

RADIOXENON ATMOSPHERIC MEASUREMENTS IN NORTH LAS VEGAS, NV

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ABSTRACT

Pacific Northwest National Laboratory (PNNL) deployed the Automated Radioxenon Sampler/Analyzer (ARSA) in North Las Vegas for two weeks in February and March 2006 for the purpose of measuring the radioxenon background at a level of sensitivity much higher than previously obtained in the vicinity of the Nevada Test Site (NTS). The measurements establish what might be expected if future measurements were ever taken at the NTS itself and investigate improved methods of environmental monitoring of NTS. A second detector, the Portable Environmental Monitoring Station (PEMS), built and operated by the Desert Research Institute (DRI), was deployed in conjunction with the ARSA and contained a pressure ion chamber, aerosol collection filters, and meteorological sensors.

Some of the radioxenon measurements detected ^{133}Xe at levels up to 3 mBq/m³. This concentration of radioxenon is consistent with the observation of low levels of radioxenon emanating from distant nuclear reactors. Previous measurements in areas of high nuclear reactor concentration have shown similar results, but the western U.S., in general, does not have many nuclear reactors. Measurements of the wind direction indicate that the air carrying the radioxenon came from south of the detector and not from the NTS.

OBJECTIVES

Background

In the 1990s, PNNL developed the ARSA as a prototype sampling station for the International Monitoring System (IMS) (Bowyer et al., 2002). The IMS is a network of stations originally developed to monitor compliance with the Comprehensive Nuclear-Test-Ban Treaty (CTBT). Ultimately, the IMS will have at least 40 radioxenon sampling stations placed around the world (Protocol, 1996). Concurrent with ARSA's development, was the development of similar detectors by a few other countries: France, Russia, and Sweden. The automated nature of the detectors is crucial as several of the planned stations are to be at remote locations. The ARSA and its counterparts are much more sensitive than anything commercially available at the time of their development, with sensitivities as low as 0.1 mBq/m³ (Bowyer et al., 2002).

Radioxenon is in the mass range where large fission yields occur from uranium- or plutonium-based nuclear weapons. In a nuclear explosion, several radioxenon isotopes are produced with half-lives convenient for detection (short enough for high radioactivity, but long enough for detection): ¹³⁵Xe (9.14 hr), ¹³³Xe (5.25 d), ^{133m}Xe (2.19 d), and ^{131m}Xe (11.93 d). As an inert noble gas, xenon doesn't combine chemically with other elements (in ground or atmosphere) and is likely to escape even from an underground explosion. Nuclear reactor operations also release radioxenon, and there are some medical and industrial sources (Bowyer et al., 1998). ¹³³Xe is detected much more frequently than the other three radioxenon isotopes during normal atmospheric monitoring (Auer et al., 2004) so seeing other radioxenon isotopes is important for nuclear explosion monitoring.

Radioactive noble gas detection is internationally accepted, can accommodate significant stand-off distances, and is quite sensitive without being overly intrusive (i.e., revealing detailed information relative to weapon design). Given its importance in international monitoring and the fact that such sensitive noble gas measurements have not previously been performed at the Nevada Test Site (NTS), it was decided that measurements should be performed using the ARSA to establish a baseline of comparison to possible future NTS background measurements. There was also interest in the measurements for improved environmental monitoring purposes. Thus, it was decided to perform a pair of measurements with the ARSA, first near Las Vegas, 65 miles away from the NTS, and then later at the NTS's Mercury townsite.

Detector Systems

Two detectors were deployed: the ARSA and the PEMS.

The ARSA is about the size of two soda machines and weighs approximately 3000 pounds (see Figure 1). It takes in air at the rate of 144 m³/day (one 48 m³ sample each eight hours) from the outside and separates and concentrates the 87 ppb Xe present. The air is first compressed, filtered, cooled, and moved through dryers to remove the H₂O and CO₂. A series of adsorption charcoal traps are used to remove first the radon, and then the xenon from gases of even lower boiling points. These traps must be regenerated (outgassed using heat) after each sample. Pre-purified nitrogen is used as a carrier gas. Additional traps are used for further removal of contaminant gasses, though some radon still gets through and is the chief source of background (Bowyer et al., 1999). Radioxenon counting is performed using a beta-gamma coincidence cell based on a NaI(Tl) and plastic scintillator, which is surrounded by a lead cave to reduce background (McIntyre et al., 2001). After counting, a thermal conductivity detector (TCD) is used to quantify the amount of Xe in the sample. Archive bottles may be filled for later retrieval and lab analysis.

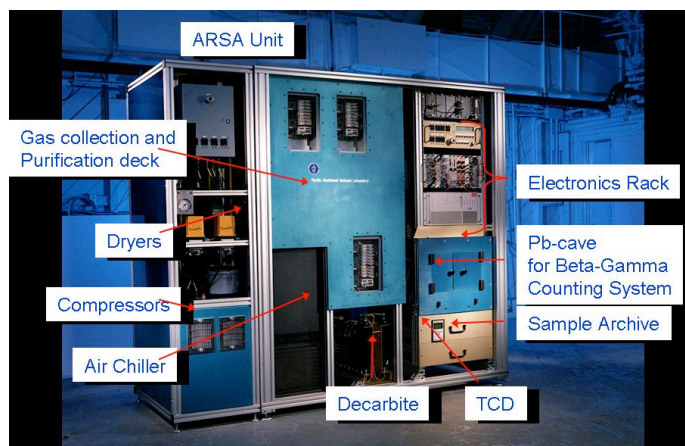


Figure 1. The ARSA system with various components labeled.

In order that ARSA be continuously monitoring the atmosphere, many of the above steps are performed with parallel subsystems. For example, there are four beta-gamma nuclear detector cells. Every 8 hours, a 24-hour count is completed for one sample. Before starting a new 24-hour sample count, an 8-hour “background” count is measured for a given cell for subsequent subtraction. This is necessary, in part, because some residual xenon and radon gas adheres to the cell walls, resulting in a “memory effect” (McIntyre et al., 2001). When operating properly, the ARSA has a sensitivity of approximately 0.1-0.2 mBq/m³ for ¹³³Xe.

In tandem with the ARSA, measurements were also made using the PEMS constructed and operated by DRI. The PEMS is modeled after the Community Environmental Monitoring Program (CEMP) stations operated by DRI around the NTS (CEMP, 2007). Figure 2 shows the PEMS detector as deployed for this measurement. The weather “tower” (on the left side of the picture) measured wind speed and direction, air temperature, humidity, barometric pressure, and precipitation. To the right in the picture, on a triangular stand, is a Reuter-Stokes pressurized ion chamber (PIC), capable of measuring gamma exposures of 2–800 μ R/hr. Rightmost on the PEMS trailer (shaped like a doghouse) is an air particulate sampler. The sampler collects particulate greater than 0.3 microns in size at a flow rate of 2.0 cubic feet per minute. Two samples of approximately one week each were taken, with the filters analyzed for gammas using well-shielded high-purity germanium (HPGe) detectors at PNNL.



Figure 2. The PEMS detector as deployed for this measurement.

RESEARCH ACCOMPLISHED

The detectors were deployed in the trailer parking lot south of the Rack Assembly Tower (bldg. A-14) at the Department of Energy's Nevada Site Office (DOE-NV) in North Las Vegas, NV, as pictured in Figure 3. The ARSA detector was transported from PNNL to North Las Vegas and deployed in a trailer (see Figure 4). In previous ARSA deployments, the detector was dismantled and shipped by container. Transport by road appeared to result in some minor damage to ARSA upon arrival, which PNNL staff were able to fix after some effort. Thus, the ARSA measurements reported are for March 1–7, 2006, when the ARSA was running successfully, while the PEMS measurements reported begin on February 22, 2006.

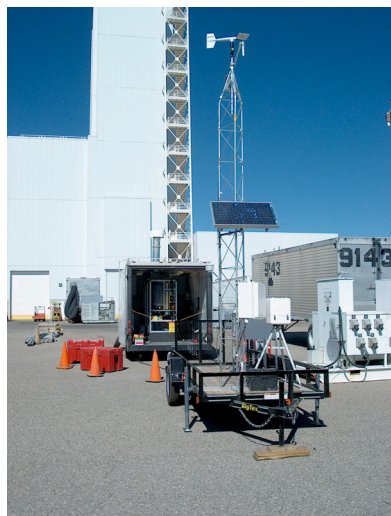


Figure 3. The PEMS (foreground) and ARSA detectors in front of the Rack Assembly Tower at DOE-NV.



Figure 4. The trailer ARSA was deployed in.

ARSA Measurements

Figure 5 shows the detected ^{133}Xe concentrations, including statistical errors for the time period. The times shown on the x-axis refer to the midpoint of the sample collection period in local time, which occurred at approximately 5 a.m., 1 p.m., and 9 p.m. each day. For example, the first measurement occurred at 5 a.m., March 1, and is plotted as 1.21 on the x-axis. There is a gap of four measurements between 9 p.m., March 1, and 1 p.m., March 3, due to an ARSA shutdown. The pink line shows the minimum detectable concentration (MDC) values, which are typically between 0.1 and 0.2 mBq/m³. The MDC rises at the end due to the memory effect of xenon from the earlier spike remaining in the detection cells. The other three xenon radioisotopes (^{135}Xe , $^{133\text{m}}\text{Xe}$, and $^{131\text{m}}\text{Xe}$) were not observed during this measurement campaign, and are thus not shown. The average MDC value for ^{135}Xe was 2.1 mBq/m³. Elevated levels of the other xenon radioisotopes in combination with ^{133}Xe are expected from nuclear explosions.

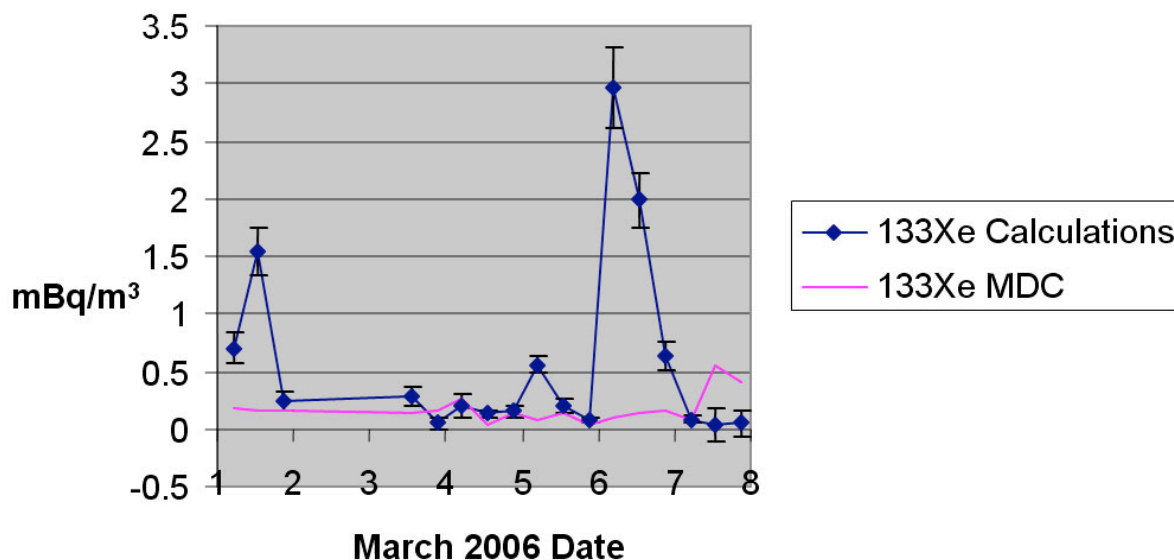


Figure 5. ^{133}Xe measurements by ARSA, beginning at 5:00 a.m., March 1, 2006.

^{133}Xe was clearly seen a few times at a relatively low level, with the largest spike (almost 3.0 mBq/m^3) occurring on March 6th. Previous, much-longer measurements at Freiburg, Germany (Auer et al., 2004), and Allentown, Pennsylvania (Bowyer et al., 1997), frequently measured values of $1\text{--}3 \text{ mBq/m}^3$. Those two locations were chosen for their proximity to many commercial nuclear reactors. Comparatively, the western United States has much fewer nuclear reactors. Measurements at PNNL, which is only 10 miles from the Columbia Nuclear Generating Station (though usually upwind), only observed radioxenon infrequently (McIntyre et al., 2001).

Nuclear reactors release radioxenon as a fission gas, either through cracks in fuel rods, or from neutron-induced fission in uranium on the outside of the fuel rods or in the cooling water (Auer et al., 2004). Especially large amounts may be released during core refueling (McIntyre et al., 2001), but the observed xenon in this measurement was not large (at Freiburg, ^{133}Xe concentrations up to 100 mBq/m^3 were observed [Auer et al., 2004]). Although reactors generally have some mechanism in place to trap released radioxenon (Bowyer et al., 1997), it is still the dominant source of released atmospheric radiation for nuclear reactors (Bowyer et al., 1998). Radioxenon is also used for some medical applications.

PEMS Measurements

Figure 6 displays the PIC data from the PEMS detector for the entire time deployment period. The values measured are consistent with those observed at the CEMP station located at DRI in Las Vegas, NV, approximately 10 miles away and available online (CEMP Station, 2007). Displayed are the ten-minute average measurements (in units of $\mu\text{R/hr}$) of the gamma activity made by the PIC. The measurements run from 8:30 a.m., February 22, through 8:30 a.m., March 9 (144 measurements per day, as shown on the x-axis). The diurnal variation is quite apparent. The chief source of this radiation is the natural radon background, and any xenon contribution is miniscule.

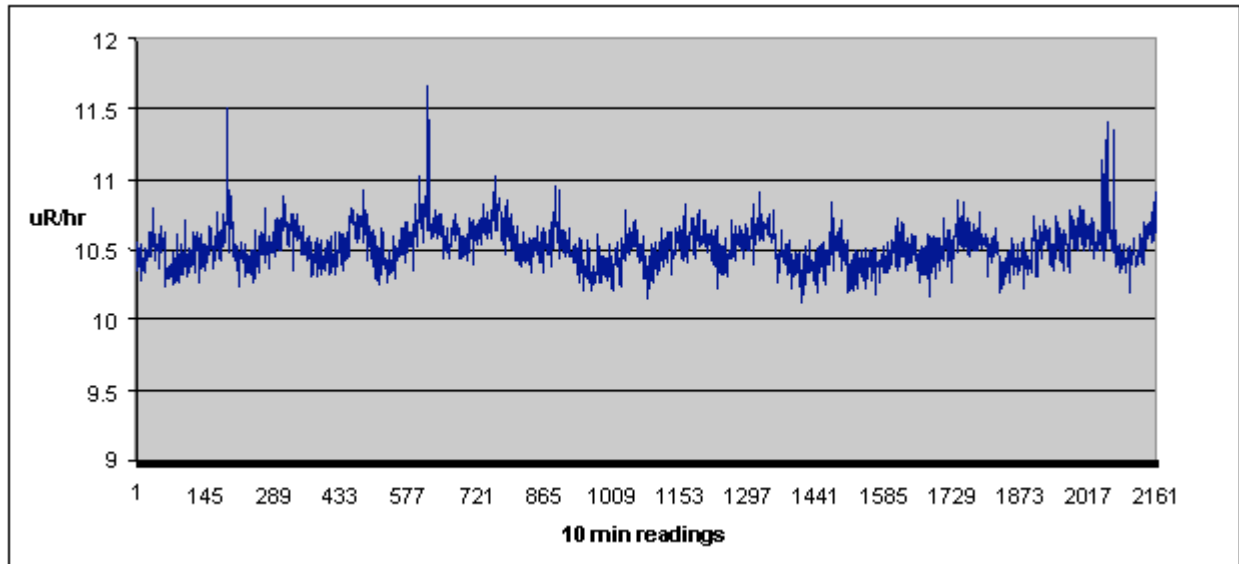


Figure 6. Ten-minute average PIC measurements by the PEMS, from 8:30 a.m., February 22, through 8:30 a.m., March 19, 2006, in north Las Vegas.

The filters collected by the PEMS were allowed to decay for one week (to reduce the radon contribution) and then measured for approximately two days using a well-shielded HPGe detector. Nothing out of the ordinary was observed. In particular, in the 2–10 pCi/sample level, ^{208}Tl , ^{212}Pb , ^{228}Ac , and ^{214}Pb were observed. ^7Be and ^{40}K were measured at approximately 200 pCi/sample. These radioisotopes are naturally occurring and commonly detected in air samples.

The PEMS station also measured meteorological data. Figure 7 shows the wind rose from March 6, the day of the largest xenon spike observed. Winds were from the SSW. The NTS is NW of north Las Vegas and thus unlikely to be the source of the radioxenon.

Las Vegas Nevada (DOE-LV)

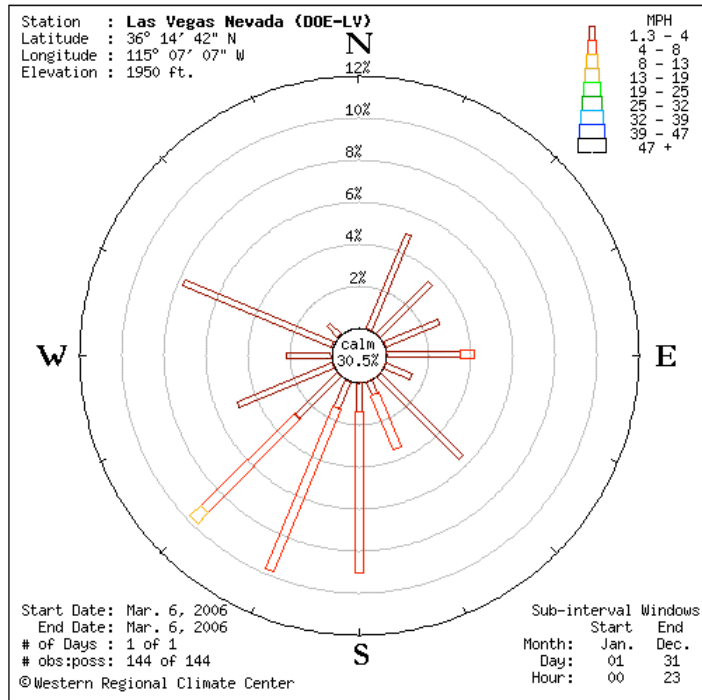


Figure 7. Wind rose for March 6, 2006.

CONCLUSIONS AND RECOMMENDATIONS

The ARSA detector was deployed in North Las Vegas for the purpose of measuring the radioxenon background at a level of sensitivity much higher than previously done in the vicinity of the NTS. Such measurements are relevant in case similar measurements were to be performed in the future at the NTS itself. A second detector, the PEMS, was deployed in conjunction with the ARSA and contained a PIC, aerosol collection filters, and meteorological sensors. Some ARSA measurements detected ^{133}Xe at levels up to 3 mBq/m^3 . While this is certainly not a large concentration of Xe, it was somewhat of a surprise. Previous measurements in areas of high nuclear reactor concentrations have shown similar results, but the western U.S., in general, does not have many nuclear reactors. Wind directions at the time indicated that the radioxenon came from a location south of the detector, and not from the NTS.

The result that the radioxenon background levels in southern NV are generally quite low, but that radioxenon can occasionally be detected is important to any future measurements that might occur at the NTS. Follow-up measurements at the NTS and over a longer period (several months, including overlapping wind-direction seasons) would be beneficial to establish expected baseline background levels. This exercise also provided valuable experience to PNNL concerning the deployment of the ARSA in a more mobile setting than it had in the past, and several improvements would have to be made prior to a subsequent deployment at NTS.

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REFERENCES

- Auer, M., A. Axelsson, X. Blanchard, T. W. Bowyer, G. Brachet, I. Bulowski, Y. Dubasov, K. Elmgren, J. P. Fontaine, W. Harms, J. C. Hayes, T. R. Heimbigner, J. I. McIntyre, M. E. Panisko, Y. Popov, A. Ringbom, H. Sartorius, S. Schmid, J. Schulze, C. Schlosser, T. Taffary, W. Weiss, and W. Wernsperger (2004). Intercomparison experiments of systems for the measurement of xenon radionuclides in the atmosphere, *App. Rad. and Isotopes* 60: 863–877.
- Bowyer, T. W., K. H. Abel, W. K. Hensley, M. E. Panisko, and R. W. Perkins (1997). Ambient ^{133}Xe levels in the northeast US, *J. Env. Rad.* 37: 143–153.
- Bowyer, T. W., C. Schlosser, K. H. Abel, M. Auer, J. C. Hayes, T. R. Heimbigner, J. I. McIntyre, M. E. Panisko, P. L. Reeder, H. Sartorius, J. Schulze, and W. Weiss (2002). Detection and analysis of xenon isotopes for the Comprehensive Nuclear-Test-Ban Treaty international monitoring system, *J. Env. Rad.* 59: 139–151.
- Bowyer, T. W., K. H. Abel, C. W. Hubbard, M. E. Panisko, P. L. Reeder, R. C. Thompson, and R. A. Warner (1999). Field testing of collection and measurement of radioxenon for the Comprehensive Nuclear-Test-Ban Treaty, *J. Radioanal. and Nuc. Chem.* 240: 109–122.
- Bowyer, T. W., R. W. Perkins, K. H. Abel, W. K. Hensley, C. W. Hubbard, A. D. McKinnon, M. E. Panisko, P. L. Reeder, R. C. Thompson, and R. A. Warner (1998). Xenon radionuclides, atmospheric: Monitoring in *Encyclopedia of Environmental Analysis and Remediation*, R. Meyers (Ed.), pp. 5295–5314. New York City: John Wiley & Sons.
- CEMP (2007). <http://www.cemp.dri.edu/>.
- CEMP Station Summary (2007). http://www.cemp.dri.edu/cgi-bin/cemp_stations.pl?stn=lasv.
- McIntyre, J. I., K. H. Abel, T. W. Bowyer, J. C. Hayes, T. R. Heimbigner, M. E. Panisko, P. L. Reeder, and R. C. Thompson (2001). Measurements of ambient radioxenon levels using the automated radioxenon sampler/analyzer (ARSA), *J. Radioanal. and Nuc. Chem.* 248: 629–635.
- Protocol to the Comprehensive Nuclear-Test-Ban Treaty* (1996). Part 1, Section C, Paragraph 10.