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Air Monitoring of Emissions from the Fukushima Daiichi Reactor

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Abstract

We report air-monitoring measurements of the fission products released from the Fukushima-Daiichi nuclear-power-plant accident in 2011. Clear gamma-spectrometry peaks were observed from Cs-134, Cs-136, Cs-137, I-131, I132, Te-132, and Te-129m. These data, together with measurements of other radionuclides, are adequate for an assessment and assure us that radionuclides from Fukushima Daiichi did not present a threat to human health at or near Los Alamos. The data demonstrate the capabilities of the Los Alamos air-monitoring systems.

Introduction

In response to the disasters in Japan on March 11, 2011, and the subsequent emissions from Fukushima-Daiichi, we monitored the air near Los Alamos using four air-monitoring systems: the standard AIRNET samplers, the standard rad-NESHAP samplers, the NEWNET system, and high-volume air samplers. Each of these systems has advantages and disadvantages. In combination, they provide a comprehensive set of measurements of airborne radionuclides near Los Alamos during the weeks following March 11.

Noble Gases and Volatile Elements

The radioactive releases from the Fukushima Daiichi nuclear power plant consisted of (a) radioactive noble gases such as krypton and xenon, (b) volatile elements such as iodine, cesium, and tellurium, and (c) less volatile elements such as strontium. Based on previous accidents such as Three-Mile Island and Chernobyl, the activity of the noble-gases was expected to be orders-of-magnitude greater than the volatile-element activity, and the activity of refractory elements such as plutonium was expected to be orders-of-magnitude smaller (Imanaka 2012.) These expectations were confirmed by measurements in Japan and throughout the world; refer to the special issue of the Health Physics journal, volume 102, No. 5 (Health Physics 2012.) According to Table 1 of Sugiyama 2012, the xenon-133 activity was two to three orders of magnitude greater than the cesium-134 and cesium-137 activities. And according to Shanks 2012 (page 532) the ratios of cesium-137 to strontium-89 to strontium-90 were 700 to 175 to 1. Measurements and models agree that the ratio of cesium-134 to cesium-137 was approximately 1 to 1 (Musolino 2012, Imanaka 2012.)

Noble gases are not collected by filters or charcoal cartridges. However, their gamma emissions are detected by the NEWNET pressurized ion chambers. These data are discussed below and are available at http://newnet.lanl.gov .

Iodine-131 (I-131) is not optimally measured by our standard polypropylene filters; it is best measured using charcoal cartridges. So we supplemented the filter data with charcoal-cartridge data. These data, together with measurements of other radionuclides, are adequate for an assessment and assure us that radionuclides from Fukushima Daiichi did not present a threat to human health at or near Los Alamos.

Radioactive materials were released during many separate events during the weeks following the March-11 disaster. The largest releases probably occurred on March 15-16. The expected plume (Figure

1) was calculated by the Comprehensive Nuclear Test Ban Treaty Organization http://www.ctbto.org/verification-regime/the-11-march-japan-disaster/ and published in the New York Times on March 16, 2011. Similar plumes are shown in Figure 2 of Sugiyama 2012. These calculations predicted that the plume would arrive in California on March 18, 2011 and in New Mexico soon after.

These predictions are consistent with the NEWNET data (Figure 2), which show an increase of 0.1 micro-R/h, perhaps as early as March 19, followed by further increases over the next few days. NEWNET uses universal time, UTC or GMT, so the March-19 NEWNET data (day 9) correspond to 5 pm March 18 to 5 pm March 19, MST in New Mexico.

The predictions are also consistent with the air sampling data: fission products were first detected in the March 17-21 High-Volume samples (Tables 1-2), in the March 15-22 stack samples (Table 3), and in the March 15-29 AIRNET samples (Table 4).

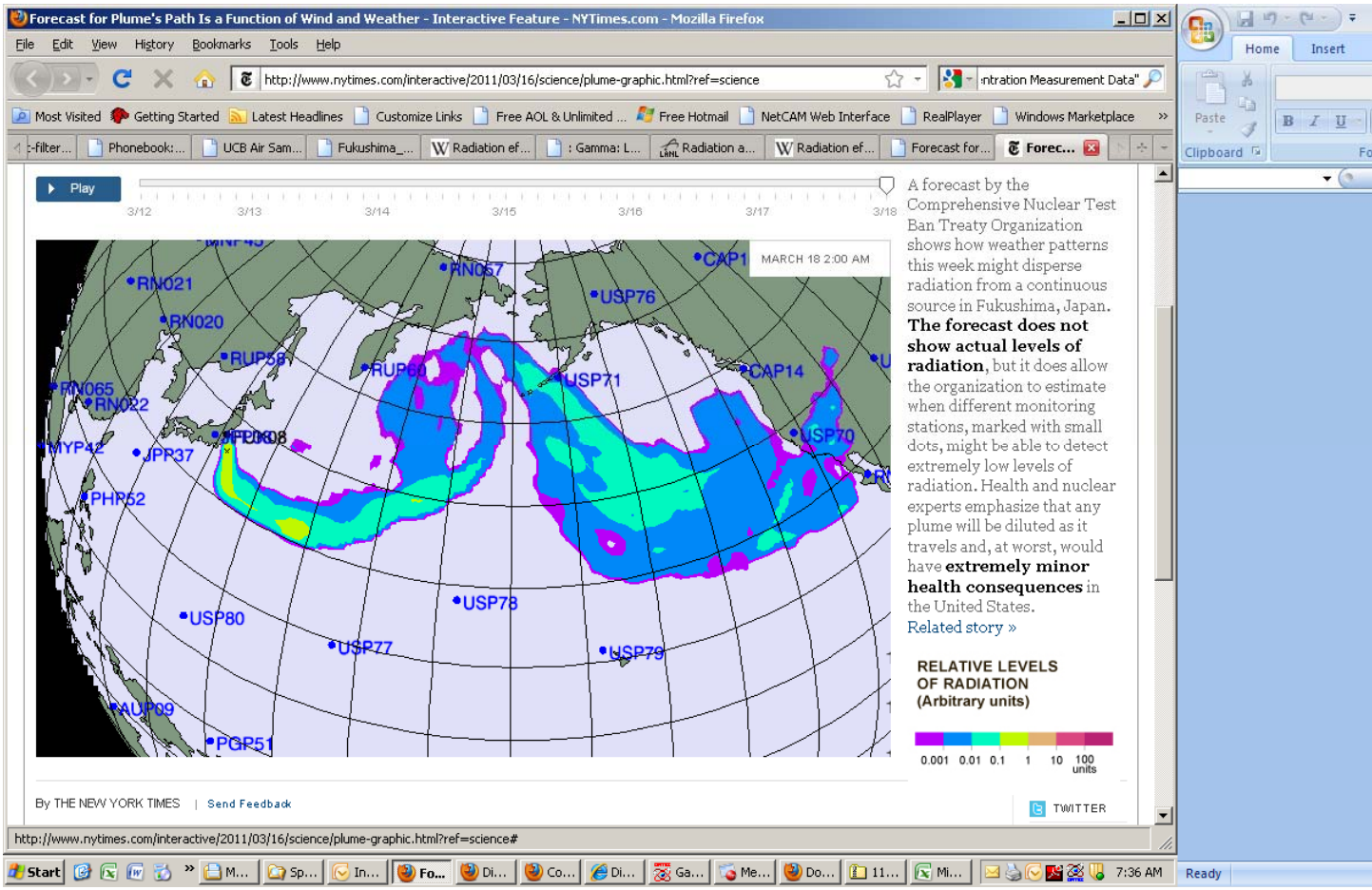

Figure 1. Predicted plume from Fukushima Daiichi, prepared by the Comprehensive Nuclear Test Ban Treaty Organization and published by the New York Times. It predicted the arrival of the plume in California on March 18, 2011.

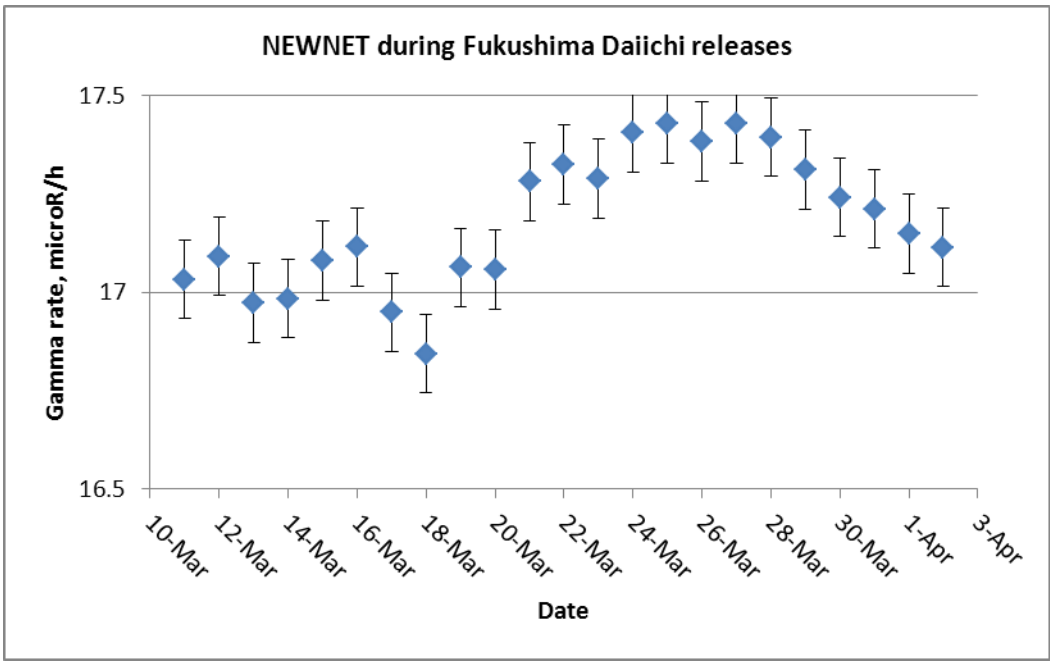

Figure 2. NEWNET data following the Japanese earthquake and tsunami. The error bars correspond to the normal fluctuations caused by radon concentrations that increase during atmospheric inversions and decrease during more turbulent conditions. The increased gamma rates from 21-March to 2-April are probably caused by fission products from Fukushima Daiichi.

High Volume Samplers

The locations of the three high-volume air samplers are listed in Table 1.

Station #	Location
#167	White Rock Old Fire Station, Los Alamos County, NM 87544
#173	LANL Technical Area 49 Gate, NM 87545
#211	Los Alamos Medical Center, NM 87544

Table 1. Locations of the three high-volume samplers within Los Alamos County.

These high-volume air samplers were operated according to standard procedures by the LANL Environmental Stewardship group, ENV-ES. The samplers use standard polypropylene filters with a nominal flow rate of 40 cubic feet per minute (1.1 m^3/minute). The actual flow rates and times were measured: each run was typically 3 to 4 days and the volume for each run was approximately 5000 to 6000 m^3.

These filters were counted for 14 hours and analyzed according to standard procedures by the Health Physics Analysis Laboratory of LANL group RP-2. The data were corrected for decay from the middle of the 3- or 4-day sampling period.

Station #	Date Collected	Radionuclide and Concentration (pCi/m^3)						
		Cs-134	Cs-136	Cs-137	I-131	I-132	Te-132	Te129m
all	3/17/2011	<	<	<	<	<	<	<
#167	3/21/2011	0.002	<	0.003	0.120	0.003	0.003	0.005
#173	3/21/2011	0.002	<	0.003	0.083	0.004	0.003	0.006
#211	3/21/2011	0.002	<	0.003	0.090	0.003	0.003	<
#167	3/24/2011	<	<	<	0.017	<	<	<
#173	3/24/2011	<	<	<	0.057	<	<	<
#211	3/24/2011	<	<	<	0.052	<	<	<
#167	3/28/2011	0.08	0.011	0.09	0.336	0.040	0.038	0.086
#173	3/28/2011	0.07	0.012	0.08	0.257	0.034	0.031	0.063
#211	3/28/2011	0.07	0.009	0.08	0.277	0.034	0.034	0.066
#167	3/31/2011	<	<	<	0.012	<	<	<
#173	3/31/2011	<	<	<	0.012	<	<	<
#211	3/31/2011	<	<	<	0.015	<	<	<
#167	4/4/2011	<	<	<	0.0048	<	<	<
#173	4/4/2011	<	<	<	0.0052	<	<	<
#211	4/4/2011	<	<	<	0.0050	<	<	<
#167	4/7/2011	<	<	<	0.0028	<	<	<
#173	4/7/2011	<	<	<	0.0029	<	<	<
#211	4/7/2011	<	<	<	0.0033	<	<	<
#167	4/11/2011	<	<	<	0.0015	<	<	<
#173	4/11/2011	<	<	<	0.0018	<	<	<
#211	4/11/2011	<	<	<	0.0016	<	<	<
#167	4/14/2011	<	<	<	0.0021	<	<	<
#173	4/14/2011	<	<	<	0.0030	<	<	<
#211	4/14/2011	<	<	<	0.0018	<	<	<
#167	4/18/2011	<	<	<	0.0062	<	<	<
#173	4/18/2011	<	<	<	0.0050	<	<	<
#211	4/18/2011	<	<	<	0.0044	<	<	<
all	4/21/2011	<	<	<	<	<	<	<
all	4/25/2011	<	<	<	<	<	<	<

Table 2. Concentrations (pCi/m^3) of various radionuclides by sampler number and the date the sample was collected. Typical uncertainty is indicated by the agreement among the three samplers, which is about 20%. Data are decay-corrected to the middle of the sampling period. The < symbol means no statistically significant concentrations were observed.

Data Quality

The data are robust and internally consistent. In total, 37 gamma peaks from fission products were observed: 8 from Cs-134, 9 from Cs-136, 1 from Cs-137, 7 from I-131, 8 from I-132, 2 from Te-129m, and 2 from Te-132. All significant peaks were identified so there were no other measurable radionuclides. For example, part of one gamma spectrum from the high-volume filters is shown in Figure 3.

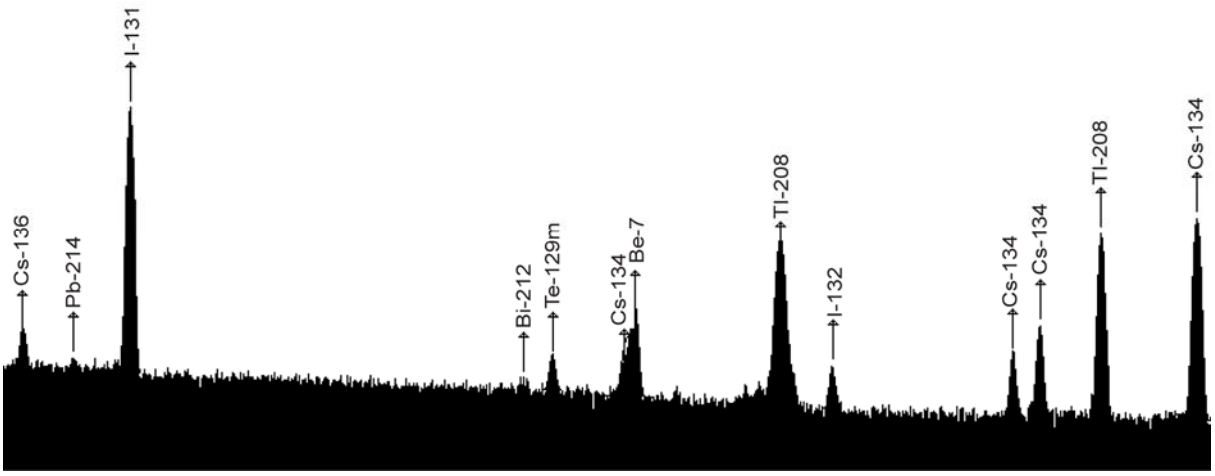

Figure 3. Part of a gamma spectrum showing peaks from fission products. The Cs-136 peak at the left is at 340.5 keV and the Cs-134 peak at the right is at 604.7 keV. The largest peak is the 364.5-keV peak from I-131.

The relative concentrations were generally consistent with expectations. For example, I-132 was in secular equilibrium with Te-132 as expected, and the Cs-134 to Cs-137 ratio was approximately 1 to 1 as expected.

In addition to the gamma peaks, an interesting feature was the presence of X-rays from about 28 to 37 keV, corresponding to the expected X-rays from the decay of tellurium, iodine, and cesium.

The data from the three High-Volume stations are consistent; the average standard deviation for each set of three samples is 20%. This indicates a uniform concentration in the vicinity of Los Alamos County.

Noble Gases, Vapors, and Volatile Elements

The predominant materials released from a nuclear reactor accident tend to be noble gases such as krypton and xenon, and volatile elements such as iodine, tellurium, and cesium. Based on past experience (Chernobyl and Three Mile Island) the activities of noble gas are likely to be more than an order of magnitude greater than those of volatile elements, and refractory elements such as plutonium are likely to be orders of magnitude smaller. These expectations are confirmed by the measurements reported in the Special Issue of the Health Physics journal, Health Physics 2012, and also in Imanaka 2012.

Noble gases are not collected on filters, and were not detected on any of the filter media. However, their gamma emissions were probably detected by all the NEWNET stations, as shown in Figure 2. NEWNET demonstrates that the dose rate from noble gases was no more than 0.3 micro-rem/hour, which amounts to less than 0.1 mrem over 2 weeks.

Vapors such as iodine are not efficiently collected by the polypropylene filters used with the AIRNET, Rad-NESHAP, and Hi-Volume samplers, so for vapors, the filter data are supplemented by charcoal-cartridge data, as follows.

Charcoal Cartridges

We supplement the filter data reported above with data from charcoal-cartridges deployed according to standard procedures by the Rad-NESHAP team (ENV-ES). The charcoal cartridges are preceded by HEPA filters and/or glass-fiber particulate filters so they measure only radio-iodine in the vapor phase. TA-48-1-07 also includes a charcoal bed upstream of the charcoal cartridge so these data were excluded.

Each sample was collected for one week at a flow rate of 2 cubic feet per minute, so the air volume through each cartridge was 571 m^3. The collection efficiency of the charcoal cartridge is 85%; for the largest I-131 peak, the gamma emission probability is 82%; detection efficiency is 5.1%; and each sample was counted for 3600 s.

We demonstrate the calculation using the 110322 data (March 22-29) as an example.

The net counts in the 364-keV peak for the ES-3, ES-2, and the duplicate sample, were obtained from the data packages, as follows.

ES-3: 1087 counts
ES-2: 832 counts
ES-2 duplicate: 956 counts

The average of these is 958 counts, with a standard deviation of 13%.

The sample activity (Bq) is calculated using the charcoal collection efficiency (0.85), gamma emission probability (0.82), detector efficiency (0.051), and count time (3600 s).

$$\text{Sample activity} = 958/(0.85 \times 0.82 \times 0.051 \times 3600) = 7.49 \text{ Bq}$$

Divide by 0.037 Bq/pCi to convert from Bq to pCi:

$$(7.49 \text{ Bq}) / (0.037 \text{ Bq/pCi}) = 202 \text{ pCi}$$

Divide by the volume (571 m^3) to calculate the concentration.

$$\text{Concentration} = (202 \text{ pCi}) / (571 \text{ m}^3) = 0.35 \text{ pCi/m}^3.$$

Samples were collected for 7 days and counted for 3600 s, 1.2 days after collection. The half life of I-131 is 8.0 days so we include a decay correction factor for half a week plus 1.2 days: 1.2 + 7/2 = 4.7 days. $2^{4.7/8} = 1.50$.

$$\text{Decay corrected concentration} = 1.50 \times 0.35 = 0.53 \text{ pCi/m}^3.$$

The charcoal-cartridge data are summarized in Table 3, below.

File	Start Date	End Date	Net Count in 364-keV peak	std dev	I-131 Concentration (pCi/m^3)	I-131 Decay-corrected concentration (pCi/m^3)
110315	3/15	3/22	422	6%	0.16	0.24
110317	3/17	3/24	265	21%	0.10	0.15
110322	3/22	3/29	958	13%	0.35	0.53
110324	3/24	3/31	510	15%	0.19	0.29
110331	3/31	4/7	37	37%	0.01	0.02

Table 3. Decay-corrected I-131 Concentrations (pCi/m^3) as measured by charcoal cartridges.

Notice that the Table-3 data are higher than the Table 2 data for I-131. This indicates that most of the iodine was in a vapor phase that passed through the polypropylene filters of the High-Volume samplers, and also passed through the filters upstream of the charcoal filters. In principle, we sum the Table-2 and Table-3 data.

Filters

There are also data from the stack filters. Interpretation of these data is difficult because the stacks are designed to measure the emissions from LANL facilities, and the inlet to the facilities is often filtered, so the stack samplers do not necessarily measure the outdoor air concentrations. However, in some cases, the stack fan was off during March so outdoor air diffused from the top of the stack into the air sampler, thus providing some indications of ambient air concentrations. Indications of Cs-137 are consistent with the ambient air measurements of AIRNET, described below.

These filters were analyzed for strontium using the standard method of chemical separation followed by beta counting. However, this method does not discriminate between Sr-89 and Sr-90, so although they were reported as Sr-90, they include Sr-89. According to Shanks 2012, more than 99% of the radioactive strontium from Fukushima Daiichi was Sr-89. After correcting for decay, the LANL stack data indicate a two-week-average concentration of 0.01 pCi/m^3, and ratios of Sr-89 to Cs-137 and Cs-134 that are consistent with the Shanks 2012 data. Calculations for a boiling water reactor (BWR) support these estimates (NAP 1996.) Thus, we conclude that the data reported as Sr-90 were mostly Sr-89 from Fukushima Daiichi.

AIRNET

We also have extensive data collected by more than 60 AIRNET samplers. Samples were collected for 14 days and counted 11 days later so the decay time varies from 11 to (11+14) days. The I-131 data in Table 4 have been multiplied by 4.8 to correct for an average of 18 days of decay: $2^{18/8} = 4.8$. For the 110328 data (March 15-29), this is conservative because the largest concentrations occurred in the later part of the two-week sampling period.

File	Start Date	End Date	std dev	Cs-134	Cs-137	I-131 Concentration (pCi/m^3)	I-131 Decay-corrected concentration (pCi/m^3)
110314	3/1	3/15	<	<	<	<	<
110328	3/15	3/29	22%	0.0175	0.0207	0.0319	0.1
110411	3/29	4/12	21%	0.0001	0.0002	0.0005	0.002
110425	4/12	4/26	29%	0.0002	0.0002	0.0001	0.0004
110509	3/26	5/10	<	<	<	<	<

Table 4. AIRNET measurements of fission products. Decay corrections for Cs-134 and Cs-137 are negligible and have not been applied.

The samples were counted for 11 hours and analyzed for the standard gamma suite of radionuclides that includes Cs-134, Cs-137, and I-131. The results are generally consistent with those reported in Table 2, though the ratios of I-131 to Cs-134 and Cs-137 indicate that the decay correction for 110328 (March

15-29) is conservative. The AIRNET data are internally consistent with a standard deviation of about 25%, indicating similar concentrations throughout the region.

In addition, many other peaks are visible in the 110328 AIRNET data but were not converted to concentrations. For example, the 110328 spectra show 7 peaks for I-131, 8 for Cs-134, 9 for Cs-136, 2 for Te-132, 4 for I-132, and 2 for Te129m.

Human Health

To estimate the dose, we use the EPA standards listed in Appendix E, Table 2, of 40 CFR 61. For I-131, we use the sum of the filter data and the cartridge data. For Te-132 we use the sum of I-132 and Te-132. The results are listed in Table 4.

Radionuclide	EPA Standard (pCi/m^3)	Dose (mrem)
Cs-134	0.027	0.3
Cs-136	0.53	0.01
Cs-137	0.019	0.5
I-131	0.21	0.9
Sr-89	1.8	0.003
Te-132	0.71	0.01
Te-129m	0.14	0.06
Total		1.8

Table 4. Total dose (mrem) from each radionuclide.

These doses are much smaller than the annual dose received from natural background radiation (approximately 500 mrem/year in Los Alamos) and are not expected to result in measurable health effects.

Conclusion

The data demonstrate the capabilities of the Los Alamos National Laboratory air-monitoring systems. Concentrations as small as 0.001 pCi/m^3, corresponding to doses as small as 0.001 mrem, were clearly observed. Results were consistent among the various air-samplers located throughout the region, demonstrating uniform concentrations and robust data.

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