# RADIONUCLIDE MEASURMENTS FOR NUCLEAR EXPLOSION MONITORING

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## ABSTRACT

The Automated Radioxenon Sampler/Analyzer (ARSA) and Radionuclide Aerosol Sampler/Analyzer (RASA) are radionuclide detection technologies developed by the Department of Energy (DOE)/National Nuclear Security Administration (NNSA) Program at the Pacific Northwest National Laboratory for monitoring for radionuclides in the atmosphere for signs of atmospheric and underground nuclear explosions.

In March 2001,the ARSA completed an 18-month field test in Freiburg, Germany, as part of the International Noble Gas Experiment (INGE). During this test, the ARSA

- made over 1,200 automated measurements of $^{131m}$Xe, $^{133}$Xe, $^{133m}$Xe, and $^{135}$Xe,
- had an up time of approximately 95%,and
- achieved a minimum detectable concentration of about 0.10 to 0.15 mBq/m$^3$ 3 times per day for $^{133}$Xe.

By measurement of the relative concentrations of the xenon isotopes, we were able to determine with high confidence that reactors were the source of the radioxenon observed during this test.

Analysis of nearly 1,700 RASA samples has shown that $^7$Be is the only radionuclide with good quality-assurance properties likely to be detected at an International Monitoring System (IMS) radionuclide analysis laboratory. Concentration of $^7$Be in ~70% of samples is adequate for an effective quality assurance program. The $^7$Be half-life, energy, and typical atmospheric concentrations allow precise and relevant follow-up laboratory measurements. These results highlight the importance of station-to-station calibration and emphasize the need for inter-laboratory comparison exercises.

**KEY WORDS:** ARSA, noble gas, RASA, $^7$Be, $^{133}$Xe, $^{135}$Xe, $^{133m}$Xe, radionuclide, aerosol, fission, isotope, atmospheric, Radionuclide Laboratory, International Monitoring System, quality

## OBJECTIVE

A key function of the radionuclide portion of the IMS includes monitoring the atmosphere for radionuclides emitted as fission or activation product aerosols or gases (radioxenons), since detection of appropriate isotopes can discriminate nuclear from non-nuclear events.

## RESEARCH ACCOMPLISHED

A first international demonstration of the ARSA xenon noble gas collection and analysis capabilities (Bowyer, 2001) and other xenon noble gas systems was performed in Freiburg, Germany, in 1999 and 2000. This paper describes the measurements obtained by the ARSA (Bowyer, 1999). Also, since 1998, the RASA system has been collecting and analyzing automated aerosol measurements in Richland, WA, USA to, among other things, determine useable radionuclides for quality assurance/quality control (QA/QC) monitoring for the IMS. The results of these measurements are shown below.

**Noble Gas Demonstration**

Starting in 1999, a noble gas equipment test was conducted to assess the state-of-the-art of the current equipment available for radioxenon measurement and to assess its suitability for the International Monitoring System (IMS) radionuclide network. In addition to the ARSA, three other technologies were selected for evaluation, including: the ARIX from the V.G. Klopkin Radium Institute in St. Petersburg, Russia, the SAUNA from the Defense Research Establishment — FOI 41, in Stockholm, Sweden, and the SPALAX from the Commissariat l'Energie Atomique, Direction des Applications Militaires, D partement Analyses Surveillance Environnement (CEA/DASE) in Paris, France. The test location at the Institute for Atmospheric Radioactivity (IAR) in Freiburg, Germany, has an average activity concentration of $^{133}$Xe, a few mBq/m_ (Stockburger, 1977) due its proximity to many operating nuclear reactors.

The ARSA system measures all of the relevant xenon isotopes from nuclear detonations; $^{131m}$Xe, $^{133}$Xe, $^{133m}$Xe, and $^{135}$Xe, by measuring the energies of both emitted particles (betas and gammas) from each decay (Reeder, 1998). The ARSA system measured all of the radioactive xenon concentrations reliably without operator intervention for nearly 18 months. The ARSA automatically measured over 1,200 atmospheric samples of radioactive xenon 3 times per day. In addition, several weeks were dedicated to manually injecting xenon isotopes into the air stream to measure isotopic ratios and for comparison to other xenon systems. The results obtained by the ARSA were compared to an independent analysis of the samples performed by IAR personnel from the xenon stored by the ARSA after measurement. There was a good agreement between the automated ARSA results (~10%), and the independent analysis (Bowyer, 2001).

The calculated up time for the ARSA was approximately 95% (Bowyer, 2001), defined as the total number of cycles that the equipment was not producing, collecting, and subsequently analyzing coincidence spectra divided by the total number of cycles available, minus time taken for spike tests, travel time of personnel, and delays from part availability or customs delays. It is notable that the system down time was mainly due to a single failure mode not repaired during the test. This failure mechanism was traced to a poor connection inside a line replaceable unit that was not replaced during the test due to lack of spares.

Xe-131m was measured occasionally at a few mBq/m$^3$ level with a few runs where there was nearly pure $^{131m}$Xe without accompanying high concentrations of $^{133}$Xe. Although this was not expected and more data are necessary, there are mechanisms that could account for a nearly pure (without $^{133}$Xe) $^{131m}$Xe. The possibilities include medical sources of radioxenon, and old reactor xenon that could have been held up at reactors before release, and hence the long-lived $^{131m}$Xe is the only radioxenon present.

Xe-133 was detected nearly every day during this test averaging 1 to 2 mBq/m$^3$, though levels exceeding 100 mBq/m$^3$ were observed with plume passage times of <10 hours (Bowyer, 2001). Figure 1 shows a plot of the measured $^{133}$Xe concentration in Freiburg for the last 12 months of the 18-month test. There are several locations on the plot where no data are shown, corresponding to down times of the equipment for spike tests, maintenance, or equipment failure. The high concentrations near the end of the test were measured after spike testing was completed, and are most likely due to radioxenon spikes being collected by the ARSA from the room. Other systems being tested in Freiburg also saw the elevated levels with substantial amounts of $^{133m}$Xe, indicating that the spikes were the cause of the elevated levels.

Xe-133m was not observed during most of the test, with the exception of the final week, which was probably due to the preparation of very high activity radioxenon spikes used in spike calibration testing (Bowyer, 2001).

Xe-135 was detected nearly a dozen times during the testing period at levels as high as 4 mBq/m$^3$. These concentrations of $^{135}$Xe were usually accompanied by $^{133}$Xe concentrations of several tens of mBq/m3, with one notable exception in which the $^{135}$Xe/$^{133}$Xe concentration ration was 2:1. This ratio is only expected following a nuclear detonation or during reactor startup. The $^{133m}$Xe/$^{133}$Xe activity ratio for this period was consistent with reactor startup.

Figure 2 shows the distribution of the concentration data collected in Freiburg during the year 2000. The distribution of concentrations shows an approximately lognormal shape. A three-parameter lognormal distribution,

$$f(x) = \frac{1}{(x-a)\sigma\sqrt{2\pi}} e^{-\frac{1}{2}\left[\frac{\ln(x-a)-\mu}{\sigma}\right]^2}$$

fits the data quite a bit better than the two-parameter lognormal function, which could be due to the fact that the minimum detectable concentration of $^{133}$Xe is near 0.1 to 0.15 mBq/m$^3$ (McIntyre, 1999), or close to the cut-off parameter, *a*, in the lognormal distribution.. This behavior is not inconsistent with other atmospheric dilution observations (Mage, 1974; Ott, 1995).

**Laboratory Quality Assurance Measurements**

Members of the international community have discussed the role of Radionuclide Laboratories for performing a network-wide quality assurance function. However, the details of this function have been lacking. The recent effort to determine the financial resources needed for the labs has driven an analysis of the nature and frequency of QA measurements. The work reported here is the analysis of aerosol samples in a scenario that represents the expected field and laboratory sample measurement sensitivity and timing. Timing is quite important, because most of the isotopes embedded in the aerosol filter are short-lived and cannot therefore survive the transit time of the aerosol samples to the laboratory.

To simulate realistic field-lab measurement timing, a RASA filter taken in Richland, WA, was allowed to decay for 10 days. Both the field and lab systems were counted for 1 day. The field system employed a 90% relative efficiency P-type detector while the lab system was an ultra-low background 40% P-type. Figure 3 shows the spectrum from the field measurement (the red or highest spectrum), the typical blank from the same RASA (in blue with diamonds at each point, or the second highest spectrum) and the lab system measurement of the same filter with 10 days decay (the green spectrum with triangles at each point, or the lowest spectrum). The filed measurement of the filter shows $^7$Be, a cosmic-ray product in the upper atmosphere, as well as many common natural isotopes ($^{212}$Bi, $^{212}$Pb, $^{214}$Bi, $^{214}$Pb, $^{208}$Tl, and $^{40}$K, for example). Natural isotopes are found in the detector background, but the $^7$Be is not. The $^7$Be is found in the lab measurement, but the natural isotopes are mostly not.

The conclusions that could be drawn from this figure are that natural isotopes will have decayed away by the time a lab measurement is made and thus have no constructive role in QA functions. Beryllium-7, whose half-life, T_ is 53.4 day, and which has a primary gamma-ray energy at 477.6 keV, may be the only isotope that is dependably present whose source is certainly on the filter and not the field or lab background. While it would be preferable to have several isotopes present in the lab measurement from ubiquitous airborne radioactivity, $^7$Be is not an altogether unfortunate isotope to be the only survivor since it has a good half-life and a primary gamma-ray energy reasonably close to that of $^{140}$Ba (primary gamma ray energy at 537.4 keV), the isotope used to act as the standard for field and lab system sensitivity.

To make effective comparisons between field measurements and lab measurements, the statistical uncertainty of measurable isotopes must be reasonable. To objectively assess the quality of the nuclide information in the spectra in Figure 3, all were subjected to an automatic analysis. The results in Table 1demonstrate that the only reliable isotope is $^7$Be and that the statistical uncertainty of the $^7$Be determination were less than 2%. (Systematic biases such as efficiency calibration errors have not been taken into account, but must eventually be controlled by the use of intra-lab calibration and careful certification of station efficiency.)

While the above example shows that reasonable uncertainties may be sometimes achieved, the question remains as to what fraction of the samples taken in the IMS network will have reasonable $^7$Be activity such that the statistical uncertainty in the field and the lab are similarly reasonable. To determine this fraction, approximately 1700 RASA daily samples were analyzed using an automated analysis code. The $^7$Be activity and the corresponding statistical uncertainty were tabulated and histogrammed on a frequency chart with a logarithmic x-axis. Figure 4 shows the fraction of the spectra with $^7$Be uncertainties in bins from 1% error to 100% error. The sum of the fractional bins from 1% to 100% is also shown. It can be seen that approximately 70% of the spectra have $^7$Be uncertainties below 3%.

## CONCLUSIONS AND RECOMMENDATIONS

Even though automated, high-sensitivity noble gas monitoring is relatively new, our recent demonstration has shown that reliable long-term operation of a system to collect and analyze the xenon isotopes $^{131m}$Xe, $^{133}$Xe, $^{133m}$Xe, and $^{135}$Xe is now possible. Recent data collected are consistent with normal reactor operations during start-ups and at equilibrium; however, more data should be collected to characterize radioxenon concentrations in parts of the world where radioxenon levels have never been measured. These data are important to determine the background that will be encountered for radioxenon monitoring on a world-wide scale.

In addition, it has been shown that $^{7}$Be will be the major focus of the radionuclide lab QA function, due to the lack of other reasonably long-lived and reasonably active isotopes in the atmosphere. The uncontrolled quantities of other natural backgrounds preclude the use of typical natural isotopes, even if they have not totally decayed by the time of the lab measurement. This leads to the final conclusion that routine samples collected from IMS stations should be quite simple to analyze presuming the backgrounds of the lab measurement system are sufficiently low.

## ACKNOWLEDGEMENTS

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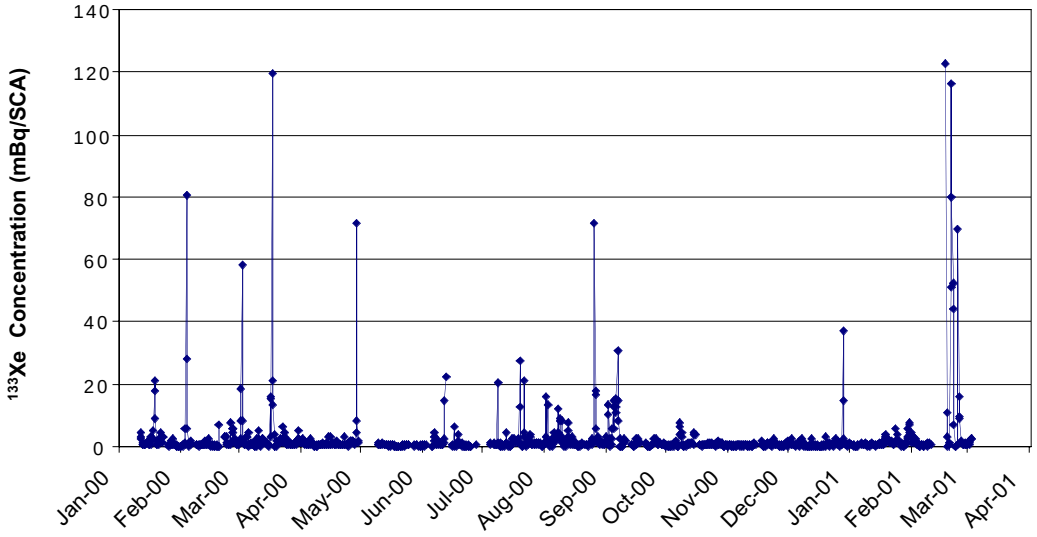


**Figure 1**. Plot of the concentration of $^{133}$Xe taken by the ARSA throughout CY 2000.

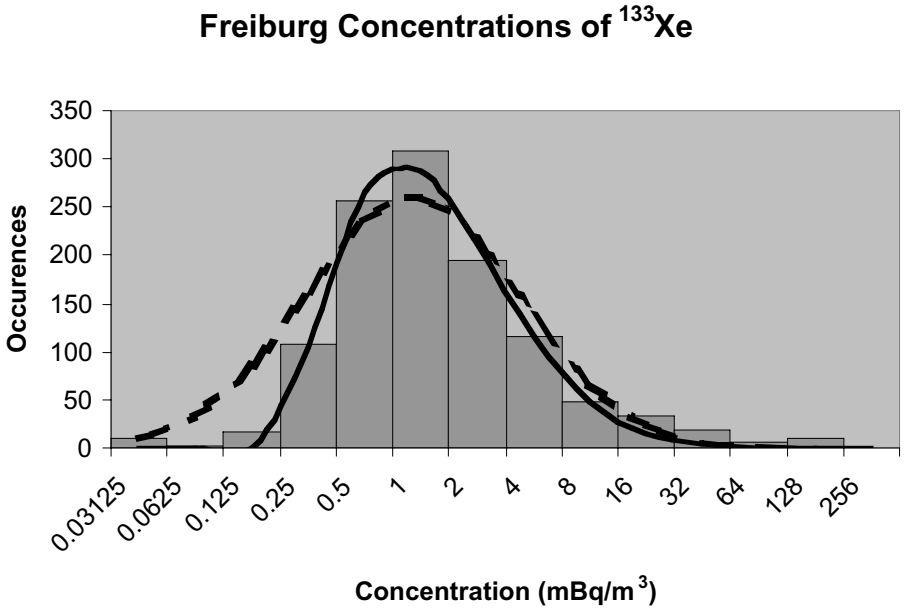


**Figure 2.** Distribution of the observed concentrations of $^{133}$Xe during the year 2000 and fits of this distribution to two different lognormal distributions. The bars show the number of occurrences of a given measured concentration, the dashed line is a fit of a two-parameter lognormal distribution, and the solid line is a fit of a three parameter lognormal distribution.

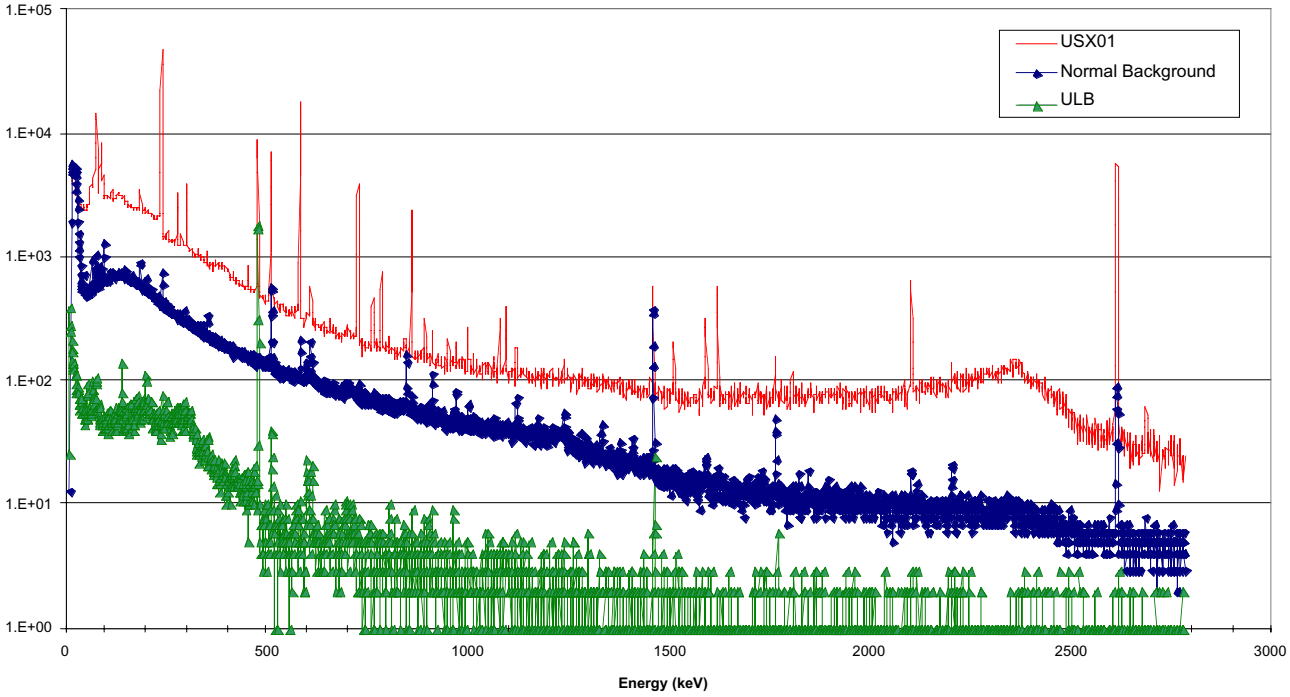


**Figure 3.** Gamma ray spectrum obtained under three conditions. A RASA field measurement (USX01) is shown in red, or is the highest spectrum. The detector background for that RASA is shown as the blue spectrum with diamonds at each point. The re-measurement of the same filter ten days later with an ultra-low background (ULB) detector is the green spectrum with triangles at each point, or is the lowest spectrum.

| ***Isotope*** | ***Station*** | ° | ° | ° | ***Lab*** | ° |
|---|---|---|---|---|---|---|
| | (Bq) | error | | | (Bq) | error |
| Be-7 | 31.72 | 1.32% | | | 27.85 | 1.88% |
| Pb-212 | 40.21 | 2.98% | | < | 0.2067 | |
| Bi-212 | 43.64 | 3.20% | | < | 2.163 | |
| K-40 | 7.054 | 3.37% | | | 1.101 | 17.12% |
| Tl-208 | 15.39 | 5.95% | | < | 0.07066 | |
| U-235 | 0.7126 | 6.05% | | < | 0.1106 | |
| U-238 | 20.62 | 13.34% | | < | 11.66 | |
| Ac-228 | 0.3922 | 15.19% | | < | 0.3593 | |
| Pb-214 | 0.2822 | 18.55% | | < | 0.03777 | |
| Bi-214 | 0.534 | 20.42% | | < | 0.09837 | |

**Table 1.** The results of automatic analysis of the original RASA spectrum and the lab spectrum. The only positive detection isotopes in the lab were $^{7}$Be and $^{40}$K.

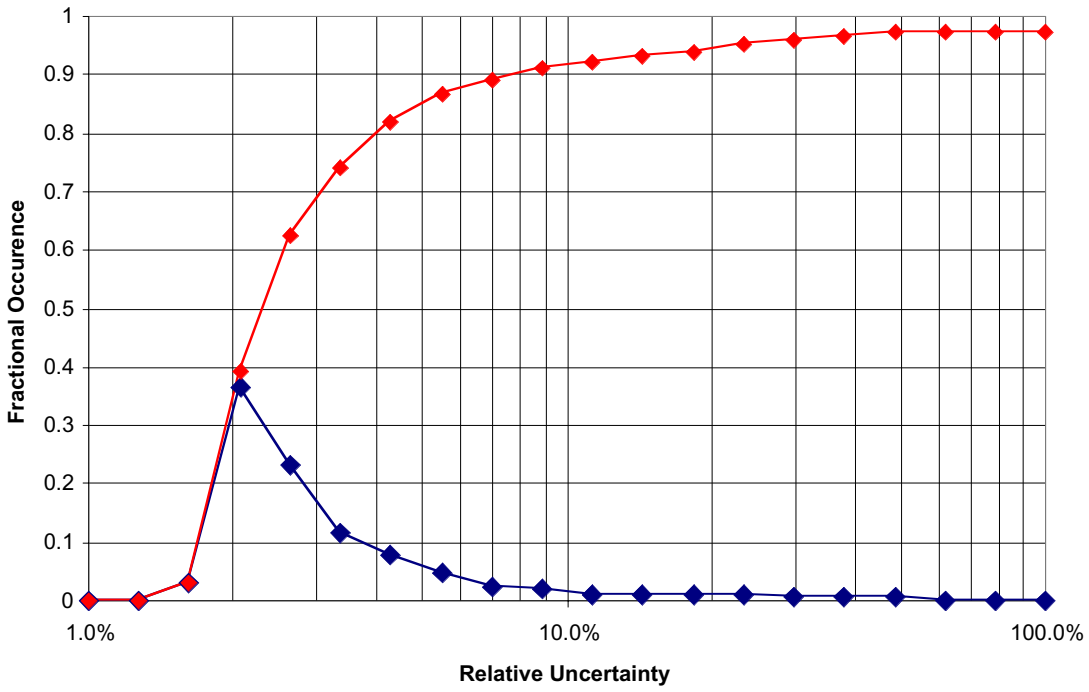


**Figure 4.** Frequency chart showing the fraction of 1700 RASA spectra with $^{7}$Be activity uncertainties in bins from 1% 10 100% uncertainty. The sum of the fractional occurrences from 1% to 100% is also shown. This demonstrates that about 70% of observed $^{7}$Be activities have a 3% uncertainty or less.