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Observation of ^{135}Xe with the PNNL ARSA System

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SUMMARY

The automated radioxenon sampler-analyzer (ARSA) developed by PNNL and with funding and support from the DOE NN-20 CTBT research and development program, observed 9.1-hr ^{135}Xe in a sample of New York City air obtained on April 4th, 1997. The report below briefly describes the ARSA system and the first ever reported measurement of the short-lived ^{135}Xe isotope from an automated radioxenon system.

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Introduction

The ARSA system is an autonomous radioxenon sampler-analyzer capable of measuring the radioxenon content of the air several times per day, storing the sample, and automatically sending the data to an appropriate data center¹. Currently, the ARSA is undergoing field-testing at the Environmental Measurements Laboratory (EML) for future use as part of an International Monitoring System (IMS) for the Comprehensive Nuclear test Ban Treaty (CTBT).

The ARSA automatically samples air continuously and measures the radioxenon component of the gas very soon after the sample has been purified. The rapid collection and analysis (nuclear counting begins about 10 hrs after the sample has been collected) of the samples allows for measurement of the shorter-lived radioxenon species that may be present in measurable levels near nuclear power plants or after a nuclear detonation. The nuclear counting system employed in the ARSA is a beta-gamma coincidence spectrometer developed at PNNL and is described in detail elsewhere².

Even though ^{133}Xe and ^{135}Xe activity levels near our testing site in New York City are expected³ to be detectable, ^{135}Xe activity levels from an automated xenon sampler-analyzer have never been reported. This may be due to the fact the short half-life of the isotope previously made it impossible for either automated or manual systems to collect and start analysis of the sample quickly enough to measure it. Also, other than the ARSA, there are no operating fully automated xenon samplers reported in the open literature. During routine testing of the ARSA at EML, located in New York City, we observed a detectable signal from ^{135}Xe .

Beta-Gamma Coincidence Data

The beta-gamma coincidence spectrometer used in the ARSA² produces a pair of beta-gated gamma-ray energy spectra three times per day from its dual crystal NaI(Tl)-based spectrometer.

Gamma spectra for the sum of the left and right NaI crystals for this sample are shown in figure 1 for April 4th 1997. The beta-gated gamma spectrum clearly shows the expected two peaks from ^{133}Xe at 31 keV and 81 keV. The peak centered at channel 304 corresponds to an energy of 250 ± 3 keV in close agreement with the known peak from ^{135}Xe at 249.8 keV.

The ARSA system allows gamma spectra to be gated by a window on low-amplitude beta pulses as well as by all beta pulses. The spectra gated by the low-amplitude beta pulses are enhanced in ^{133}Xe relative to background events which tend to have large amplitude beta pulses. The beta spectrum associated with the 249.8 keV peak has an endpoint energy of 900 keV which is much greater than the endpoint energy of 346 keV for the beta spectrum from ^{133}Xe . The relative abundance of the 250-keV peak in the spectrum gated by low-amplitude betas was compared to its relative abundance in

the spectrum gated by all betas. The 250-keV peak was found to be enhanced in the spectrum gated by high-amplitude beta pulses which is consistent with its assignment to ^{135}Xe .

The concentrations of ^{133}Xe and ^{135}Xe were calculated based on the current best estimates of various efficiency factors. The counts in a given peak were determined as the net area under a peak after correcting for tails above and below the peak in the beta-gated spectra for the sum of the two NaI crystals. The concentrations were calculated by dividing the net counts by the following factors and are reported in units of milli-Becquerels per cubic meter of air (mBq/m³).

For the 31-keV peak (assumed to be entirely due to ^{133}Xe):

Net counts	= 5504
Counting time	= 58,560 s
Beta efficiency (31)	= 0.75
Gamma eff. (31)	= 0.64
Abundance (31)	= 0.4894
Chemical yield	= 0.5 (estimate)
Vol. of air sample	= 25 m ³

$$\text{Concentration } ^{133}\text{Xe} = 32.1 \text{ mBq/m}^3$$

For the 81-keV peak - ^{133}Xe :

Net counts	= 3487
Counting time	= 58,560 s
Beta efficiency (81)	= 0.50
Gamma eff. (81)	= 0.80
Abundance (81)	= 0.37
Chemical yield	= 0.5 (estimate)
Vol. of air sample	= 25 m ³

$$\text{Concentration } ^{133}\text{Xe} = 32.2 \text{ mBq/m}^3$$

For the 250-keV peak - ^{135}Xe :

Net counts	= 459
Counting time	= 58,560 s
Beta efficiency (250)	= 0.75 (estimate)
Gamma eff. (250)	= 0.80 (estimate)
Abundance (250)	= 0.90
Chemical yield	= 0.5 (estimate)
Vol. of air sample	= 25 m ³

$$(\text{Concentration } ^{135}\text{Xe} = 1.16 \text{ mBq/m}^3)$$

Because of the 9.1-hr half-life ^{135}Xe , the concentration of ^{135}Xe must be corrected for decay during the 16.3-hr counting time. Dividing by the correction factor,

$$(1 - e^{-\lambda t})/\lambda t = 0.573,$$

gives 2.03 mBq/m^3 at the beginning of the counting period. An additional correction ($e^{-\lambda t} = 0.543$) is needed for decay of the sample between the end of collection and the beginning of the count (8 hr). At the end of the sample collection, the concentration of ^{135}Xe was 3.7 mBq/m^3 . The equivalent decay corrections for ^{133}Xe are much smaller ($<5\%$) and have not been applied here.

If the observed Xe fission products are produced by leaks from an operating nuclear reactor, one might expect that the relative amounts of ^{133}Xe and ^{135}Xe are given by their respective cumulative fission yields, but due mainly to burnup of ^{135}Xe , one would expect⁴ the ^{135}Xe level to be somewhat lower. Assuming a thermal or fission spectrum neutron fission of ^{235}U , the cumulative fission yields, and normal burn-up of ^{135}Xe in a pressurized water reactor (PWR) in equilibrium, the expected ratio $^{135}\text{Xe}/^{133}\text{Xe}$ at $t=0$ is 0.37. The experimentally observed ratio of $^{135}\text{Xe}/^{133}\text{Xe} = 3.7/32 = 0.12$ implies that the ^{135}Xe has decayed by about 1.4 half-lives while traveling from the source to the sampling station. Other complications such as hold-up at the source after release or variable concentrations during the 8-hr sampling period were not considered here.

Figure 2 is an example of background gamma spectra taken during the 24 hours prior to the sample shown in figure 1. Note that the background does show some residual ^{133}Xe from previous samples in the cell. Tests at PNNL have shown that about 5% of a Xe sample remains in the cell after pumping the cell to remove the sample. However, note that there is no evidence of a background peak in the vicinity of the 250-keV peak from ^{135}Xe .

Figure 1. Xe sample obtained from New York City air on April 2, 1997 showing ^{135}Xe peak (channel 304) in addition to ^{133}Xe peaks (channels 39 and 102). Top curve is ungated spectrum. Bottom curve is beta-gated spectrum. Both curves are sum of left and right NaI crystals.

Figure 2. Background spectra (sum of both gamma detectors) for cell 3. Top curve is ungated spectrum. Bottom curve is beta-gated spectrum. Note residual peaks from prior samples of ^{133}Xe and absence of ^{135}Xe peak in beta-gated spectrum.

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