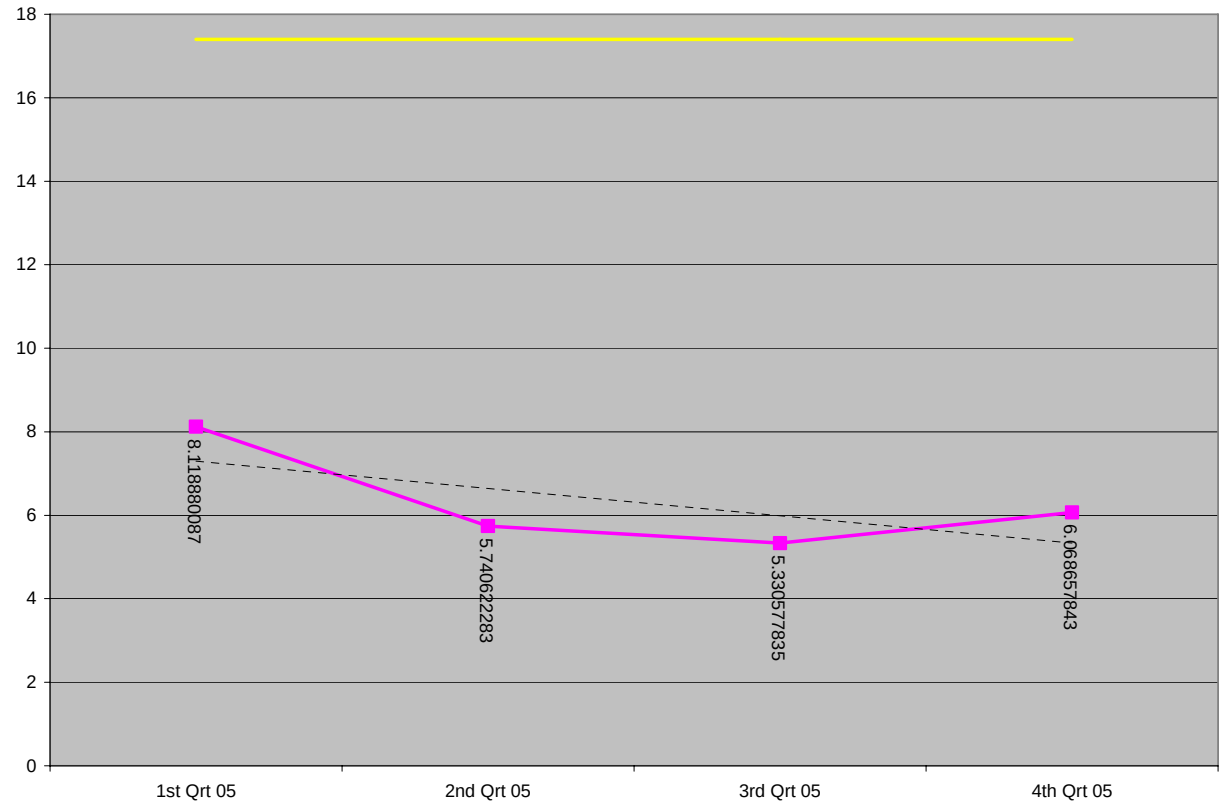
Table 2 documents the concentrations of Gross Beta radiation measured by the AIRNET station in this study. The data included in the table represent only those measurements exceeding the 95% confidence interval. The dotted trend line (representing *method of least squares* calculation) indicates that the concentrations generally increased throughout the project period. Unlike the Gross Alpha measurements, however, the Gross Beta data do not exceed either NCRP or LANL averages (with one exception). The wind deposition hypothesis also is not supported by the Gross Beta data since the higher concentrations were measured during the summer months when the prevailing wind is out of the south and southwest.

[9] National Council on Radiation Protection Measurements

4.3. Composite Sample Results

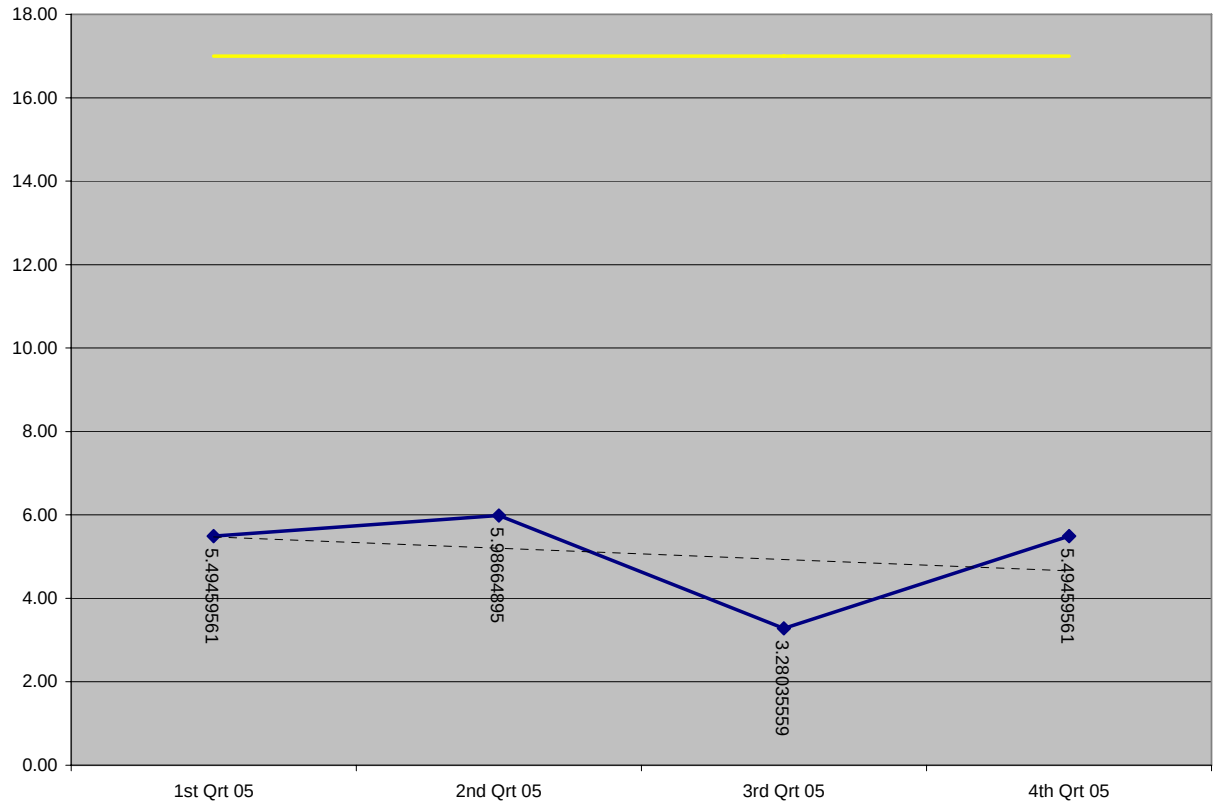
Samples comprised of halves of filters used to collect Gross Alpha and Beta samples were composited on a quarterly basis. Each composite sample was analyzed for Americium-241, Plutonium-238, 239 and Uranium-234, 235, and 238. Once qualified data, lab QA/QC sample data and data that did not meet the 95% confidence level test were removed from the data set, only U-234 and 238 had acceptable data for all four quarters. The data are summarized in Tables 3 and 4 below. These data indicate that concentrations of the uranium isotopes are well below the regional mean measured by LANL and also below the DOE's guidelines for air exposure. Further, as the year progressed, the concentrations of Uranium isotopes diminished.

Table 3. Average Quarterly U-234 Concentration in aCi/m^3 from Caldera AIRNET Station 2005



DOE Derived Concentration Guidelines for Air Inhalation of U-234 by the Public = 90,000 aCi/m^3
LANL-measured Mean Regional U-234 Concentration, 2004
Measured U-234 concentrations
95% Confidence Interval +/-1.22 aCi/m^3

Table 4. Average Quarterly U-238 Concentration in aCi/m^3 from Caldera AIRNET Station 2005



DOE Derived Concentration Guidelines for Air Inhalation of U-238 by the Public = 10,000 aCi/m^3
LANL-measured Mean Regional U-238 Concentration, 2004
Measured U-238 concentrations
95% Confidence Interval +/-1.19 aCi/m^3

Am-241 was detected in only one composite sample that met all of the tests for acceptability in this study. The same is true for Pu-239/240. These results follow:

Radioisotope	**Result in aCi/m^3**	**Quarter Measured**	**LANL-measured Mean Regional U-234 Concentration, 2004 in aCi/m^3**	**DOE Derived Concentration Guidelines for Air Inhalation by the Public in aCi/m^3**
Am-241	1.476160016	1st 2005	-.4700	20,000 per week
Pu-239/240	0.656071118	3rd 2005	-.0700	20,000 per week

These results exceed the regional average for the respective isotopes but are considerably lower than the DOE's guidelines for air exposure. It is worth noting however, that both Am-241 and Pu-239/240 are man-made, alpha-emitting, fission materials. Thus, the higher than average

concentrations cannot be attributed to naturally-occurring conditions such as radioisotopes in soil. Further, since the mean concentrations measured by LANL include amounts contributed by global fallout, the heavier concentrations measured in this study may have been contributed by LANL operations given its proximity to the sampling location.

5. Discussion and Conclusions

The data derived from this study create more questions than they answer. Why, for instance, were the highest concentrations for both Gross Alpha and Beta measured in the warmer months of the year when there are no weather inversions to keep radon close to the ground? Why are the concentrations higher when the prevailing wind would tend to keep any LANL-emitted radioisotopes from entering the Caldera?

It is possible that winds aloft could move differently than wind measured at ground level and thus our presumption about the prevailing wind direction is inaccurate. It is also possible that naturally occurring Alpha and Beta emitters are more mobile in warm, dry weather conditions within the Caldera than during the winter months. It should be noted that the winter months of 2005 were above average for snowfall especially compared to recent years when a severe drought has kept snowfall to a minimum. This may have kept higher numbers of both Alpha and Beta particulates from reaching the sampler during the winter.

What the study has confirmed are elevated concentrations of Gross Alpha radiation and the presence of higher than average concentrations of Am-241 and Pu-239/240, both Alpha-emitters, in the Caldera. It has also shown that it is unlikely that LANL's *current* Gross Alpha emissions are the cause of the measured elevated concentrations given the weather conditions at the time the measurements were made. It is possible that these isotopes were in the Caldera prior to this study and were mobilized by weather conditions and trapped on the AIRNET filter. The fact that the two datum for Am-241 and Pu-239/240 were collected during the 1st and 3rd quarters of 2005 respectively, both warmer dryer periods of 2005, supports this theory.

As is frequently the case in environmental science, additional work is need to evaluate possible sources for the elevated Gross Alpha measurements in the Caldera and to characterize isotope deposition throughout the Preserve. The Pueblo of Jemez greatly appreciates the assistance of the Citizens Monitoring and Technical Assistance Fund for making this study possible.

6. References

NCRP, 1975: National Council on Radiation Protection and Measurements, "Natural Background Radiation in the United States, National Council on Radiation Protection and Measurements, Report No. 45 (November, 1975).

NCRP, 1987: National Council on Radiation Protection and Measurements, *"Recommendations on Limits for Exposure to Ionizing Radiation,"* NCRP Report No. 91 (June, 1987).

LANL ESH-17 2000: Air Quality Group, *"Sampling and Analysis Plan For Radiological Air Sampling Network (AIRNET),"* LANL Air Quality Group document EHS-17-AIRNET (August, 2000).

Rothman, 1997: *"On Rims and Ridges: The Los Alamos Area Since 1880,"* Hal K. Rothman, University of Nebraska Press, 1997.

LANL, 2002: McLin, Stephen G. and Lyons, Dale W., "Background Radioactivity in River and Reservoir Sediments Near Los Alamos, New Mexico," Los Alamos National Laboratory Report LA-13603-MA

LANL, 2004: Los Alamos National Laboratory Report LA-14239-ENV "*Environmental Surveillance at Los Alamos during 2004."*