ENVIRONMENTAL MONITORING OF RADIOXENON IN SUPPORT OF THE RADIONUCLIDE MEASUREMENT SYSTEM OF THE INTERNATIONAL MONITORING SYSTEM

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ABSTRACT

The Automated Radioxenon Analyzer/Sampler (ARSA) has been deployed at several locations throughout the world: Richland, WA; New York City, NY; Charlottesville, VA; Freiburg, Germany; and, most recently, Guangzhou, China. In each of these locations, the ARSA has measured varying concentrations of ^{131m}Xe, ^{133}Xe, ^{133m}Xe, and ^{135}Xe. These concentrations of radioxenon come from a variety of sources such as nuclear reactors, medical hospitals, and nuclear fuel reprocessing; the concentrations vary by location. This makes it necessary to utilize the isotope ratios as well as their concentrations to differentiate ambient radioxenon emissions from those released by clandestine underground nuclear explosions. High concentrations and multiple isotope identification within a single sample are good measures of potentially suspect radioxenon emissions. Utilizing the ratios of the concentrations of ^{135}Xe to ^{133}Xe and ^{133}Xe to ^{133m}Xe enhances the separation of the more mundane emissions from clandestine underground nuclear detonations. This paper presents concentration data collected from each of the sites and explores the ability of ratios to discriminate between reactor effluents and underground nuclear tests. Analyses to date indicate that concentration levels, multiple radioxenon isotopes in a single sample, the presence of ^{133m}Xe and several isotopic ratios will all be good indicators of clandestine underground nuclear explosions and the combination of two or more of these measures will provide strong evidence of such activities.

OBJECTIVE

Several versions of the ARSA, developed at the Pacific Northwest National Laboratory (PNNL), have been deployed at several locations throughout the world during the past five years. Deployments were first undertaken at the Environmental Monitoring Laboratory (EML) in New York City during 1997 (Bowyer 1997), second in Orlando, Florida in 1999, and third in Freiburg, Germany in 1999-2001 (Bowyer 2002). Additional systems were run in Orlando Florida and Charlottesville, VA in 2001-2002 with one of the units deployed in Guangzhou, China in late July 2002; a PNNL model has run continually in Richland, WA area since May of 2001. All systems have measured ^{133}Xe and ^{135}Xe, which represent two of the three most important radioxenon isotopes that are indicative of a nuclear explosion. In some locations, concentrations of ^{131m}Xe have been measured and less frequently ^{133m}Xe has been measured.

These measured radioxenon concentrations represent an ambient background that will be present at some of the planned deployment locations outlined by the Provisional Technical Secretariat (PTS). These emissions can be attributed to nuclear power reactor operations, hospital use of ^{133}Xe for medical testing, and fuel reprocessing activities to a lesser extent (Bowyer and Perkins 1998). In all instances the measured concentrations for ^{133}Xe have been well above the 1 mBq/m^3 minimum detectable concentration (MDC) requirement adopted by the Comprehensive Nuclear-Test-Ban-Treaty Organization (CTBTO) Preparatory Commission (Preparatory 1999). For several of the deployed locations (Florida, Charlottesville, Germany, and New York City) the ARSA, which has a ^{133}Xe MDC below 0.2 mBq/m^3, had a significant number of samples with ^{133}Xe concentrations above the 0.2 mBq/m^3 MDC level. For the samples collected and measured in the Freiburg, Germany location over 95% had a detection of ^{133}Xe, see Figure 1. Though no MDC's have been set for ^{131m}Xe and ^{133m}Xe the PTS required that any of the deployed system be capable of measuring them and the ARSA has achieved MDC's for the two metastable isotopes of less than 0.3 mBq/m^3 and for ^{135}Xe below 1.0 mBq/m^3.

The measured concentrations and the identification of multiple isotopes within a single sample at all of the sampling locations merited further analysis of the data to determine the isotopic ratios from non-nuclear detonation sources. In the cases of multiple isotopes, the ^{135}Xe/^{133}Xe ratio clearly indicated that the plumes were from a source other than a nuclear explosion. The further absence of statistically significant amounts of ^{133m}Xe also provided a key indication that the plumes were from background sources. The most recent set of data obtained in Richland, Washington illustrates the identification problem; through the use of isotope identification and the ^{135}Xe/^{133}Xe ratio, the collected sample in Richland has been unambiguously assigned to the local nuclear reactor.

The discussion here is centered on nuclear reactor emissions, but it is important to point out that the two other likely sources of radioxenon isotopes will have unique ratios as well. For both medical uses and fuel processing activities the ^{131m}Xe/^{133}Xe plays an important role. For medical uses, only ^{133}Xe is used and 20 mCi is the typical dose used. These samples also contain ~0.5% ^{131m}Xe concentrations upon arrival (within 5 days of generation). After several weeks the ratio of ^{131m}Xe/^{133}Xe becomes very large (>> 10:1) and samples that are not used will be released to the atmosphere as part of the disposal process. For nuclear fuel reprocessing the ^{131m}Xe/^{133}Xe ratio will also be high because it is stored for several months to allow the decay of short-lived radionuclides before it is processed. Both of these activities will not have significant ^{135}Xe or ^{133m}Xe concentrations.

RESEARCH ACCOMPLISHED

Monitoring ambient levels of some radioxenons has a long history and the introduction of the Comprehensive-Nuclear-Test-Ban-Treaty in 1996 increased the interest in measuring atmospheric concentrations of the four radioxenons that are most likely to escape from an underground nuclear explosion and last long enough to be measured by a world wide radioxenon network. Historically, both anthropogenic and cosmogenic sources of these radioxenon isotopes were investigated and initial work indicated that concentrations from even small (1kT) subsurface nuclear explosions would be much greater than those produced by other means (Bowyer and Perkins 1998). Figure 2 shows the ^{133}Xe/^{133m}Xe and ^{135}Xe/^{133}Xe ratios that are expected for an operating nuclear reactor in equilibrium and those expected for an underground nuclear explosion. It is clear that the ratios are several orders of magnitude higher for a nuclear detonation than they are for a nuclear reactor.

New York City 1997

The first reported environmental measurement of 135Xe concentration were taken by the first prototype ARSA unit at the Environmental Monitoring Laboratory in New York City during April of 1997(see Figure 3). The 135Xe/133Xe ratio for this sample was 0.13 and, along with the lack of any measurable 133mXe concentration, indicates that this plume was from a local reactor, Xe-133 concentrations were also seen during this field test.

Freiburg Germany (1999-2001)

The PTS Phase II testing was done at the Institute for Atmospheric Research (IAR) and included systems from Sweden (SAUNA), France (SPALAX), Russia (ARIX) and the ARSA system. IAR hosted the test and provided analysis of the archived samples that each of the systems collected. Comparisons of the concentration data from each of the systems were in good agreement with one another and will be detailed in a subsequent publication.

During this test the ambient concentrations of ^{133}Xe fluctuated between approximately 1.0 and 120 mBq/m^3 on a daily basis (see Figure 1). Xenon-135 was seen in several of the samples during the 15-month test and the other two isotopes of interest ^{131m}Xe and ^{133m}Xe were seen as well. For one of the samples collected during this time a ^{135}Xe/^{133}Xe ratio of 2.1 was measured, which is almost four times higher than that expected for a nuclear reactor operating in equilibrium. This was a clear indication that additional reactor operations analysis was required to account for a sample that was not from an underground nuclear detonation.

High ratios of ^{131m}Xe/^{133}Xe were seen on two occasions, which indicates either a release from a nuclear fuel reprocessing plant or a release of an old (>2 months) medical sample of ^{133}Xe by a nearby hospital.

Richland 2001

The running of the ARSA system in Richland has typically yielded radioxenon concentrations below the MDC levels of the system. Low radioxenon concentrations are expected in several of the PTS radionuclide station locations in the Southern Hemisphere due to the low numbers of nuclear reactors

The Richland location of the ARSA offers a unique test ground for ratios as it is in very close proximity of a nuclear reactor run by Columbia Generating Station (the WNP-II 2-GW BWR is less than 8 miles from the sampling station) with a prevailing wind direction blowing effluents away from the sampling site. During July of 2002, the reactor had a scheduled shutdown for biannual maintenance and refueling. A spike of ^{133}Xe and ^{135}Xe was seen at the sampling site within hours of reactor shutdown. The ^{133}Xe concentration was fairly low (0.74 mBq/m^3) and the ^{135}Xe concentration was (3.1 mBq/m^3), which was high for the Richland area, as ^{135}Xe had not been detected previously. This gave a ^{135}Xe/^{133}Xe ratio of ~4.2, which is the highest that has been measured by the ARSA. There were no measurable quantities of ^{133m}Xe or ^{131m}Xe found in the two samples that had the ^{133}Xe and ^{135}Xe. With the high ratio of ^{135}Xe/^{133}Xe and the lack of ^{133m}Xe, the samples indicate the source was a nuclear reactor that was not in equilibrium.

These results are consistent with results obtained by irradiation of 10.1 grams of HEU using a 9.0 x 10^8 neutron/sec PuBe source at PNNL. The HEU was sealed in a gastight container, irradiated for several days and, thus, provided ^{133}Xe and ^{135}Xe. The head gas was drawn off and directly injected into the ARSA beta cells to provide calibration data for the beta energies (see Figure 4). The ^{133}Xe/^{135}Xe ratio ranges between 6 and 10, and depends on how soon the sample is injected into the ARSA detector after irradiation and the length of the irradiation.

Nuclear Reactor Shutdown

During a reactor shutdown the rapid decrease in the neutron flux allows the buildup of ^{135}Xe within the fuel rods, i.e. the ^{135}Xe is no longer being burned. Thermal stresses related to the reactor cool down facilitate the release of the trapped radioxenons within the fuel rods. Figure 5 shows the increase in the ^{135}Xe after shutdown of the reactor. The concentration peaks after about 10 hours by a factor of approximately 20 from normal operations; thus the ^{135}Xe/^{133}Xe ratio will go from about 0.5 to 10 during the first day after shutdown. The July 28, 2001 release from WNP-II 2-GW BWR was 4.2, which is well matched to the expected results.

Nuclear Reactor Startup

During the startup of a nuclear reactor the ^{135}Xe builds quickly compared to ^{133}Xe because the shorter half-lives of the ^{135}Te and ^{133}Te grandparents and ^{135}I and ^{133}I parents that feed into their respective production. This coupled with an increase in gas leakage due to thermal stressing of the fuel rods leads to high ratios and high concentrations of both xenon isotopes. The reactor is slowly ramped up over a 10-day period in 10 daily steps of 10%. Figure 6 shows the various ratios determined for a pressurized water reactor. It takes approximately 3 weeks to be within 10% of equilibrium for the ^{135}Xe/^{133}Xe ratio, more than 4 weeks for the ^{133}Xe/^{131m}Xe ratio to be in equilibrium and the ^{133}Xe/^{133m}Xe ratio is stable shortly after startup.

As the reactor comes up to full power, the production of ^{135}Xe is suppressed because of its extremely high neutron capture cross-section (2.6×10^6 barns) and the ^{135}Xe/^{133}Xe ratio will drop to those levels indicated for a stable reactor in Figure 6. After the reactor has reached equilibrium the ^{135}Xe/^{133}Xe ratio is approximately 0.5 and the ^{135}Xe/^{133m}Xe ratio is approximately 11. The release of additional ^{131m}Xe, ^{133m}Xe and ^{133}Xe and ^{135}Xe is further suppressed because the reactor/fuel rods have reached thermal equilibrium.

CONCLUSIONS AND RECOMMENDATIONS

As more data are acquired from the several radioxenon monitoring stations currently operating analyses will be required to discriminate between reactor related radioxenon and that expected from possible underground nuclear detonations. Analyses to date indicate that concentration levels, multiple radioxenon isotopes in a single sample, the presence of ^{133m}Xe and several isotopic ratios will all be good indicators of clandestine underground nuclear explosions and the combination of two or more of these measures will provide strong evidence of such activities.

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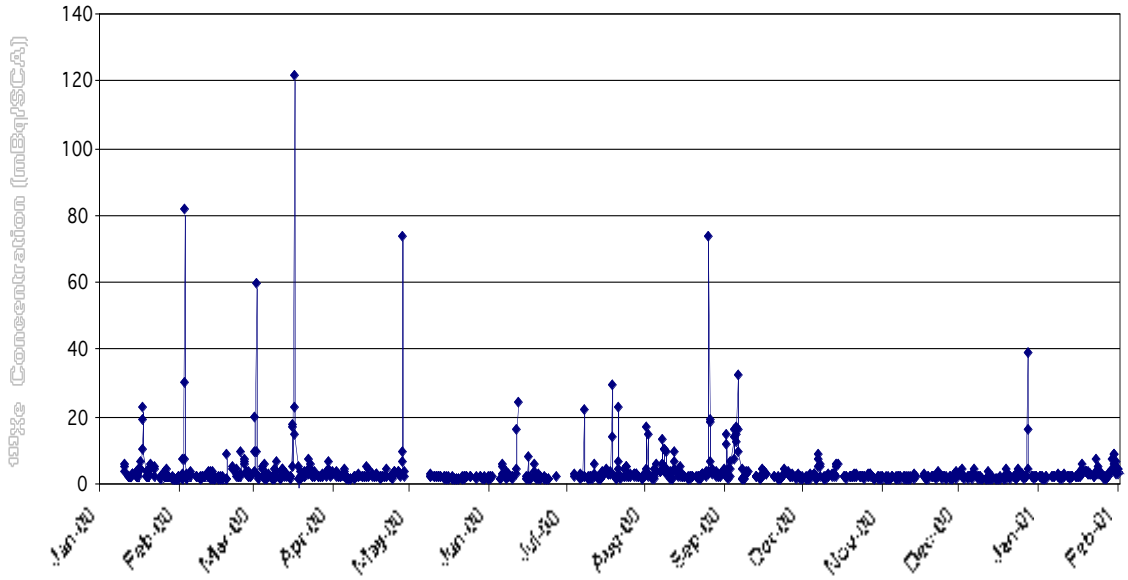

Figure 1. Freiburg ^{133}Xe concentration results for the period January 2000 to February 2001. Over 1100 samples were collected and measured during the test. The chart clearly indicates that for radioxenon measurement systems located in areas with large numbers of nuclear reactors high concentration of ^{133}Xe will be seen regularly.

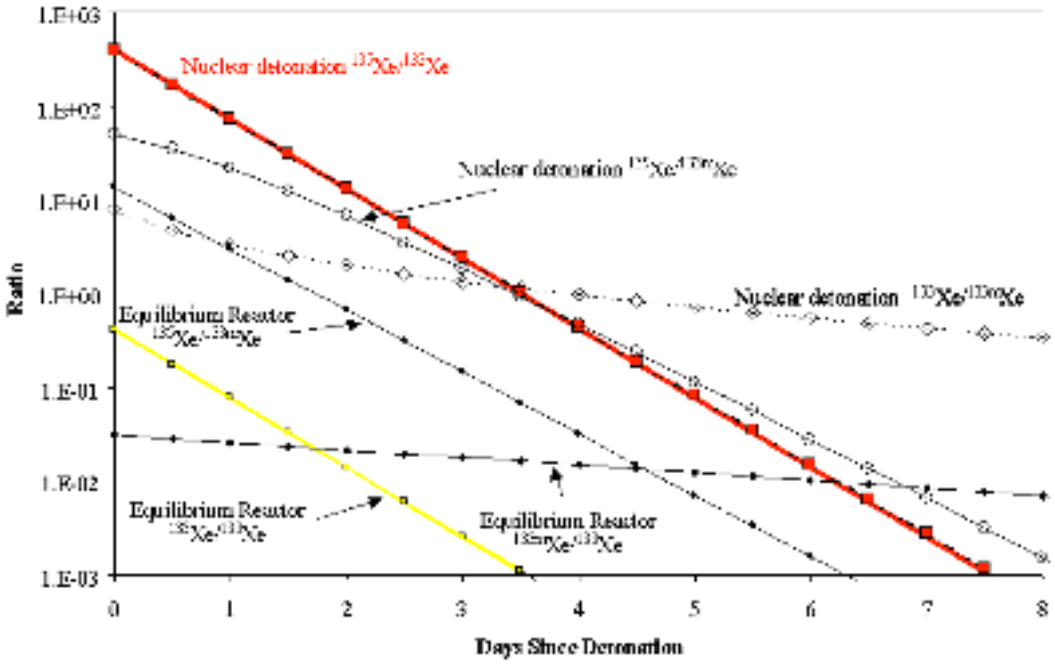

Figure 2. Expected activity ratios for ^{133}Xe/^{133}mXe, ^{133}Xe/^{133}mXe and ^{135}Xe/^{133}Xe with respect to release time from a subsurface nuclear detonation and from an operating reactor. All of the reactor ratios are lower than the nuclear detonation ratios, and in some cases by several orders of magnitude for several days.

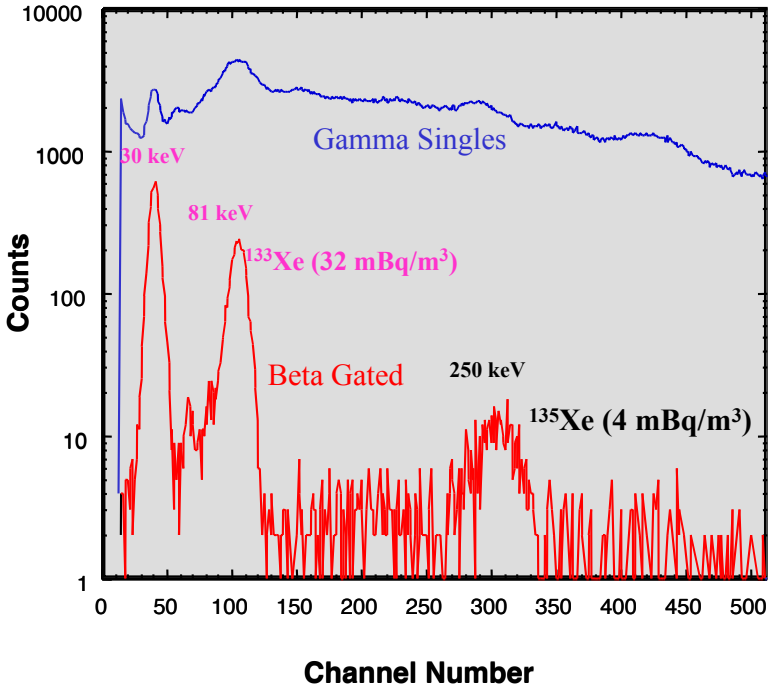

Figure 3. Gamma-ray spectrum of first reported environmental concentration measurement of ^{135}Xe along with ^{133}Xe. The lower red curve is the beta-gated gamma spectrum and the higher blue curve is the ungated gamma spectrum. This data was taken by the first prototype ARSA unit at the Environmental Monitoring Laboratory in New York City, 1997. The ^{135}Xe/^{133}Xe ratio for this sample was 0.13 that is indicative of a release from a stable reactor or a plume that has traveled large distances from either a reactor that was powering up or down.

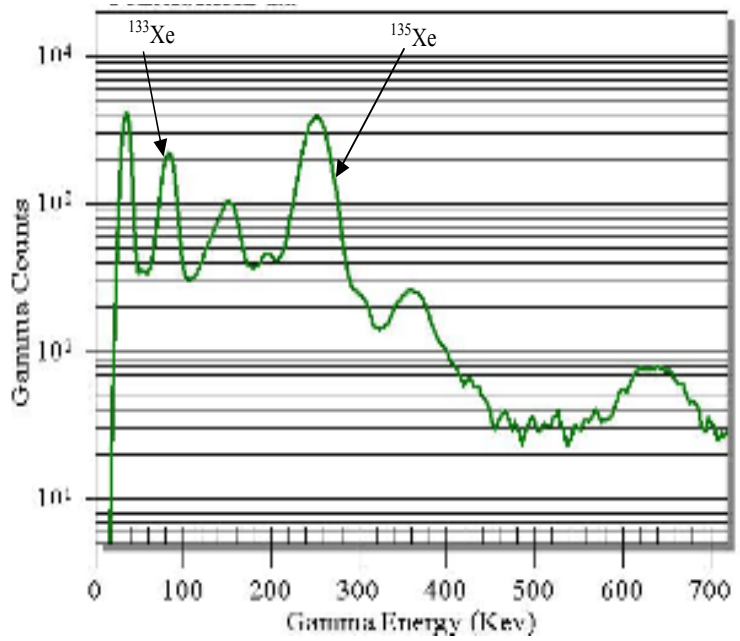

Figure 4. A beta-gated gamma spectrum from ARSA obtained using the fission product gases produced by irradiating HEU with a PuBe neutron source. The two radioxenon isotopes ^{133}Xe and ^{135}Xe are present at 30 and 81-keV and 250-keV respectively.

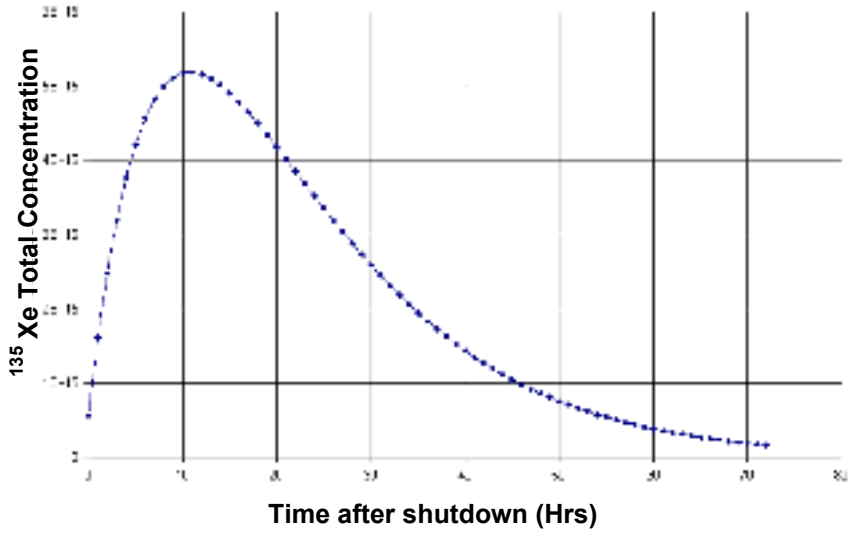

Figure 5. Xenon-135 concentration with respect to time after a full power shutdown of PWR reactor. The increase is due to the lack of burn up caused by neutrons in the reactor core.

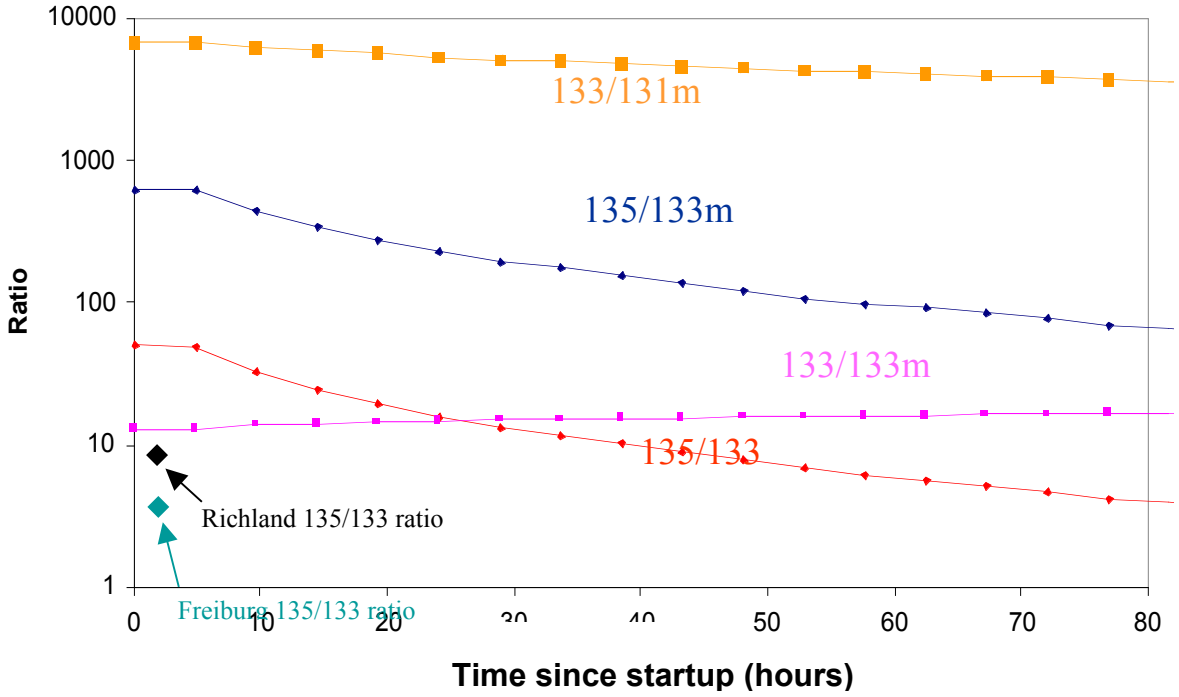

Figure 6. Radioxenon ratios during startup of a pressure-water reactor. The two data points represent the measured ratios for the Freiburg and Richland radioxenon samples.