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AMBIENT AIR SAMPLING FOR RADIOACTIVE AIR CONTAMINANTS AT LOS ALAMOS NATIONAL LABORATORY: A LARGE RESEARCH AND DEVELOPMENT FACILITY

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Ambient Air Sampling for Radioactive Air Contaminants at Los Alamos National Laboratory: A Large Research and Development Facility

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ABSTRACT

This paper describes the ambient air sampling program for collection, analysis, and reporting of radioactive air contaminants in and around Los Alamos National Laboratory (LANL). Particulate matter and water vapor are sampled continuously at more than 50 sites. These samples are collected every two weeks and then analyzed for tritium, and gross alpha, gross beta, and gamma ray radiation. The alpha, beta, and gamma measurements are used to detect unexpected radionuclide releases. Quarterly composites are analyzed for isotopes of uranium (^{234}U, ^{235}U, ^{238}U), plutonium (^{238}Pu, $^{239/240}$Pu), and americium (^{241}Am). All of the data is stored in a relational database with hard copies as the official records. Data used to determine environmental concentrations are validated and verified before being used in any calculations. This evaluation demonstrates that the sampling and analysis process can detect tritium, uranium, plutonium, and americium at levels much less than one percent of the public dose limit of 10 millirems. The isotopic results also indicate that, except for tritium, off-site concentrations of radionuclides potentially released from LANL are similar to typical background measurements.

INTRODUCTION

Los Alamos National Laboratory is a large research and development facility located in northern New Mexico operated by the University of California for the U.S. Department of Energy (DOE). It covers 43 square miles (111 km^2) and encompasses hundreds of buildings with approximately 7 million square feet of floor space. The variety of operations using radioactive materials is rivaled by few, if any, other facilities in the world. However, only a few operations have the potential to release enough material that could cause a dose greater than 0.1 millirem, one percent of the standard, to a member of the public. Continuous stack samples are collected and analyzed for these sources. Unlike a production facility or a nuclear power plant, where potentially large quantities of a few air contaminants could be released from a few locations, there are many smaller sources of radionuclides that cannot be monitored. Even if it was possible to measure and then model all atmospheric releases, the complex meteorology makes it difficult to accurately predict atmospheric concentrations. Therefore, it is necessary to measure atmospheric radionuclides to estimate the impact of LANL emissions on the public.

Ambient measurements have been collected for more than 25 years to meet the Department of Energy requirements (1) and guidance (2) for measuring the impact of radioactive emissions on local and regional air concentrations. The adoption of a 10 millirem effective dose equivalent (40 CFR Subpart H - National Emission Standards for Emissions of Radionuclides Other Than Radon from Department of Energy Facilities) on December 15, 1989 has accentuated the need for fully documented and verified measurements. Even though ambient monitoring is not required by Subpart H, two notices of noncompliance were received from the EPA in 1991 and 1992 that precipitated a signed Federal Facility Agreement in 1996 which includes requirements for ambient monitoring.

SAMPLING METHODOLOGY

LANL operates more than 50 air samplers in and around its site to collect water vapor and particulate matter. This AIR sampling NETwork is referred to as AIRNET. On-site, the samplers are generally located where there is the potential for detecting high concentrations such as near, or even in, radioactive waste disposal areas. Off-site samplers are located at the perimeter of the Laboratory, in nearby local communities including several of the Pueblos, and at regional locations.

Particulate matter and water vapor samples are collected using a rotary carbon vane vacuum pump capable of pulling 10 ft^3 of air per minute. Parallel sampling lines are used to collect particulate matter and water vapor samples for subsequent radiochemical analyses. The sampling equipment is located in an aluminum all-weather housing with louvered openings on all four sides to restrict entrance of rain and snow while providing little or no air flow restriction (3). Samples are continuously collected for two weeks unless there are power outages or equipment malfunctions. Sampling time at each location is measured by using an elapsed sample time indicator activated by an in-line vacuum-sensitive switch. Rotameters are used to measure airflow through the sampling collection media. Air volume is calculated by averaging the start and stop flow rates and multiplying this number by the hours recorded by the elapsed time indicator. All field data are entered into a spreadsheet on a palmtop computer in the field to avoid transcription errors when the data is uploaded into the AIRNET database. As an additional error prevention technique, this spreadsheet requires confirmation for measurements outside of typical values.

A documented chain-of-custody is maintained for all samples collected from the AIRNET samplers. This can include physical possession, positive control such as being locked up, or stored in a secure area where access is limited to authorized and accountable personnel. Preprinted forms or electronic entry of data are used to generate records that document field data, sample collection, and initial chain-of-custody information. Samples received by analytical laboratories are also handled according to procedures established to meet EPA chain-of-custody requirements.

Particulate Matter

Particulate matter samples are collected on a 47 mm circular polypropylene filter with about 13 cm^2 effective surface area. The filter is situated vertically within the all-weather housing. Flow rate through the filter is normally set to 4 cfm resulting in a typical face velocity of about 150 cm/sec. Particle size collection efficiency is estimated to be 100% percent for particles with an aerodynamic diameter of 0.5 μm or larger. Particles with an aerodynamic diameter less than this diameter are collected at lower efficiencies reaching a minimum collection efficiency of 80 to 90 percent at an aerodynamic diameter of about 0.1 μm (4). After collection, the samples are cut in half with half promptly sent to an analytical laboratory for alpha, beta, and gamma analysis. The other half is retained and composited quarterly for isotopic analysis.

Water Vapor

Water vapor samples are collected in vertically mounted canisters containing about 135 grams of silica gel with a volume of 220 cm^3. This silica gel is dried in a drying oven before use in the field to remove any residual water. The silica gel is used as a desiccant to remove moisture from the air, followed by distillation, condensation, collection as a liquid, and analysis for tritium. It is recognized as a method for collecting atmospheric moisture (2) (5). The air flow rate is typically 200 cm^3/min. The water mass is determined by the difference in the start and ending weights of the silica gel canisters or by the measuring the water collected by condensation of the distillate.

Radiochemistry

Individual biweekly samples are measured for gross radioactivity after each collection period. Gamma-ray spectrometry is used to measure specific gamma-ray emitting nuclides on groups of half filters. After this analysis, individual particulate matter filters are dissolved and the inorganic residue deposited onto counting planchettes for gross alpha and gross beta measurements by gas flow proportional counting.

The other half of the filters are composited, on a site-by-site basis over a calendar quarter for radiochemical analysis. These composites are analyzed for isotopes of uranium (^{234}U, ^{235}U, ^{238}U), plutonium (^{238}Pu, $^{239/240}$Pu), and americium (^{241}Am) by dissolving them in an acid solution, chemically separating and concentrating the radionuclides onto sample planchettes, and counting the concentrated samples with an alpha spectrometer.

The silica gel is heated to vaporize the water which is then collected by condensation. Tritium in the water, as an oxide (HTO and T_2O), is measured by liquid scintillation counting.

MONITORING NETWORK

As of January 1, 1998, 54 AIRNET samplers were being operated by LANL. Eighteen samplers are compliance samplers located in a standard 16 sector radial array (22.5° sector angle) with one sampler and a backup at the predicted highest dose or MEI (Maximum Exposed Individual) location. Each of these compliance samplers is sited in a location, that, within the sector, is predicted to be at or near the point of highest public impact for LANL emissions. Five off-site samplers can be considered regional stations (two in Santa Fe and one each in Espanola, Pojoaque, and Jemez Pueblo). Four additional off-site samplers are located in Los Alamos (two), San Ildefonso Pueblo, and Bandelier National Monument. The remaining 27 samplers are located within the boundaries of LANL. Figure 1 is a map that shows the locations of the AIRNET samplers on LANL property and in Los Alamos.

RESULTS

Data from three stations are reviewed in this paper. These three stations are located as follows: Santa Fe, NM a background site; Eastgate, the MEI site, located in the eastern part of Los Alamos near the LANL perimeter; and a site within Area G which is an active radioactive waste disposal site. Table 1 summarizes the ambient radionuclide concentrations for the three sites. The ninety-five percent confidence intervals in this table are based on all data that has been validated and accepted. As such, the confidence interval includes environmental, sampling, and analytical variability. Some of the high values in the data sets were apparently caused by contamination during the sample handling and analysis, but unless a specific cause was identified the data were not rejected. Therefore, the high individual values, the average concentrations, and the confidence intervals may be overestimated. Additional data summaries for all of the AIRNET sites can be found at the web page maintained by the Air Quality Group (http://www.air-quality.lanl.gov).

Tritium

Tritium is present in the atmosphere primarily due to cosmogenic production and above-ground testing of nuclear explosives. Typical global concentrations are about 0.04 pCi/m^3 (6). Local concentrations vary with the amount of moisture in the air and the sources such as operations at LANL. Figures 2 and 3 are time series graphs of the tritium data, but on different scales. Figure 2 clearly shows that tritium concentrations are several orders of magnitude greater at the Area G waste disposal site than at the other two sites. The cause of these high tritium concentrations is the tritium, from buried radioactive waste, that is diffusing through the soil and into the ambient air. This graph also shows that the diffusion rate dramatically increases during the summer months due to higher temperatures and increased evaporation and transpiration. There is clearly an on-site impact, but the average concentrations at the Area G site are only about 20 percent of the concentration equivalent to an annual dose of 10 millirems (1500 pCi/m^3).

Figure 3 is a plot of the same data, but the scale has been reduced by a factor of a hundred. A few low winter tritium values at Area G show on this scale and are comparable to the highest values recorded for Santa Fe and Eastgate. Even though both the Santa Fe and Eastgate stations have very low concentrations of tritium (normally less than 2 pCi/m^3), the Eastgate station is still significantly higher than the Santa Fe station indicating a LANL impact at this location. In addition, most of the measurements of tritium at the Eastgate site are above the MDA (Minimum Detectable Activity) whereas most of the measurements at the Santa Fe site are below the MDA. This qualitatively indicates that there are measurable concentrations at Eastgate, but not at Santa Fe given the analytical process.

Uranium

Three isotopes of uranium are naturally present in the ambient air: uranium-234, uranium-235, and uranium-238. The natural sources of uranium are crustal rocks and soils. Therefore, the ambient concentrations are dependent on the mass of suspended particulate matter, the uranium concentrations in the parent material, and any local sources. Typical uranium crustal concentrations range from 0.5 ppm to 5 ppm, but local concentrations can be well outside this range (6).

Ambient uranium concentrations are similar at the three sites, but the average concentrations of all three isotopes are higher at the Santa Fe site as shown in Table 1. Figures 4 and 6 also show that the uranium-234 and uranium-238 concentrations are consistently higher for individual measurements at the Santa Fe site. However, only the uranium-234 concentrations for Santa Fe are significantly higher than the other two sites. The uranium-235 concentrations (Figure 5) should show a similar pattern to the other two isotopes, because the relative abundances are constant, but they do not. This pattern is probably obscured because most of the measurements are below the MDA.

Low grade uranium ore bodies are known to exist in the Santa Fe area. This potential source of uranium, along with the higher ambient air concentrations of particulate matter in Santa Fe, are the probable cause of the higher uranium levels at the Santa Fe site when compared to the

Eastgate and Area G sites. The total uranium concentrations at the highest site, Santa Fe, are only about 0.4 percent of the maximum allowed dose of 10 millirems per year.

Plutonium

Plutonium is not naturally present in measurable quantities in the ambient air. All measurable sources are from nuclear explosions, the nuclear fuel cycle, and other related activities. With a few local exceptions, world-wide fallout from atmospheric testing of nuclear explosives is the primary source of plutonium. Four isotopes of concern can be present in the atmosphere: plutonium-238, plutonium-239, plutonium-240, and plutonium-241. However, plutonium-241 is not measured because it is an insignificant alpha emitter that decays by beta emission to americium-241. This beta decay in not only hard to measure, but the dose is insignificant when compared to americium-241. Plutonium-239 and plutonium-240 are indistinguishable by alpha spectroscopy and are clumped together for analytical purposes.

The majority of plutonium-238 and plutonium-239/240 measurements are less than the MDA, but there are enough measurements for the three selected sites to indicate that there is measurable plutonium at all of them. Area G is highest followed by Santa Fe and then Eastgate for both isotopes. None of these differences are significant. All concentrations are less than 0.1 percent of the maximum allowed dose of 10 millirems per year.

Americium

The americium-241 concentrations are the primary source of dose caused by the release of plutonium-241. As a decay product of plutonium-241, measurable sources are from nuclear explosions, the nuclear fuel cycle, and other related activities. Americium-241 concentrations are highest at Area G followed by Eastgate and then Santa Fe. This may be due to releases from LANL, but the noted differences between sites are not significant. All concentrations are less than 0.1 percent of the maximum allowed dose of 10 millirems per year.

CONCLUSION

This evaluation demonstrates that the AIRNET samplers operated by LANL are capable of detecting tritium, uranium, plutonium, and americium at levels much less than one percent of the public dose limit of 10 millirems. Other than very short-lived activated air emissions from accelerator operations, these radionuclides are the primary radioactive air emissions from LANL.

ACKNOWLEDGEMENTS

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Table 1. Average Radionuclide Concentrations with 95 percent Confidence Intervals

Sampling Location	Units*	Santa Fe	Eastgate	Area G
1997 Sampling Site Number		3	10	35
Tritium (as an oxide)	pCi/m^3	0.33 ± 0.28	2.5 ± 0.62	314. ± 67.
Uranium-234	aCi/m^3	14. ± 3.8	6.2 ± 1.6	9.3 ± 3.9
Uranium-235	aCi/m^3	1.2 ± 0.76	0.6 ± 0.28	0.7 ± 0.30
Uranium-238	aCi/m^3	13. ± 3.7	8.5 ± 4.9	11. ± 9.5
Plutonium-238	aCi/m^3	0.56 ± 0.37	0.36 ± 0.19	0.74 ± 0.49
Plutonium-239/240	aCi/m^3	1.3 ± 0.85	0.94 ± 0.69	1.5 ± 1.3
Americium-241	aCi/m^3	1.4 ± 0.37	1.6 ± 0.48	1.7 ± 0.54

* pCi = picocurie = 10^{-12} curies = .037 becquerels = 0.037 disintegrations per second
aCi = attocurie = 10^{-18} curies = 3.7 X 10^{-8} becquerels = 3.7 X 10^{-8} disintegrations per second

Figure 1.
Los Alamos National Laboratory
AIRNET Monitoring Sites

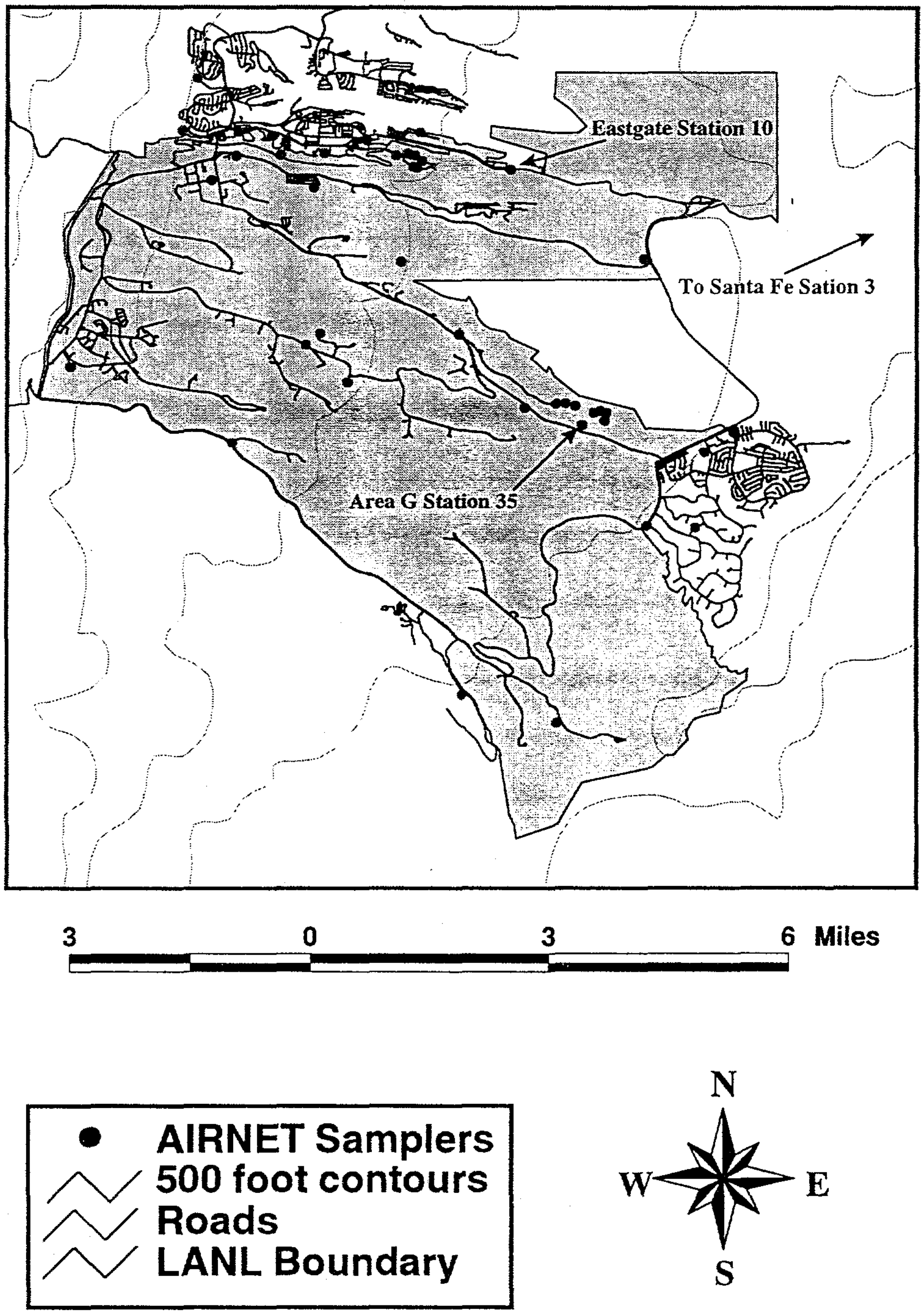

Figure 2. Tritium Concentrations in the Ambient Air (large scale)

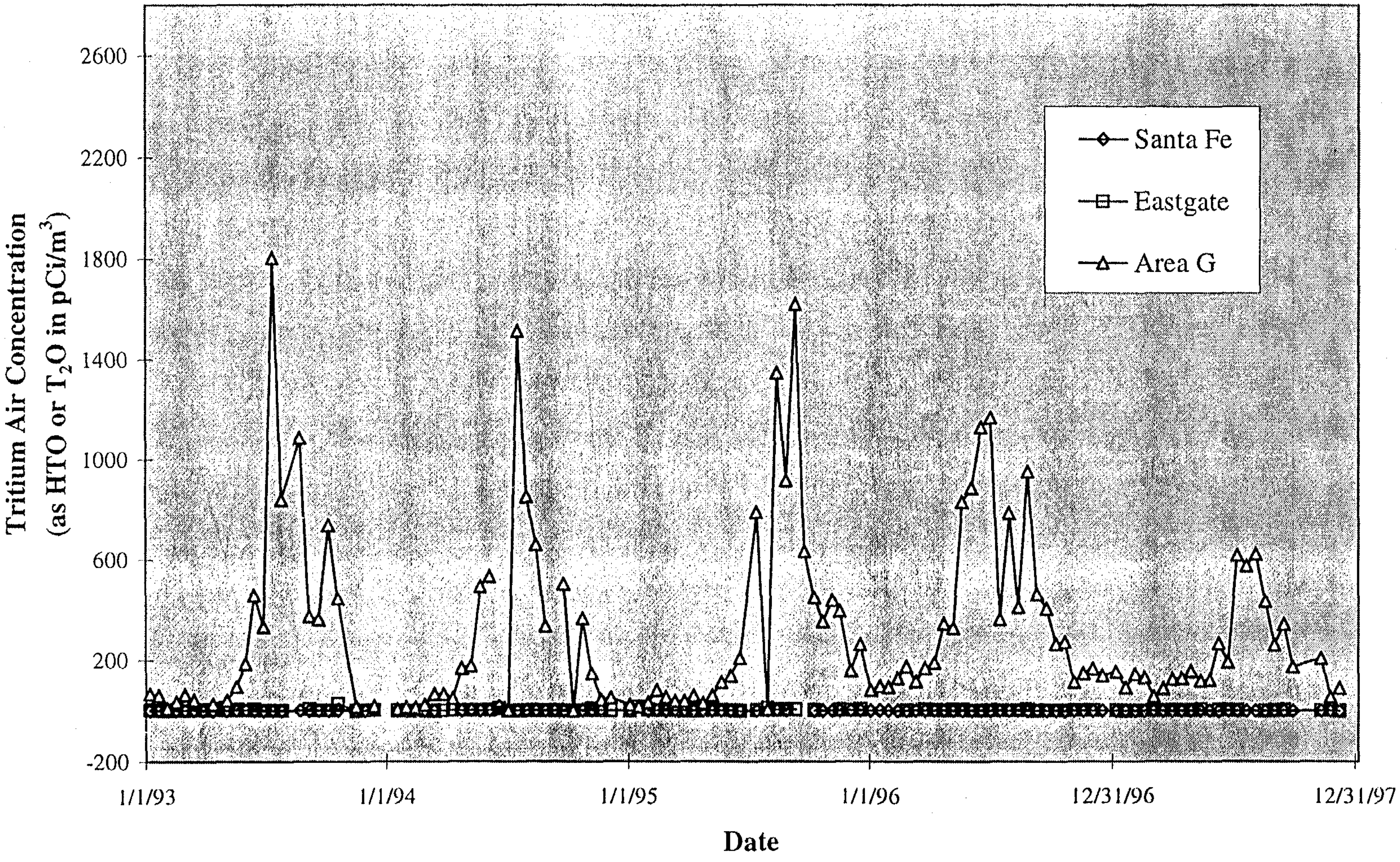

Figure 3. Tritium Concentrations in the Ambient Air
(small scale)

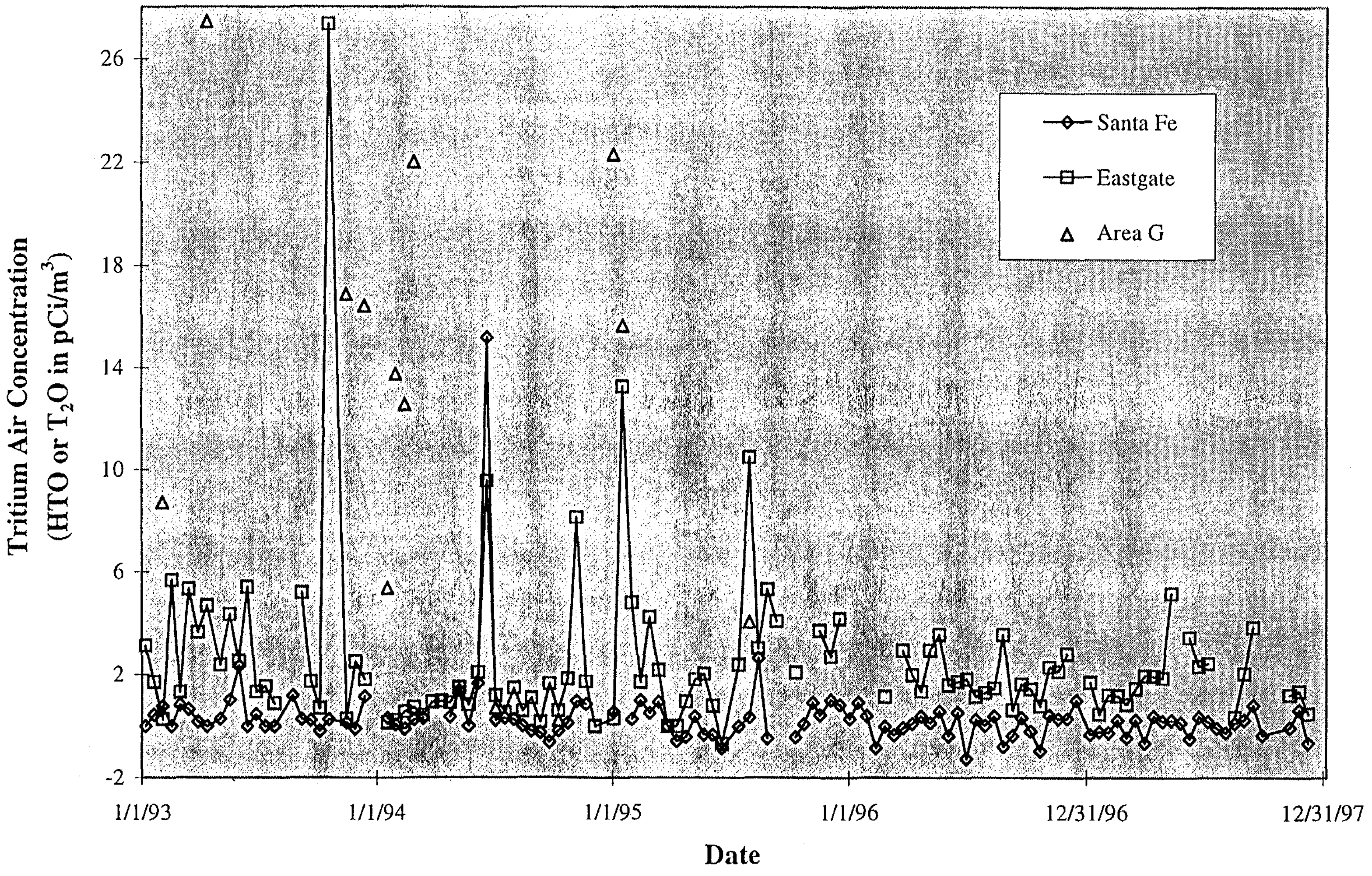

Figure 4. Uranium-234 Concentrations
in the Ambient Air

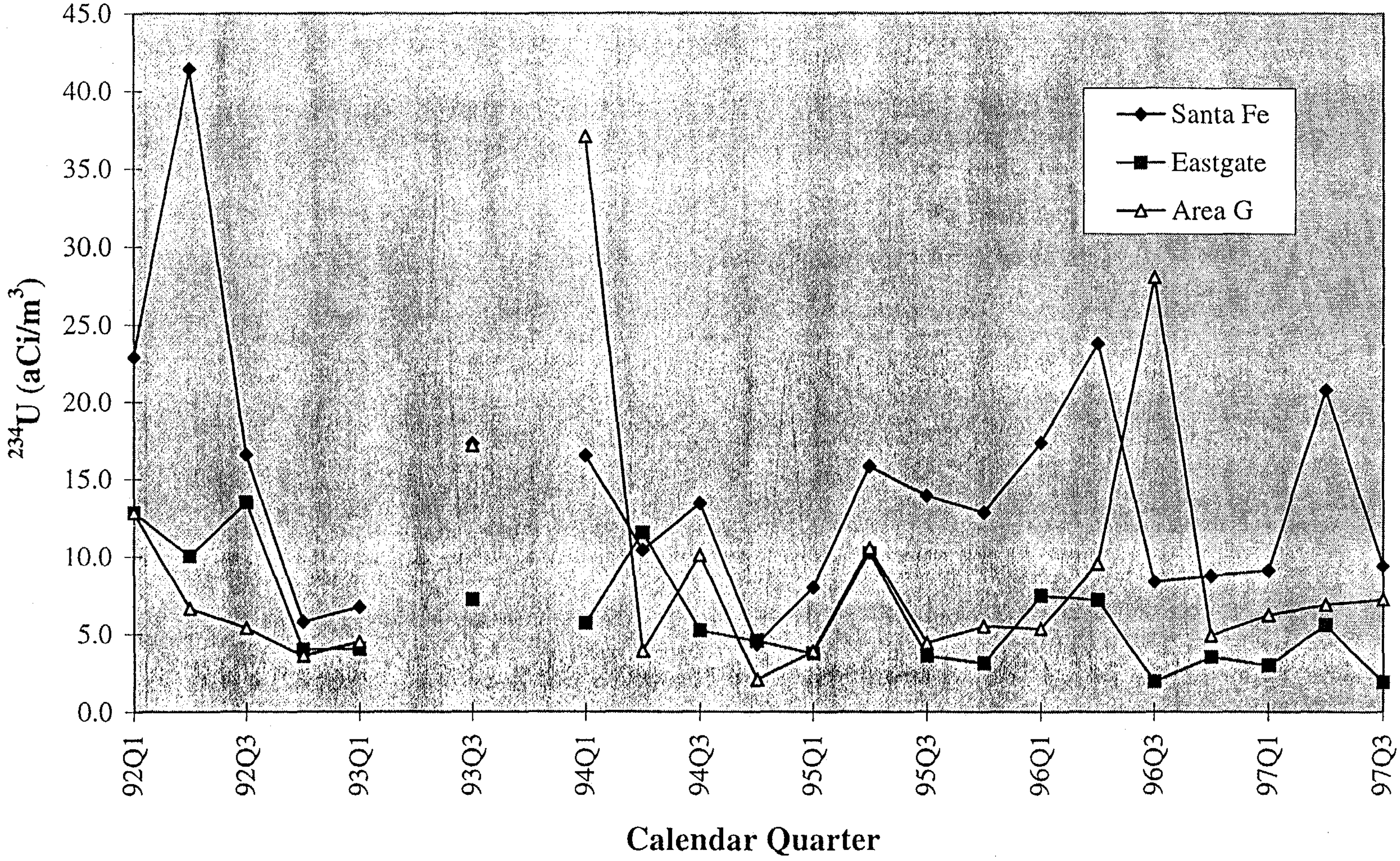

Figure 5. Uranium-235 Concentrations in the Ambient Air

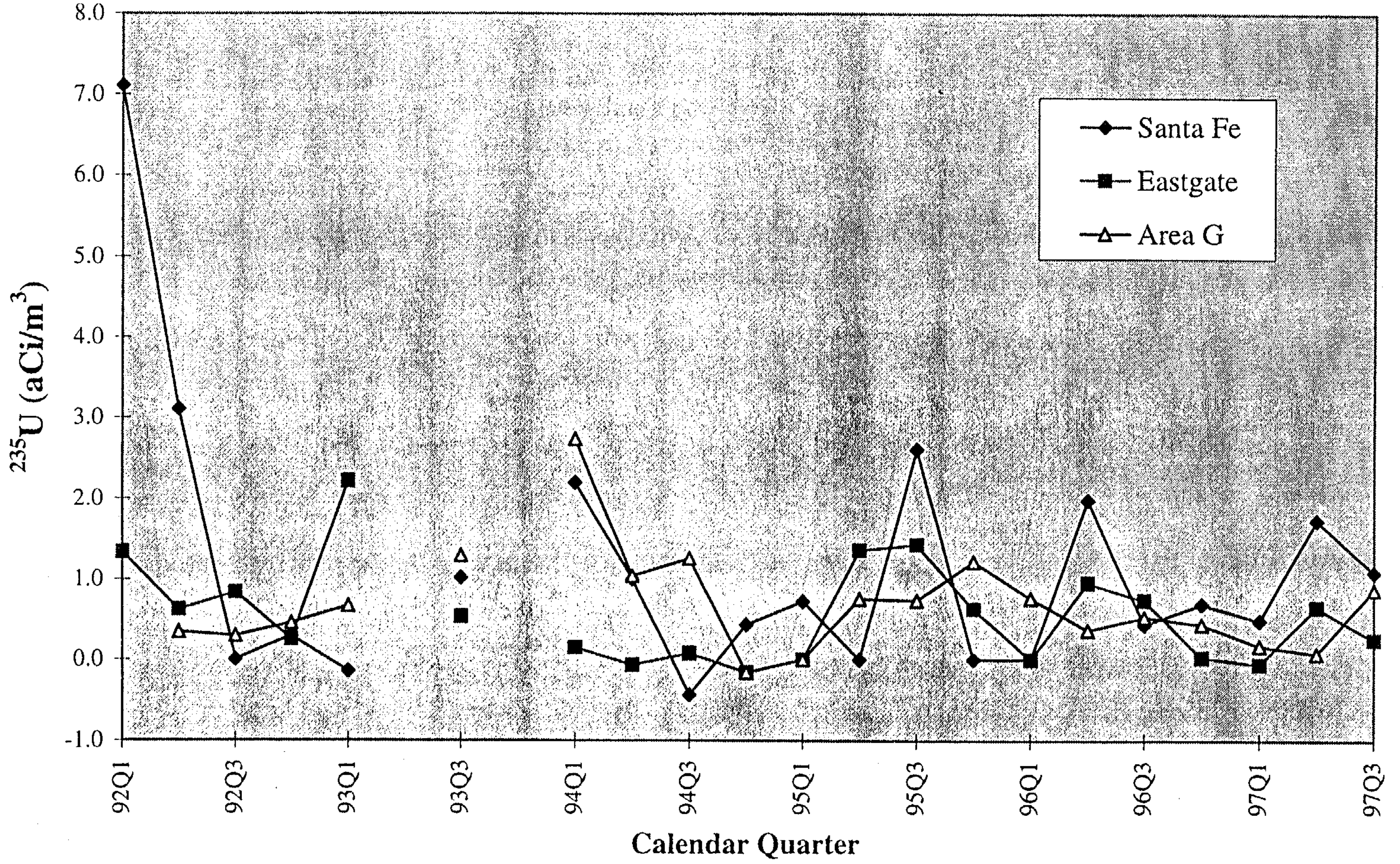

Figure 6. Uranium-238 Concentrations in the Ambient Air

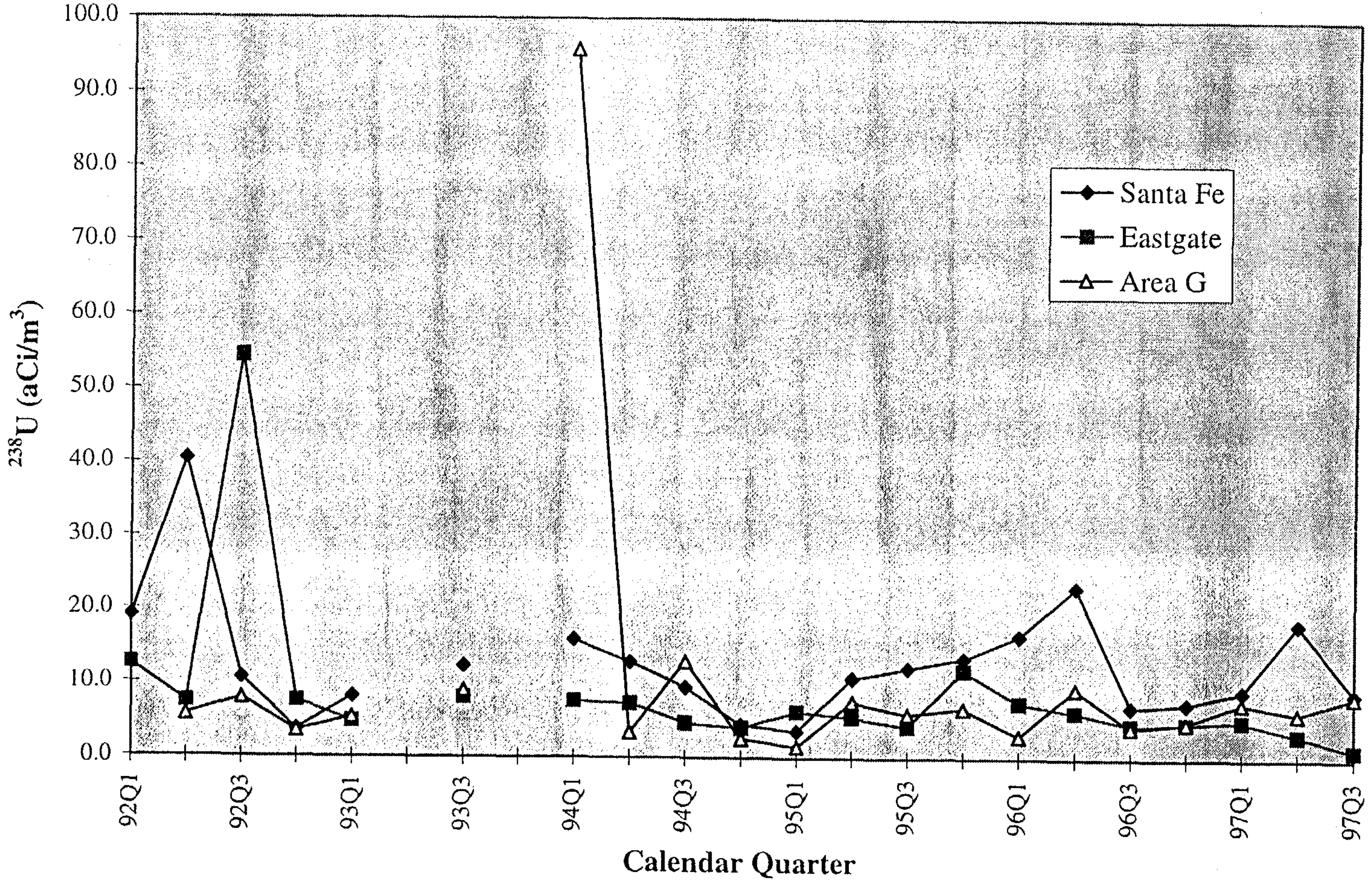

Figure 7. Plutonium-238 Concentrations in the Ambient Air

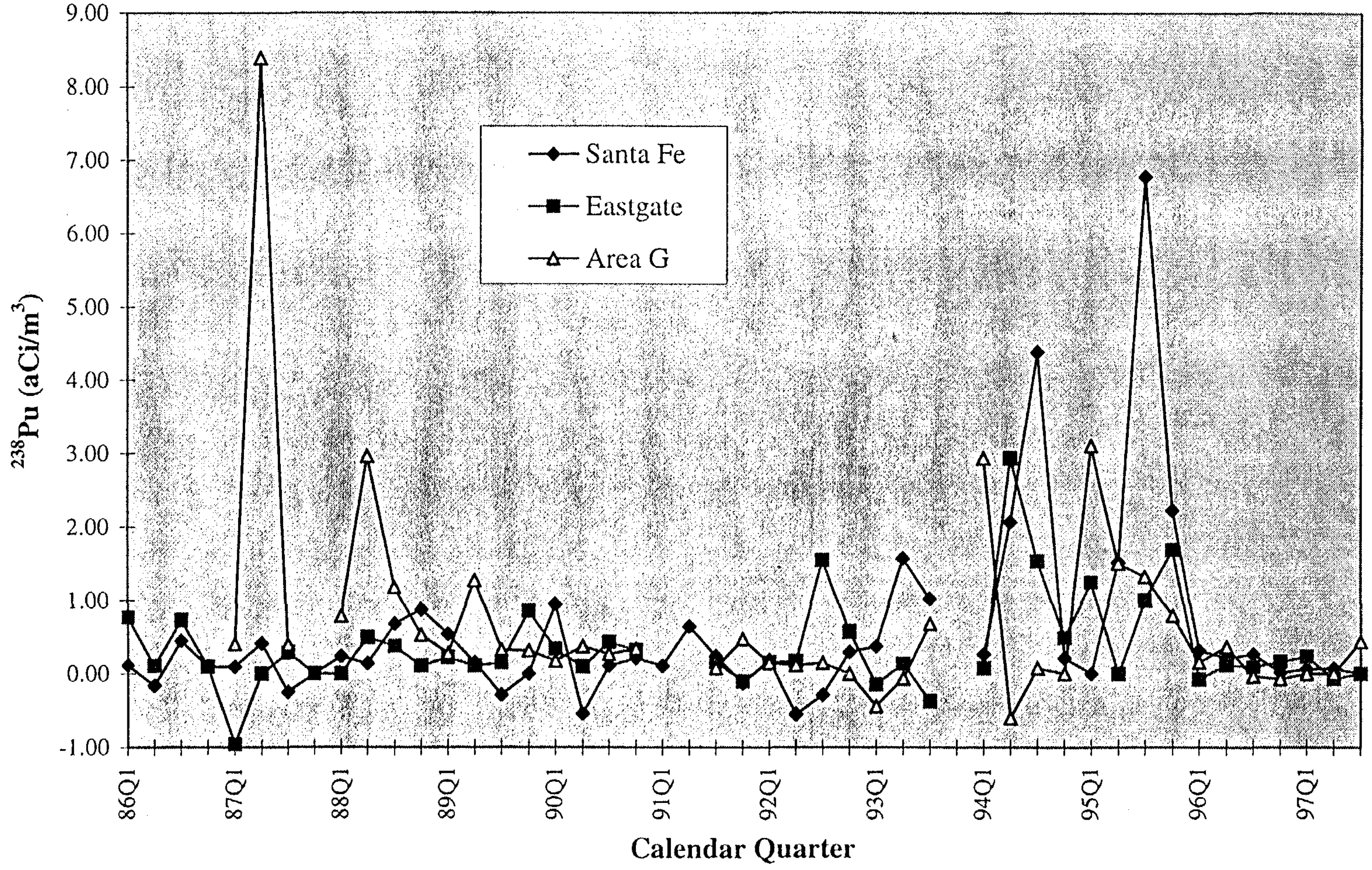

Figure 8. Plutonium-239/240 Concentrations in the Ambient Air

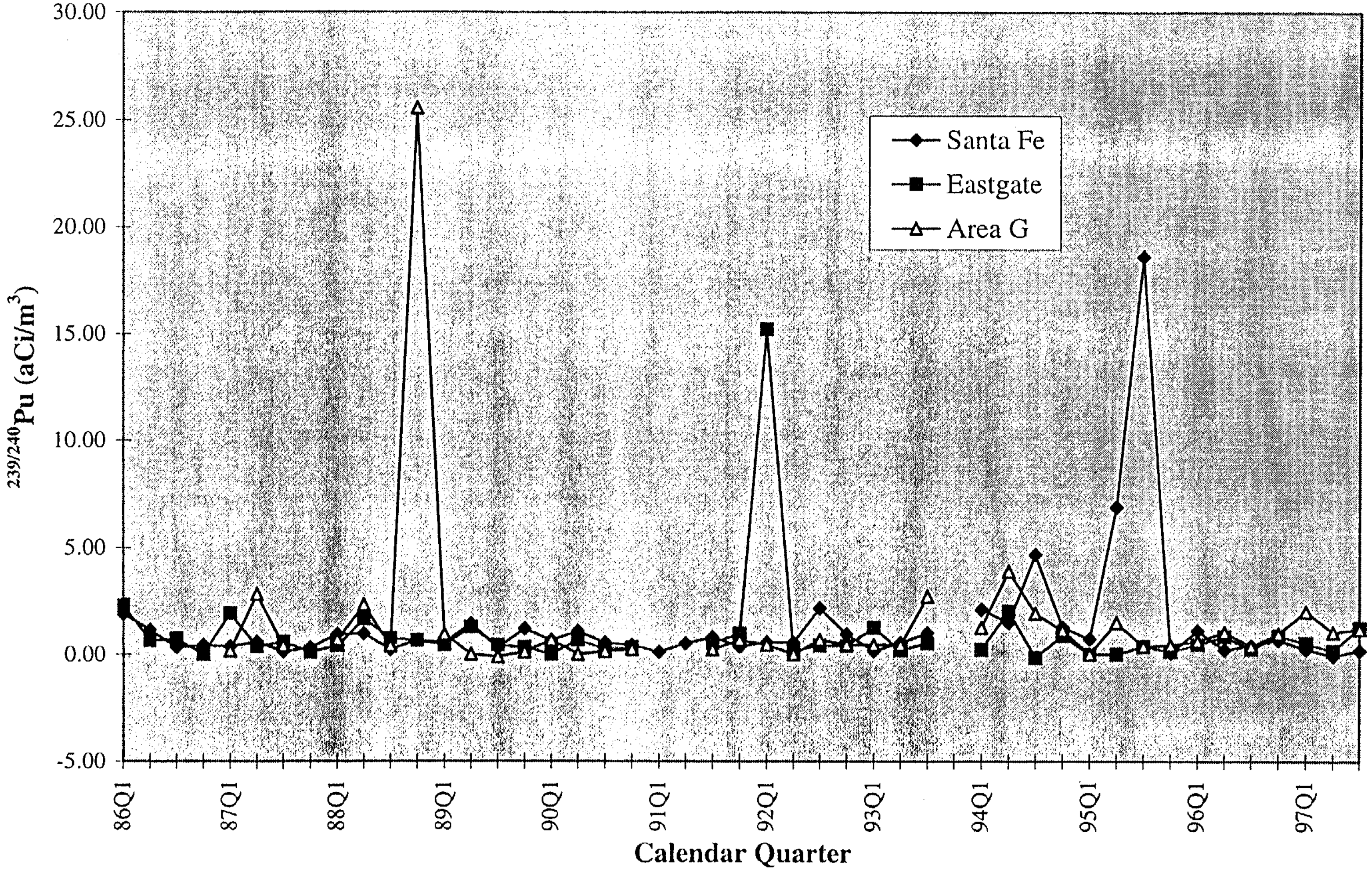

Figure 9. Americium-241 Concentrations in the Ambient Air