

Measurements of radioxenon in ground level air in South Korea following the claimed nuclear test in North Korea on October 9, 2006

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Received: 1 July 2009 / Published online: 25 July 2009
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Abstract Following the claimed nuclear test in the Democratic People's Republic of Korea (DPRK) on October 9, 2006, and a reported seismic event, a mobile system for sampling of atmospheric xenon was transported to the Republic of South Korea (ROK) in an attempt to detect possible emissions of radioxenon in the region from a presumed test. Five samples were collected in the ROK during October 11–14, 2006 near the ROK–DPRK border, and thereafter transported to the Swedish Defense Research Agency (FOI) in Stockholm, Sweden, for analysis. Following the initial measurements, an automatic radioxenon

sampling and analysis system was installed at the same location in the ROK, and measurements on the ambient atmospheric radioxenon background in the region were performed during November 2006 to February 2007. The measured radioxenon concentrations strongly indicate that the explosion in October 9, 2006 was a nuclear test. The conclusion is further strengthened by atmospheric transport models. Radioactive xenon measurement was the only independent confirmation that the supposed test was in fact a nuclear explosion and not a conventional (chemical) explosive.

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Keywords Nuclear test · Radioxenon · Noble gas ·
CTBT · ROK · DPRK

Introduction

Following the claimed nuclear test in the Democratic People's Republic of Korea (DPRK) on October 9, 2006, a mobile system for sampling of atmospheric xenon was transported to the Republic of South Korea (ROK) in an attempt to detect possible emissions of radioxenon in the region. Five air samples were collected in ROK during October 11–14, 2006, and thereafter transported to the Swedish Defense Research Agency (FOI) in Stockholm, Sweden, for processing, activity concentration measurement, and analysis. In order to gain further confidence in the analysis and interpretation of these measurements, an automatic radioxenon sampling and analysis system was installed at the same location in ROK, performing measurements on the normal atmospheric radioxenon background in the region. The background measurements were performed during November 2006 to February 2007. This paper describes the measurements performed, the data

analysis, and a discussion of the results. Seismic measurements were reported a few days after the announcement by the International Monitoring System of the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO), though that evidence did not prove that the event was a nuclear explosion. Only through the detection of fission and/or activation products is it possible to determine conclusively that a suspected event was in fact nuclear. Specifically in the case of underground detonations, the detection of radioactive xenon is crucial because it is the mostly likely radionuclear emitted in this case.

Apparatus and measurement procedure

Apparatus

The measurements presented in this report were conducted using two pieces of equipment; a mobile system for xenon sampling, and a stationary system for automatic radioxenon sampling, processing and analysis (SAUNA II—Swedish Automatic Unit for Noble gas Acquisition, version II). The SAUNA prototype was originally developed by FOI [1], and the system is now produced commercially by Gamadata AB, Uppsala, Sweden. Compared to the prototype, the commercial system has been modified in several important ways. In particular, the SAUNA II is capable of producing xenon samples of maximum 1.4 ml, to be compared to 0.5 ml for the prototype, thus yielding an approximate factor of three improvement in the detection limit. The system is used at several monitoring stations within the International Monitoring System (IMS) being established by CTBTO, and has undergone extensive testing and evaluation within the International Noble Gas Experiment (INGE) [2].

The SAUNA II system consists of three main units, performing air sampling, processing, and activity concentration measurement. Air sampling is performed by flowing dried compressed air at an air flow of about 1.5 m³/h through columns containing molecular sieves and activated charcoal. One 12 h sample contains xenon from about 15 m³ of air. After purification to remove CO₂, water vapor, and radon, the sample is introduced into a gas chromatograph for further radon separation and quantification of the stable xenon content. Helium is used as carrier gas in all steps. After quantification of the stable xenon the sample is transferred to the measurement cell in a beta-gamma detector system. The beta-gamma coincidence detector consists of a 6.4 cm³ plastic scintillator cell used both as a container for the sample and as beta detector. The gamma- and X-rays from the decays are recorded using a 5 in. cylindrical NaI (TI) detector surrounding the beta cell. The read-out electronics records the pulse heights from the

two detectors in coincidence, resulting in a two-dimensional spectrum, allowing activity concentration analysis using beta- and gamma spectroscopy.

The collection of the five initial samples was performed by a mobile SAUNA xenon sampler. The mobile system can perform uninterrupted sampling in 12-h cycles, similarly to the stationary SAUNA II system. The collected sample is desorbed and moved onto a charcoal transport column for transport to the laboratory unit where xenon separation, quantification, and activity concentration measurement are performed. The laboratory unit is identical to the processing and activity analysis part of the SAUNA II system described above.

Measurement procedure

At midnight local time on October 11, the mobile sampler arrived at the selected sampling site in ROK. The sampling site, located close to the DPRK–ROK border, was chosen on the basis of meteorological forecasts. Collection in a continuous mode was started at 21:02, October 11 (UTC-time). In total, five 12-h xenon samples were collected in this first measurement campaign. The samples arrived to the laboratory at FOI in Stockholm, Sweden in two batches. The first sample arrived to FOI in Sweden on October 14, and was immediately processed and analyzed. The remaining four air samples arrived on October 17, and were processed during the following days. The time schedule for sampling and measurement is shown in Fig. 1.

Preceding each sample activity concentration measurement a gas background measurement was performed. The data acquisition times for these gas background measurements were similar to those for the samples (15 h or more). In addition to the gas background measurements, an earlier recording of the ambient detector background was used in the analysis. A more detailed description of the sampling and analysis procedure can be found in Ref. [3]. Following the initial measurement campaign, additional measurements at the same location were carried out. The main motivation was to establish the general background conditions in the area in order to improve the analysis of the initial data set. The stationary SAUNA II system was transported to ROK at the end of October 2006, installed at the site, and first data were produced on November 7, 2006. In total, 82 background samples were measured until January 26, 2007.

Data analysis

The routine analysis method for SAUNA samples is the so called net-count method, developed within INGE. A detailed description of this analysis method [4] and the

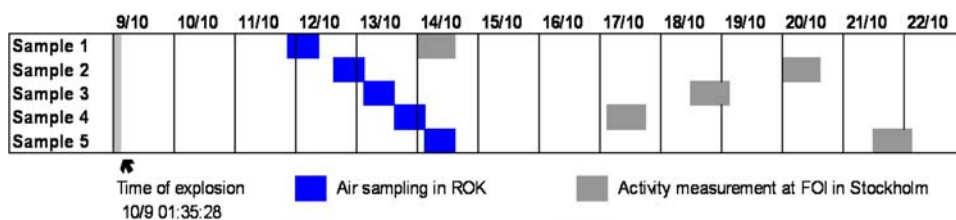


Fig. 1 Time schedule (UTC) for the sampling and activity concentration measurements of the five samples collected in ROK shortly following the claimed nuclear test in DPRK. The time of explosion is the one reported by the International Data Center (IDC), CTBTO, Vienna

software [5] can be found elsewhere, only the main principles are discussed here.

The air concentration of xenon radioactivity C is calculated using the formula

$$C = \frac{n \lambda^2 t_c}{\epsilon_{\beta\gamma} \beta \lambda F_C F_P F_A V} \tag{1}$$

where n is the background-corrected number of counts in the region of interest (ROI) corresponding to beta-gamma coincidences of the isotope in question, λ is the decay constant of the isotope, $\epsilon_{\beta\gamma}$ is the energy-averaged detection efficiency for the ROI, $\beta\gamma$ is the combined beta-gamma branching ratio to the decay, t_c is the sample collection time, and V is the sampled air volume. Decay corrections during sampling, processing and activity concentration measurement are accounted for by the factors F_C , F_P , and F_A , where $F_C = 1 - e^{-\lambda t_c}$, $F_P = e^{-\lambda t_p}$, and $F_A = 1 - e^{-\lambda t_a}$, where the processing time is denoted by t_p and the data acquisition time by t_a . Equation 1 is based on the assumption that the concentration and sampling rate is constant during sampling and that all isotopes decays independently. The latter is not true if ^{133m}Xe is present in the sample, since this isotope decays into ^{133}Xe . In this case it can be shown that the atmospheric concentration of ^{133}Xe is given by

$$C_{133} = C_{133}^0 - \xi \cdot C_{133m}, \tag{2}$$

where C_{133}^0 is the ^{133}Xe concentration obtained when no correction for the parent decay is performed, and ξ is a decay correction parameter depending on the collection, processing, and acquisition times (see Ref. [3] for details).

The analysis routine uses 10 ROIs selected to cover the main beta-gamma decays of the four isotopes ^{133}Xe , ^{131m}Xe , ^{133m}Xe , and ^{135}Xe . The number of net-counts for each ROI is estimated by extracting the gross counts in all ROIs for sample, gas background, and detector background, and subtracting the interfering contributions from other isotopes using experimentally determined interference factors. The analysis of the five samples using the net-count method indicated concentrations of the metastable isotopes above the critical limit for a few of the samples. Since the isotopic composition (i.e., isotopic ratios) is crucial in order to determine the nature of the source, a more detailed

investigation of this result was motivated. An alternative analysis method [3] was developed, using a high-statistics ^{133}Xe spectrum measured on October 3, 2006, with the same detector. The basic concept in the analysis is to use this spectrum as a standard spectrum for ^{133}Xe , and fit projected gamma- and beta sample spectra to this distribution, together with Gaussian functions describing the conversion electron peaks from ^{131m}Xe and ^{133m}Xe . The obtained concentrations using this technique agreed with the results from the net-count method within 1σ uncertainties. In addition, it was found that the critical limits (L_C) were lowered using standard spectrum fitting; which is not surprising since this method, in contrast to the net-count technique, uses the detailed shape of the spectrum.

The atmospheric concentrations presented in the next section are calculated using the standard-spectrum technique for the five initial samples (except for ^{135}Xe , where no standard spectrum were available), and with the net-count technique for the rest of the samples. One example from the standard spectrum analysis is given in Fig. 2, where two different fits to the beta energy spectrum gated on the 30-keV xenon X-ray peak are shown for one of the initial five samples (Sample 5 in Table 1). The left panel of Fig. 2 shows a beta energy fit in the 30-keV gamma region assuming only ^{133}Xe present in the spectrum. It is difficult to account for all signal strength assuming this is the only isotope present in the sample. The right panel shows the result when the fit includes also the two Gaussian functions describing the metastable states, and as can be seen, the two conversion electron peaks corresponding to the decays of ^{131m}Xe and ^{133m}Xe are better accounted for.

Results

The five initial samples

Results from the concentration analysis of the five initial samples are shown in Table 1, where the atmospheric concentrations for ^{133}Xe , ^{131m}Xe , ^{133m}Xe , and ^{135}Xe are given, as well as the critical limits and the minimum detectable concentrations (if applicable). The critical limits [6] (L_C) are calculated assuming a 5% risk for false positive

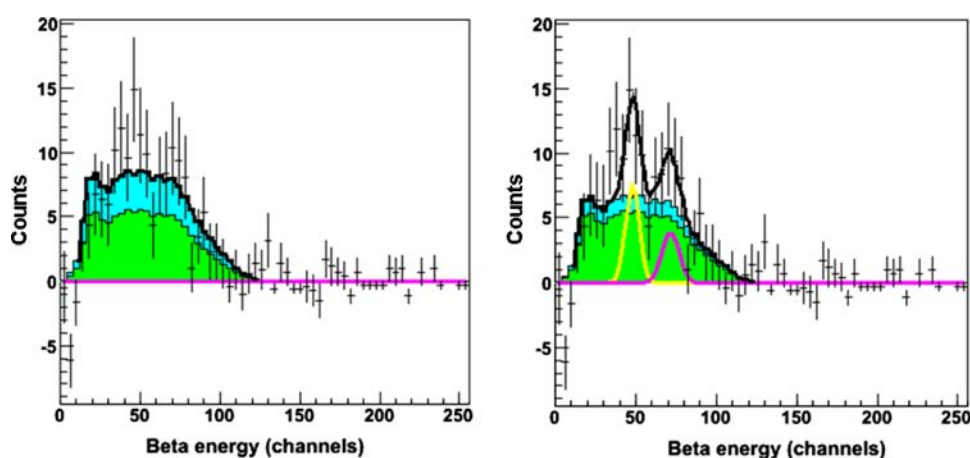


Fig. 2 Detector background subtracted beta spectra for Sample 5 in Table 1, gated over the 30-keV gamma region. The left graph shows a fit using a high statistics ^{133}Xe spectrum, and the right graph shows the result when the two Gaussians representing $^{131\text{m}}\text{Xe}$ and $^{133\text{m}}\text{Xe}$ are included in the fit. The shaded areas show the ^{133}Xe signal and

memory effect, and the thick solid line is the total result from the fit. The negative bins at the lowest channels are due to the fact that the subtracted detector background spectrum contained slightly more noise at the lowest channels compared to the sample spectrum

Table 1 Atmospheric activity concentrations (C), critical limits (L_C), and minimum detectable concentrations (MDC) for the five initial samples (in mBq/m^3)

	Collection start	^{133}Xe	$^{131\text{m}}\text{Xe}$	$^{133\text{m}}\text{Xe}$	^{135}Xe
Sample 1	10/11/2006 21:02				
C		1.48 ± 0.17	Below L_C	Below L_C	Below L_C
L_C		0.26	0.13	0.27	9.1
MDC		n/a	0.29	0.58	19
Sample 2	10/12/2006 14:40				
C		7.16 ± 0.51	Below L_C	Below L_C	$(1.4 \pm 0.7) \times 10^5$
L_C		0.75	0.23	1.62	1.1×10^5
MDC		n/a	0.50	3.48	2.4×10^5
Sample 3	10/13/2006 02:40				
C		2.00 ± 0.24	Below L_C	0.58 ± 0.38	Below L_C
L_C		0.34	0.14	0.57	2.8×10^3
MDC		n/a	0.31	n/a	6.0×10^3
Sample 4	10/13/2006 14:49				
C		1.58 ± 0.18	Below L_C	Below L_C	Below L_C
L_C		0.23	0.13	0.35	86
MDC		n/a	0.29	0.79	1.8×10^2
Sample 5	10/14/2006 02:49				
C		Below L_C	0.22 ± 0.08	0.96 ± 0.54	Below L_C
L_C		0.48	0.11	0.78	1.0×10^5
MDC		0.64	n/a	n/a	2.1×10^5

The uncertainties are at the 1σ level

and false negative detections. As can be seen in the table, four of the five samples contain clearly detectable amounts of ^{133}Xe . As expected, the critical limits for ^{135}Xe are very high due to the long transport times in combination with the short half-life for this isotope. The only sample with a reasonable low L_C for this isotope (Sample 1) did not

contain any traces of ^{135}Xe . Sample 5 was the only one having a $^{131\text{m}}\text{Xe}$ concentration above L_C . The concentration of $^{133\text{m}}\text{Xe}$ was calculated as being slightly above the L_C in Sample 3, and more significantly above in sample number 5. Another observation concerning Sample 5 is that although a clear ^{133}Xe signal was observed in the

measurement, the calculated concentration was compatible with zero. This is the result of ingrowth of ^{133}Xe from the decay of $^{133\text{m}}\text{Xe}$ (see Eq. 2) during sample transport and activity concentration measurement. Sample 3 resulted in a $^{133\text{m}}\text{Xe}$ concentration below the critical limit (1.645σ), but was detected at the 1σ level.

Results of the background measurement

The isotopic concentrations as a function of collection start time for ^{133}Xe , $^{131\text{m}}\text{Xe}$, $^{133\text{m}}\text{Xe}$, and ^{135}Xe , are shown in Fig. 3. The first five samples reported in more detail in

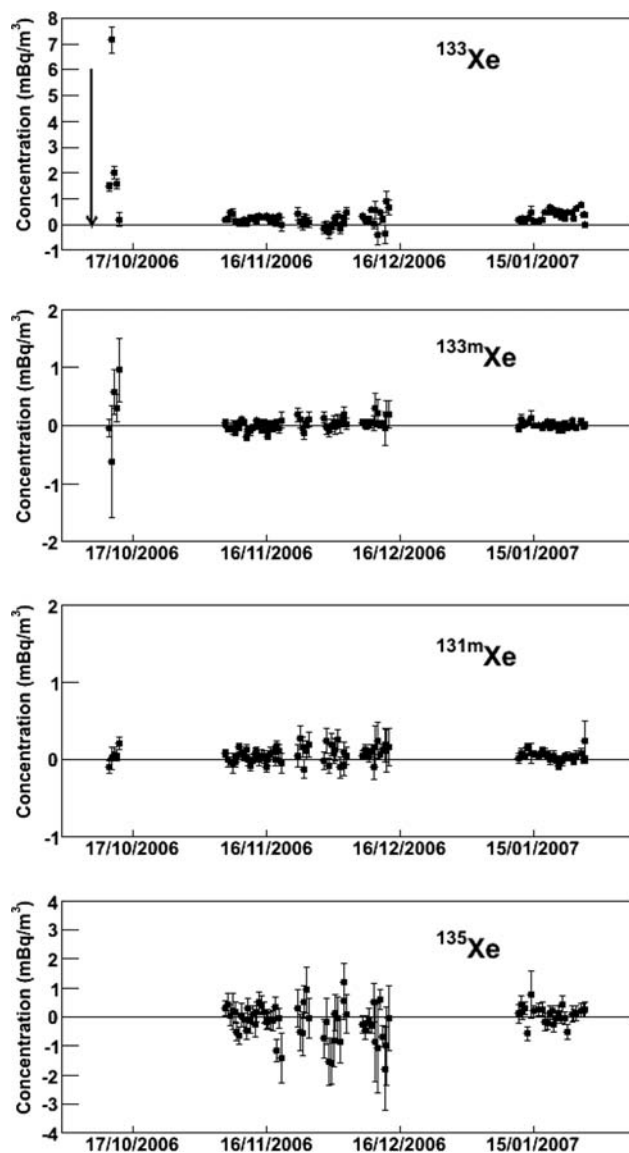


Fig. 3 Measured atmospheric concentration time series for ground level air in ROK collected between October 11, 2006, and January 26, 2007. The uncertainties are at the 1σ level. The time of the explosion is indicated by the arrow in the uppermost panel

Table 1 are visible as a clear peak in the very beginning of the time series for ^{133}Xe . Furthermore, the indications of $^{133\text{m}}\text{Xe}$ in the first measurements can be seen in the second panel.

The background data were recorded in two batches, interrupted by a time gap caused by technical problems. The average ^{133}Xe concentration in the background was 0.24 mBq/m^3 , with an observed maximum concentration of 0.9 mBq/m^3 . A total of 53 out of the 82 samples contained ^{133}Xe above the critical limit. In addition to ^{133}Xe , there is clear evidence of $^{131\text{m}}\text{Xe}$ in the background data, detected above the critical limit in 20 of the samples. To further investigate this, sample spectra for each detector were summed, and as can be seen in Fig. 4, the 129-keV conversion electron peak from $^{131\text{m}}\text{Xe}$ is clearly observed. The average concentration of $^{131\text{m}}\text{Xe}$ for the background samples was 0.06 mBq/m^3 , and the maximum concentration was 0.3 mBq/m^3 . No clear evidence of $^{133\text{m}}\text{Xe}$ could be observed in the background data. Five samples, or about 6%, resulted in concentrations slightly above the critical limit. Since this limit was defined at the 95% confidence level, 5% of the samples should result in a detected concentration for statistical reasons only. Furthermore, no indications of the $^{133\text{m}}\text{Xe}$ conversion electron peak at 199 keV could be observed in the summed sample spectra (see Fig. 4). No clear evidence of ^{135}Xe could be observed in the background data, where 3 of the 82 samples (or 3.7%) had a ^{135}Xe concentration above the L_C .

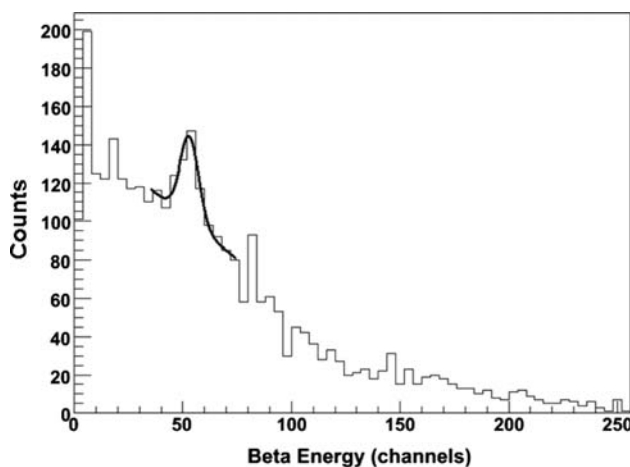


Fig. 4 Summed background beta spectra, gated over the 30-keV X-ray peak using one of the two detectors. A Gaussian fit in the expected region of the $^{131\text{m}}\text{Xe}$ peak at 129 keV is shown by a solid line. The average atmospheric concentration determined from the number of counts in the peak was $0.06 \pm 0.03 \text{ mBq/m}^3$. If present, the $^{133\text{m}}\text{Xe}$ conversion electron peak at 199 keV would have been centered at channel 72

Interpretation of results

The radioxenon source term

If all of the fission gases remained inside a cavity created by a nuclear explosion, the radioxenon activities the first five days would change as a function of time according to the decay calculations [7] shown in Fig. 5. As can be seen, the $^{133\text{m}}\text{Xe}$ activity remains relatively constant for several days, while the ^{133}Xe activity grows rapidly due to decay of the precursor ^{133}I , going mainly to the ground state of ^{133}Xe . After an initial increase during the first hours, the activity from ^{135}Xe decreases rapidly due to the short half-life of both the parent ^{135}I and ^{135}Xe itself, while the $^{131\text{m}}\text{Xe}$ activity slowly increases due to the decay of ^{131}I . A large ^{135}Xe activity is initially produced, but has to be measured within a few days following the fission in order to be detected, since the half-life is only 9 h.

In the case studied here, where the sampling was conducted 72–132 h after the explosion, and the activity concentration measurement was performed 6–13 days after time zero, only ^{133}Xe and $^{133\text{m}}\text{Xe}$ would have had the possibility to be detected.

The ratio $^{133\text{m}}\text{Xe}/^{133}\text{Xe}$ inside the cavity will decrease rapidly during the first hours following the explosion. This means that a $^{133\text{m}}\text{Xe}/^{133}\text{Xe}$ ratio measured in the atmosphere is very sensitive to the time of fractionation of xenon from its precursors. This fractionation can either occur by direct leakage to the atmosphere, or the gas can be transported away from the other fission products into cracks or other cavities but not make it all the way to the ground. A possible scenario is a prompt release of a substantial fraction of the fission gases initially produced at the time of explosion, due to the sudden pressure change, followed by seepage of xenon gas in the following days.

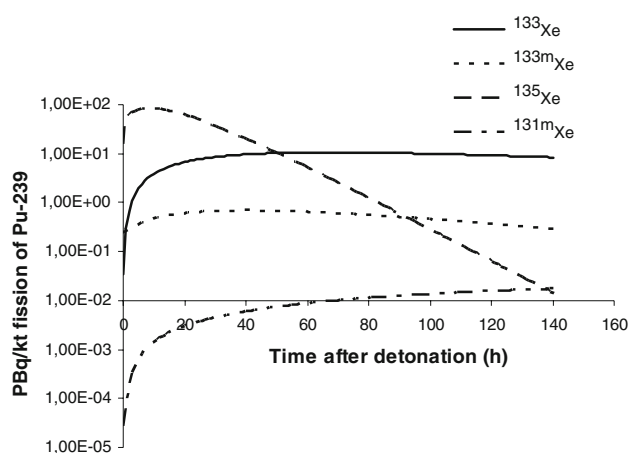


Fig. 5 Radioxenon activities as a function of time following fission of ^{239}Pu (PBq/kt). The calculations take into account the full decay chains

The gas seeping out at later times would almost exclusively consist of ^{133}Xe , since ^{135}Xe has decayed away, and a large part of the initially produced $^{133\text{m}}\text{Xe}$ would have been released initially, and only a small amount is produced later. The initial production of $^{133\text{m}}\text{Xe}$ is an order of magnitude higher for ^{239}Pu compared to ^{235}U , but the $^{133\text{m}}\text{Xe}/^{133}\text{Xe}$ ratio is about the same. This means that it is difficult to use this ratio to discriminate between fission of ^{239}Pu or ^{235}U .

Observed levels of ^{133}Xe

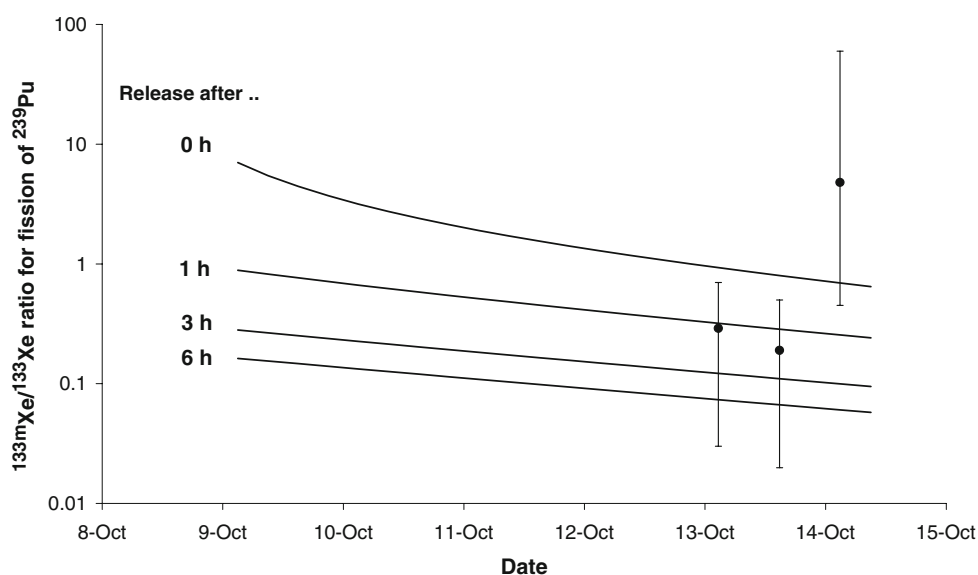
The distinct plume of ^{133}Xe with a maximum concentration of 7 mBq/m³ measured on October 11–13, 2006, together with the background data, strongly suggests a release of fission gases not normally observed in this region. Atmospheric transport modeling [8] indicates that the sampling site was particularly sensitive to the suspected test site in DPRK 2–4 days after the explosion, with winds blowing from north-northeast towards the sampling position. Notably, there are very few other possible release sources of ^{133}Xe north of the sampling point. A HYSPLIT dispersion calculation [9] assuming a 6-h release at the test site starting at 1500, October 10, results in a depletion factor at the sampling point of 10^{-16} m^{-3} at the sampling position two days later. This gives a decay corrected ^{133}Xe activity at the release point of $7 \times 10^{13} \text{ Bq}$. For a 1 kt device, the total ^{133}Xe activity is about 10^{16} Bq , and the detected activity concentration would then be compatible to a xenon seepage of 0.7% of the totally produced activity in the cavity. The uncertainties are certainly large in this estimate, but it shows that the detected ^{133}Xe activity concentration is compatible with a realistic release scenario.

Observed isotopic ratios

Figure 6 shows calculated $^{133\text{m}}\text{Xe}/^{133}\text{Xe}$ ratios as a function of time after fission of ^{239}Pu for different release scenarios. The calculations assume release from the cavity directly to the atmosphere at different times after explosion. The observed ratios are shown in the same figure. The last sample, having the highest ratio, can only be explained by a release within hours after the explosion. The ratios measured from the two other samples can be explained by a release one or a few hours after the explosion or by mixing of prompt release and fission gas from seepage one or several days after the event.

Samples 1 and 2 contained no detectable amounts of $^{133\text{m}}\text{Xe}$, and could be explained by seepage at later times, when the $^{133\text{m}}\text{Xe}/^{133}\text{Xe}$ ratio has decreased below the detection limit. As mentioned above, the possibility of mixing of ^{133}Xe by other sources should not be excluded. However, the results from the background measurements

Fig. 6 Calculated $^{133m}\text{Xe}/^{133}\text{Xe}$ ratios as a function of time after fission of ^{239}Pu for different release scenarios. The calculations assume release from the cavity directly to the atmosphere at different times after explosion. The observed ratios are shown as black circles. The uncertainties are at the 95% confidence level



discussed in “Results of the Background Measurements” section, indicates that the contribution from other sources is small. The analysis of Sample 5 also resulted in a detection of ^{131m}Xe . The observed ratios were however too high to be explained by a release from a nuclear test, and the only reasonable explanation to this observation is mixture from a second source such as medical isotope usage. This assumption is strengthened by the background observations, showing that ^{131m}Xe is frequently present in the area, at comparable concentration levels.

Summary and conclusions

Xenon measurements are key for the verification that events suspected of being nuclear tests by other means such as seismic detects are nuclear tests. Analysis of the radioxenon content in air samples collected in the ROK 2–5 days after the claimed nuclear test in DPRK showed measurable quantities of ^{133}Xe and ^{133m}Xe . The observed concentration levels and ratios can be explained by a nuclear test conducted in DPRK at the time and location registered by seismic networks.

Acknowledgements The authors wish to thank The Embassy of the Republic of South Korea in Stockholm, the ROK Ministry for Foreign Affairs and Trade, the ROK Ministry for Science and Technology, and the Korean Institute for Nuclear Safety (KINS) for fruitful cooperation. The On-Site Inspection Division at CTBTO, Vienna, is acknowledged for supporting the development of the mobile xenon sampling system. Many thanks also to Lars-Erik De Geer at FOI for valuable discussions on statistics.

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