PROGRESS REPORT

**DETERMINATION OF ATMOSPHERIC CONCENTRATIONS
OF XENON RADIOISOTOPES**

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Determination of Atmospheric Concentrations of Xenon Radioisotopes

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Determination of radioactive xenon concentrations in the atmosphere over a two year period has been performed as part of a research program to develop real-time measurement capabilities. The initial measurements were made to develop, prove, and validate our technical approach, while the longer-term measurements are being undertaken to establish natural background concentrations and variability with time. The results reported were made using noble gas fraction (typically 90% Kr and 10% Xe by weight) gas samples obtained from a commercial air-reduction plant in the north-eastern U. S. over a two-year interval beginning in the fall of 1993. The concentrated gas samples were typically obtained during a 6-8 hour interval at the commercial reduction plant and were shipped overnight to our laboratory. Analysis was typically completed approximately 24 hours after sampling. The analytical separation process typically took approximately 6 hours and gamma-ray spectrometric measurements were conducted for intervals ranging from 3 to 16 hours.

The technical approach involved removal of potentially interfering radon daughter radionuclides using a molecular sieve at room temperature, followed by cryogenic concentration of noble gases using a chilled ($-76^{\circ}\text{C}.$) activated carbon molecular sieve. During initial measurements both molecular sieve materials were contained in 30' x 1/4" gas chromatography columns for analytical separations. Krypton was separated from Xenon during the analytical procedure by warming the activated carbon molecular sieve to room temperature after initial noble gas concentration and actively pumping it away. Xenon-133 adsorbed to the activated charcoal molecular sieve was then quantified via its 81 keV gamma-ray using initially a p-type

intrinsic germanium detector and later a higher efficiency (64% relative to a 3" x 3" sodium iodide) n-type intrinsic germanium detector. Initially, Xenon-133 was analyzed directly on the separation column, but during later measurements, Xenon was transferred to a smaller mass of activated carbon molecular sieve. Separations conducted in November, 1993 utilized a spiral coil of approximately 4" in diameter; while separations during 1995 employed a cylindrical cell with a thin stainless foil window (1.5 mil), in order to obtain a higher efficiency cylinder for gamma-ray spectrometric measurements.

The atmospheric concentrations of ^{133}Xe observed in October and November, 1993 are shown in Table 1 and data from the current year are shown in Table 2. Figure 1 illustrates the radio-xenon concentration versus date observed to date in 1995. The data are tabulated in disintegrations per minute per cubic meter of air ($\text{d}/\text{m}/\text{m}^3$) and in milli-Becquerels per Standard Cubic Meter of air (mBq/SCM). Also listed at the bottom of the tables are the average concentration observed over the sampling intervals and the standard deviation of the data. The data in the tables are a factor of 4 lower than those reported by Bernstrom & De Geer (1983) for air sampled in Stockholm, which averaged $8.6 \text{ mBq}/\text{SCM}$, and an order of magnitude lower than air sampled at Ljungbyhed, which averaged $23 \text{ mBq}/\text{SCM}$. These authors cited significantly higher values for U. S. locations of $100 \text{ mBq}/\text{SCM}$ in 1975 (Kunz and Paperiello, 1976) and approximately $200 \text{ mBq}/\text{SCM}$ in 1981. The average for our data over the two-month interval, $2.0 \text{ mBq}/\text{SCM}$, is a factor of 50 lower than the 1975 average of $100 \text{ mBq}/\text{SCM}$ cited for the U. S.

TABLE 1. Xenon 133 Concentrations in Air as Determined by Cryogenic Separation and Gamma-Ray Spectrometry During 1993

<u>Sample Date</u>	<u>d/m/m³</u>	<u>mBq/SCM</u>
10/14/93	0.23	3.83
10/15/93	0.24	4.00
10/18/93	0.08	1.33
10/21/93	0.17	2.83
10/25/93	0.15	2.50
10/27/93	0.08	1.33
10/28/93	0.12	2.00
10/30/93	0.07	1.17
11/01/93	0.07	1.17
11/03/93	0.11	1.83
11/04/93	0.16	2.67
11/06/93	0.03	0.50
11/08/93	0.07	1.17
11/08/93	0.06	1.00
11/12/93	0.12	2.00
Average	0.12	1.96
Standard Deviation	0.06	1.03

TABLE 2. Xenon 133 Concentrations in Air as Determined by Cryogenic Separation and Gamma-Ray Spectrometry During 1995

<u>Sample Date</u>	<u>d/m/m³</u>	<u>mBq/SCM</u>
1/11/95	0.03	0.47
1/18/95	0.09	1.51
1/29/95	0.09	1.58
2/07/95	0.14	2.27
2/16/95	0.05	0.87
2/27/95	0.09	1.52
3/09/95	0.22	3.73
3/16/95	0.33	5.43
3/31/95	0.32	5.36
4/13/95	0.10	1.73
4/20/95	0.09	1.46
4/26/95	0.07	1.19
5/05/95	0.27	4.45
5/10/95	0.51	8.48
5/23/95	0.24	3.96
5/24/95	0.08	1.36
5/25/95	0.06	0.96
5/26/95	0.09	1.44
5/27/95	0.11	1.91
6/06/95	0.09	1.46
6/13/95	0.37	6.10
6/20/95	0.19	3.13
6/28/95	0.08	1.41
Average	0.16	2.69
Standard Deviation	0.12	2.05

Figure 1
 ^{133}Xe concentration measured in 1993.

^{133}Xe Background in the Northeast Sector of the US (1993)

Figure 2
 ^{133}Xe concentration measured in 1995.

^{133}Xe Background in the Northeast Sector of the US (1995)

REFERENCES

Bernstrom, B., and L. De Geer. 1983. Measurement of Small Amounts of Xenon-133 in Air. Defense Research Agency, Stockholm, Sweden.

Kunz, C. O., and C. J. Paperiello. 1976. "Xenon-133: Ambient Activity from Nuclear Power Situations." Science. Vol.192, p1235.

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