Discrimination of Nuclear Explosions against Civilian Sources Based on Atmospheric Xenon Isotopic Activity Ratios

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Abstract—A global monitoring system for atmospheric xenon radioactivity is being established as part of the International Monitoring System that will verify compliance with the Comprehensive Nuclear-Test-Ban Treaty (CTBT) once the treaty has entered into force. This paper studies isotopic activity ratios to support the interpretation of observed atmospheric concentrations of ^{135}Xe, ^{133m}Xe, ^{133}Xe and ^{131m}Xe. The goal is to distinguish nuclear explosion sources from civilian releases. Simulations of nuclear explosions and reactors, empirical data for both test and reactor releases as well as observations by measurement stations of the International Noble Gas Experiment (INGE) are used to provide a proof of concept for the isotopic ratio based method for source discrimination.

Key words: CTBT, environmental monitoring, international monitoring system, isotope activity ratios, noble gas, radioactivity monitoring, radioxenon, source discrimination, test ban, xenon.

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1. Introduction

Xenon isotopes and their isomers are the most likely observable radioactive signatures of underground nuclear explosions at IMS stations. However, radioactive xenon is released during normal operations of nuclear facilities. So, at stations located downwind to nuclear facilities, these isotopes can frequently be detected and be unrelated to nuclear testing. Therefore, proper source characterization is important for determining whether an event is possibly a nuclear explosion.

Xenon samples will be collected and measured on a daily basis at 40 radionuclide stations of the International Monitoring System (IMS) that is currently being established (see e.g., Bowyer *et al.*, 1998; Hoffmann *et al.*, 1999). Due to their half-lives and fission yields the xenon radionuclides ^{135}Xe, ^{133m}Xe, ^{133}Xe and ^{131m}Xe are relevant for detecting a nuclear explosion (De Geer, 2001). Though two of these are metastable isomers, for convenience, this paper refers to the entities of this quartet as the four relevant xenon isotopes.

The measurements will be analyzed at the International Data Centre (IDC) in Vienna as well as at National Data Centers (NDC). The goal of the analysis is to characterize the samples and screen them according to their relevance to the signature of a nuclear explosion, though no judgment would be made by the IDC. Appropriate criteria are needed to characterize the samples as either "definitely not a nuclear explosion (i.e., screened out)" or "consistent

with a nuclear explosion or inconclusive due to insufficient data (i.e., not screened out)". The latter would be flagged for further study. The screening-out procedure has to be done based on scientifically proven and robust criteria without any prejudice, and the final judgement is left to the member states of the CTBT. The goal is to screen out as many events as reasonably possible and to keep the number of events flagged as low as possible in order to minimize costs for additional analysis. This also allows states to concentrate on fewer important events.

Several methods for categorizing radioxenon measurements are under investigation. High concentrations of one xenon isotope (e.g., ^{133}Xe) detected distantly from nuclear reactors at locations with historically low concentrations of xenon isotopes, and no other exceptional release possibilities, could serve as a criterion. In addition, the combination with other data such as relevant anthropogenic radioactivity in air filter samples or a seismic signal (HOFFMANN et al., 1999), could increase confidence that a suspected detection results from a nuclear detonation. Information on releases from reactors or isotope production facilities as part of confidence building measures will help to clarify if the source is of a civilian nature. Determination of the specific sensitivity to known sources based on atmospheric transport simulation appears to be particularly helpful for categorization (WOTAWA et al., 2010). Another approach is the Bayesian analysis of radio xenon samples (ZÄH-RINGER and KIRCHNER, 2008).

In the past it was suggested that single xenon activity ratios would be useful both for source discrimination as well as for event timing (see e.g., BOWYER et al., 1998; FINKELSTEIN, 2001; CARMAN et al., 2002). A positive conclusion on whether a measured activity ratio is consistent with a nuclear explosion has been based on assumptions that did not take into account the full range of possible isotopic activity ratios.

The main assumptions, in previous work, have been that the extended irradiation time in a reactor leads to equilibrium operational characteristics of steady power production and no change to routine procedures for off-gas treatment and release, whereas a nuclear explosion is an instantaneous process with a very short duration of a fast neutron flux resulting in different isotopic ratios. However, initial data have shown that the theoretically derived isotopic activity ratios for reactors out of equilibrium are not uncommon (AUER et al., 2004; BOWYER et al., 2002; STOCKI et al., 2005) and therefore new tools for discrimination of nuclear explosions and reactor operations are being developed.

A robust screening method is required for IDC sample categorization. To support this, new methods for source discrimination based on the relationship of two different isotopic activity ratios are investigated in this paper. It is applicable if several of the xenon isotopes are measured in a sample and if the measured ratios originate from a single release source. It requires three or four xenon isotopes to be detected and quantified. If they are not detected they can be, in certain cases, replaced by an upper limit that is related to the detection limit. The most important feature of the screening approach suggested here is the robustness of the method, independent of decay of the species. The advantage of this time-invariance is that the age of the xenon plume that intercepts the sampling site does not need to be known.

In this paper, the combinations of isotopic ratios of various calculated and empirical data sets as well as atmospheric observations are used to validate the new screening method. These data sets are:

1. Reported gaseous effluents from light water reactors (LWRs)
2. Simulations for LWR operational cycles
3. Simulations of mixing between fresh and aged air masses
4. Other civilian sources (accidental releases from low burn-up fuel, medical isotope production)
5. Simulations of various nuclear explosion scenarios
6. Empirical release data of underground nuclear tests
7. Short-time irradiated HEU targets
8. Observations of atmospheric xenon concentrations

In the first section of this paper, all these data sets are introduced. The second section uses the data of items 1–7 to demonstrate the useability of isotopic activity ratios for source discrimination. The screening method based on the relationship of different xenon isotopic activity ratios is validated against real atmospheric measurements (data set number 8) in the third section.

2. Radioxenon Releases, Observations and Simulations

2.1. Reported Light Water Reactor Release Data

A large part of the data set on radioxenon releases from nuclear power plants is taken from the reports on activity releases to the environment of radioactive materials in airborne and liquid effluents during the period 1995–1999 (VAN DER STRICHT and JANSSENS, 2001) and 2000–2003 (EC, 2004). It covers discharges from operational nuclear power stations of capacity greater than 50 MW (electric power) in the European Union (EU). During the time covered by the first report, 73 nuclear power stations totaling 148 reactors were operational and are spread over 64 different sites within the territory of the European Union. The annual xenon release data are available for 66 nuclear reactors at 55 sites in Finland, France, Germany, Spain and Sweden. In some cases, the discharge values for noble gases are reported to be below the detection limit. In addition, release data from the four Swiss nuclear power plants are included (BAG, 2000, 2001 and 2002). Another large portion of reactor release data is taken from 41 quarterly reports of nuclear power plants in the USA. The North American Technical Center (NATC) Public Radiation Safety Research Program at the University of Illinois at Urbana-Champaign compiled these data for the US effluent database (NATC, 1999). This database is the official US database provided to the United Nations (UNSCEAR) for their global report on radiation dose to man. The special feature of this database is to report continuous and batch releases separately. Distinct differences are noted between the isotopic activity ratios appearing in these two different release modes.

It should be noted that the list of individual nuclides routinely measured might vary from one installation to another. Also, the way the corresponding activity is measured or estimated, as well as measurement accuracy and precision, can be different at each installation location or in terms of the requirements of each State. The data are cumulative annual figures and, therefore, not overly sensitive for possible outliers in single measurements. Nevertheless, single discharge events can still have isotopic ratios outside the ranges presented here, for example, when new nuclear technologies are implemented.

Figure 1 shows the distributions of six different xenon isotopic activity ratios with the isotopes ^{135}Xe, ^{133m}Xe, ^{133}Xe and ^{131m}Xe derived from all reported nuclear reactor xenon releases as described above. In total there are 340 data sets, each of which contains the activities of up to four xenon isotopes. The activity ratios are sorted by increasing value. The first sorting numbers are reserved for those data sets in which both numerator and denominator are not reported and hence no ratio is available. By this way, the plot can show the number of samples in which none of the two required activities is reported. For example, the activities of both isomers ^{131m}Xe and ^{133m}Xe are missing in 140 data sets and accordingly, in Fig. 1, the entries for the ^{133m}Xe/^{131m}Xe activity ratio begin at sorting number 141. The highest sorting numbers are used for cases in which only one of the required activities is given. Accordingly, the entries end at lower sorting numbers if more data sets with one out of the two required activities are available. In particular, it can be seen from Fig. 1 that ^{133}Xe and ^{135}Xe are available in most reports and only for a handful of data sets is the activity of exactly one of these two isotopes reported.

The reported data shown in Fig. 1 exhibit a broad range of ratios spreading over several orders of magnitude. The logarithmic mean ratio is given for each isotope. Part of the spread is likely due to radioactive decay after different residence times within the reactor containment. Other effluent data clearly indicate that the LWRs were in many cases not in equilibrium that is reached after power and neutron flux are maintained constant for several weeks.

On the other hand, previous atmospheric measurements at some distance from nuclear reactors as published by AUER et al. (2004), BOWYER et al. (2002), LE PETIT et al. (2008), STOCKI et al. (2005) and SAEY et al. (2010) suggest that remote measurement of xenon is consistent with equilibrium operation in many cases. This apparent contradiction could be explained by the fact that when measurements are made closer to a reactor, it is much more likely that all four isotopes will be at high enough concentrations so as to have a higher sensitivity to
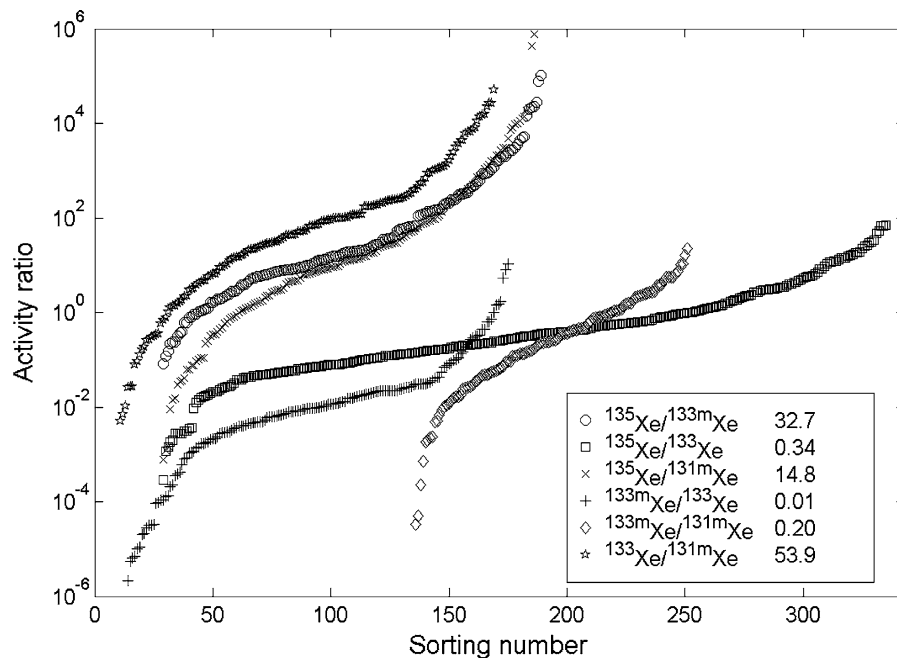
Figure 1

Distribution of released xenon isotopic activity ratios as reported from nuclear power sites. The logarithmic mean ratios are given in the text box

determining the equilibrium state of the reactor. In fact, in most cases where the measurements were made remote from reactors, only ^{133}Xe was detected, and when other isotopes were measured, usually only 2 or 3 isotopes were at high enough concentrations to allow a ratio to be formed.

A more detailed consideration on power reactor emissions can be found in KALINOWSKI and TUMA (2009).

2.2. Simulations for Light Water Reactors

The reasons for, and the potential extent of the variability of reactor releases under different operational conditions need to be understood to make progress on the screening of data collected for nuclear explosion monitoring. Figure 2 shows results of LWRs fuel burn-up simulations covering the whole operational time with the typical three-one-year power cycles with 3.2% enrichment in ^{235}U (KALINOWSKI and PISTNER, 2006).

The isotopic activity ratios depend significantly on the neutron flux and can vary over several orders of magnitude within a few days of release. Any

power change and especially shut-down and start-up of a reactor (BOWYER et al., 2002) causes a deviation from the initial values. Effluent treatment can have an impact, particularly if this leads to an extended residence time in the reactor because the isotopic ratios change by decay. Uranium enrichment, fuel burn-up and diffusion through the fuel matrix or effluent treatment filters have a comparatively low impact on the xenon ratios. Diffusion of fission gases from the fuel matrix into the free volume causes fractionation of the isotopes (see Table 4, KALINOWSKI and PISTNER, 2006).

2.3. Simulations of the Effect of Mixing between Fresh and Aged Air Masses

The process of mixing air volumes that emanate from nuclear reactors can lead to isotopic activity ratios different from any of those in the contributing air parcels. This is true even for a mixture of two parcels of air that shared the same starting ratios, but are of different ages. Mixing of this sort can occur within the contained spaces of the facility or in the atmosphere.
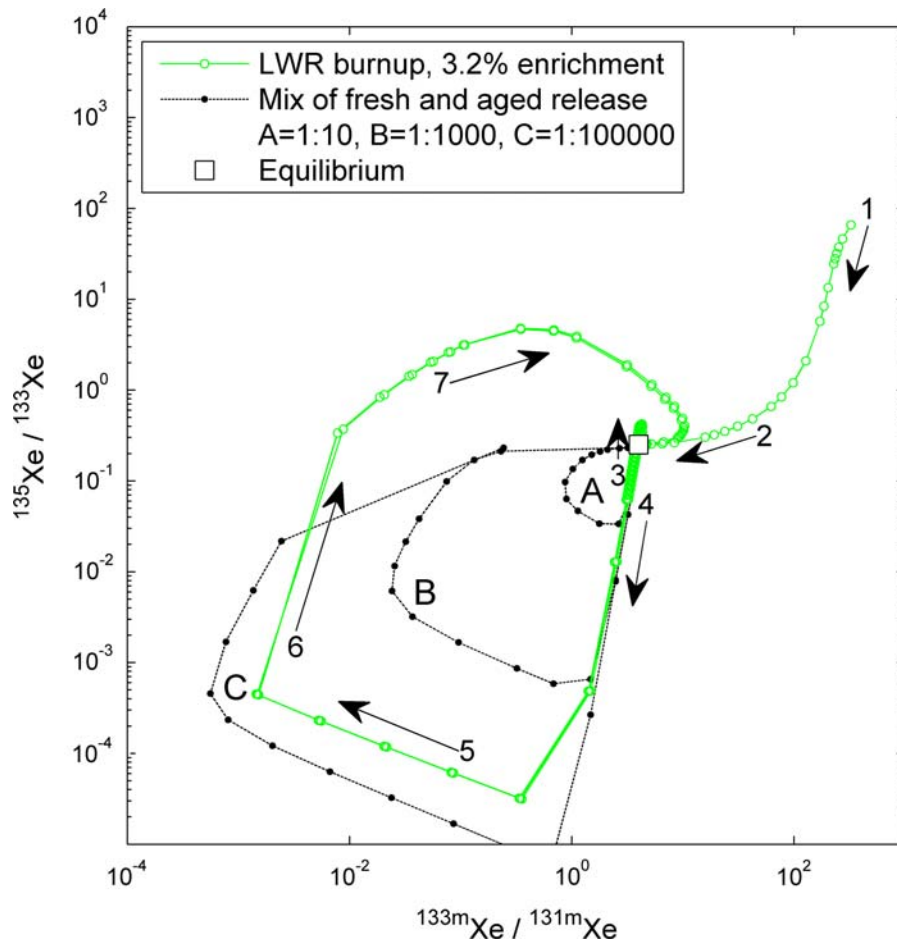
Figure 2
Reactor ratio trajectory of the first one-year power cycle of fuel plus a 30-day revision period with 3.2% enrichment in ^{235}U (KALINOWSKI and PISTNER, 2006). The equilibrium, reached after a few weeks, is marked with a large square. Description of the isotopic ratio track during the first reactor power cycle: 1. Start-up with fresh fuel only 2. Approaching equilibrium 3. Short-time increase of ^{135}Xe after power shut-down 4. Radioactive decay 5. Low neutron flux during revision period 6. Restart of reactor to full power 7. Circling back to equilibrium. The second and third power cycle repeat the track from part 3–7 very close to the first cycle. The most significant difference can be seen along part 7 of the track

In order to study the impact of mixing on the isotopic activity ratios, a number of mixing scenarios is set up. A fresh and an aged air mass are mixed in three volume ratios. Both start from the equilibrium activities in fuel with 3.2% enrichment as reached at the end of the third cycle (KALINOWSKI and PISTNER, 2006). The fresh part is kept at time zero, while the other portion is assumed to have decayed for a certain number of days. Figure 2 shows the trajectories for the three mixing scenarios. All three are similar to the no-mixing trajectory.

2.4. Other Civilian Sources

Besides discharges from nuclear reactors during normal operation and the regular large releases from medical isotope production facilities, other less frequent sources have to be considered. These include spent fuel reprocessing, medical isotope usage and disposal, and exceptional accidental releases from nuclear reactors. The latter case is of particular interest with the scenario of a short irradiation reactor, i.e., less than 7 days (FINKELSTEIN, 2001).

Normally, nuclear fuel is reprocessed with at least a year cooling time after the fuel is unloaded from the reactor core. At that time, only ^{131m}Xe and tiny traces of ^{133}Xe remain at measurable activity levels. The ^{133}Xe/^{131m}Xe isotopic ratio might still be in the order of 10^{-5} (BOWYER *et al.*, 1998) but could be significantly less due to the radioactive decay. This is already more than two orders of magnitude below the minimum found in the reactor releases (see Fig. 1). Unless the detection is very close to the reprocessing activity, or reprocessing is undertaken with an unusually short decay time, the activities will be extremely low, consequently this activity will likely have no impact on source discrimination. The equilibrium activity of ^{133}Xe reached with a burn-up of 30 GWd/MTU (Giga Watt days per metric ton of uranium) at the time of unloading the fuel is 6×10^{18} Bq/MTU, but it will be decayed to 6×10^{-3} Bq/MTU after 1 year. After that time, even the full release of all fission noble gases will most certainly not allow detection of ^{133}Xe further away than meters from an atmospheric source.

The main source of radioactive xenon isotopes for medical applications is separation of fission gases from irradiated highly enriched uranium (HEU) with a low exposure of only normally 2–11 days. In fact, radioactive xenon often is harvested as a by-product of breeding ^{99}Mo in HEU targets. Table 1 shows isotopic activity ratios for various irradiation times and days of decay. For data based on other assumptions see SAEY (2009). Some of these ratios happen to be close to the respective logarithmic mean ratio for power plant releases as shown in Fig. 1. However, other low-exposure ratios are indeed similar to explosion ratios, as found in calculations and

experiments described in Sects. 1.5 and 1.7, below, respectively (KALINOWSKI and PISTNER, 2006).

As a result, isotope production from HEU can pose a difficulty to source discrimination. It could give rise to a false alarm and requires special consideration. A worst case estimate has been created by assuming total release from a large 1 kg target exposed for 5 days with a total neutron flux of 3.5×10^{14} s^{-1}cm^{-2} following 1 day of decay. The ^{133}Xe content at the time of the release is 10^{16} Bq. Under the assumption of a 1-h release duration and a stack air volume rate of 10,000 m^3 h^{-1}, the air concentration at the point of release is 10^{12} Bq m^{-3}. This is certainly detectable at some distance (STOCKI *et al.*, 2005). The number of isotope production facilities in the world is limited, and even if the number increases their releases can be taken into consideration though it complicates the test ban verification and makes it less confident. Further information on radiopharmaceutical production facilities can be found in SAEY (2009).

Radioactive xenon isotopes for medical applications are in general not produced by exposing natural xenon to a high thermal neutron flux. Though ^{133}Xe might be produced by neutron absorption in ^{132}Xe, the content of other radioactive xenon isotopes, especially ^{125}Xe would result in an excessively high dose rate. However, the production of ^{125}I, another isotope important for medical applications, is based on the irradiation of natural xenon gas. After the nuclear reaction ^{124}Xe(n,γ)^{125}Xe, the product decays to ^{125}I. Therefore, one needs to take into account the possibility of detecting radioactive xenon resulting from this source. As an example, the isotopic composition of 5 ml natural xenon after a 6 h irradiation in a neutron flux of 7×10^{11} s^{-1} cm^{-2} is considered (KELLER, 2004). The related isotopic activity ratios are found in the nuclear explosion domain only for one of the xenon ratio relationship plots. This is the one that contains all isotopes with the exception of ^{131m}Xe.

A possible source of pure ^{131m}Xe is the medical application of its precursor, ^{131m}I, which has a half-life of 8.02 days. The ^{131m}Xe frequently observed in Ottawa, Canada originates from ^{131}I production at Chalk River Nuclear Laboratory (STOCKI *et al.*, 2005). The precursor of this iodine isotope can be generated

Table 1

Isotopic activity ratios for 93% enrichment in ^{235}U and an irradiation time of 10 and 5 days of decay and for an irradiation time of 5 and 2 days of decay

Isotopic activity ratios	Irradiation time = 10 days, decay time = 5 days	Irradiation time = 5 days decay time = 2 days
^{133m}Xe/^{133}Xe	0.02	0.04
^{133m}Xe/^{131m}Xe	6	36
^{133}Xe/^{131m}Xe	330	890

with a high purity by neutron capture: $^{130}Te(n,\gamma)$ ^{131}Te.

Another scenario that could result in isotopic ratios in the explosion domain would be an emission from a short-irradiation reactor accident. In this case, the source term could be significantly larger than in the case of medical isotope production. After an irradiation time of 15 days, the isotopic ratios are still found in the area between the reactor and the test domain. At that time, the ^{133}Xe content in one fuel element with a weight of 1 MTU results in the activity release of 10^{18} Bq. If this would be fully released within 1 h through an air volume rate of 100,000 m^3 h^{-1}, the air concentration at the stack is 10^{13} Bq m^{-3}. The simultaneous release of radioactive aerosols would certainly help to relate this kind of event to a reactor accident.

2.5. Simulations of Various Nuclear Explosion Scenarios

A nuclear explosion takes place in a very short time and the nuclear chain reaction ends in less than a second. Two extreme scenarios are possible, one in which the Xe is removed from their precursors immediately and allowed to decay, and another in which all fission products are held together, perhaps in an underground cavity, such that in-growth from precursors is allowed and the full cumulative yield of Xe is produced.

Three different neutron sources are studied with regard to the fissile material used and the neutron energy. Fission of ^{235}U and ^{239}Pu is simulated at fission neutron energies and ^{238}U with high-energy neutrons using the Bateman equations (BATEMAN, 1910) implemented in Matlab (KALINOWSKI, 2010a, b).

2.6. Empirical Release Data of Underground Nuclear Tests

SCHOENGOLD et al. (1996) report detailed atmospheric radioactivity release information for 433 nuclear tests conducted on the Nevada Test Site (NTS) from 15 September 1961 through 23 September 1992. An analysis of these data can be found in KALINOWSKI (2010a, b). Figure 3 shows the distribution of xenon isotopic activity ratios that are calculated from the reported activity releases of these
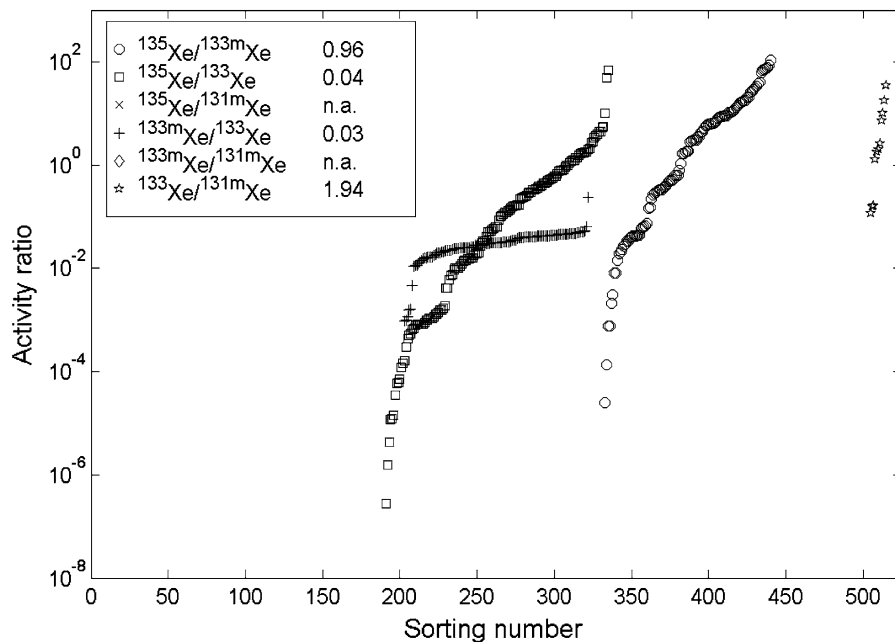
Figure 3
Distribution of released xenon isotopic activity ratios as reported for underground nuclear explosions at Nevada. The logarithmic mean ratios are given in the text box, *n.a.* not applicable

underground nuclear explosions. The data were reported at two significant figures accuracy. The spread of the activity ratios over several orders of magnitude can be explained by the delay and duration of the releases.

Only 102 of the 433 releases are considered. The selection criterion used was to pick only those events for which single release events are reported, and therefore excludes aggregated activities that are summed up from different release events at different times originating from the same test explosion.

All these 102 cases are operational releases resulting from the purging of tunnels or sometimes shafts to minimize the exposure to personnel, from drill-back operations to recover samples for diagnostic purposes, from gas sampling or from sealing the drill hole with a plug and cementing it to the surface. These releases had delays ranging between 31.5 h and 24.5 days with durations of at least 2 min and at most 5 days.

In most of the cases in which the activities of three xenon isotopes were measured and reported, the activity of ^{131m}Xe was not included. That isotope is reported only for a very few late operational and seepage release cases. The likely reason for this isomer not being measured is that its independent fission yield is 10^{-6}% or less depending on the fission reaction. In addition, the half-life of the immediate precursor is 8 days, too long for a significant build-up through the decay chain shortly after the explosion. The only exception is a late-time seepage release that occurred after operations in the relevant test area ceased with a delay of 9.5 days and a duration of 30 days. All isotopes but ^{135}Xe were measured in that case.

The two plots on top of Fig. 4 show the calculated xenon isotopic ratios that are possible for nuclear detonations using different scales; the right one being zoomed in. The change in the isotopic ratios is shown for complete fractionation involving only the independent yields (pure xenon curve) and without any fractionation converging within 1 day close to the cumulative yields (unfractionated decay chain curve). Depending on the amount of fractionation of the xenon isotopes from their parents, the actual ratios will fall between the two or precisely on one of the lines. Also shown in the figure are data points from the Nevada Test Site (NTS) (Schoengold et al.,

1996). It is notable that in all of these examples from the NTS, the xenon parents were allowed to come into equilibrium with the xenon isotopes before the measurement, as can be concluded from the fact that all points lie quite close to the unfractionated decay chain curve. This seems consistent with the fact that all samples were collected at least 31.5 h after the explosion.

Figure 4, bottom left, shows the single reported seepage release in the isotopic activity ratio relationship plot for the three isotopes excluding ^{135}Xe. No plots are shown with other ratio pairs because there are no data available with the required isotope combinations.

The agreement between the NTS measurements and the model is good, and therefore engenders confidence that the model at least bounds (see unfractionated decay chain curve in Fig. 4) the correct answer for the "worst case." Further, the comparison of data from the Nevada Test Site with the calculations illustrates that the xenon isotopes and their precursors remained basically non-fractionated before the releases took place. In other words, irrespective of the release scenario for an operational release, there is high confidence that the airborne xenon activity ratio will exhibit negligible fractionation and lie close to the unfractionated decay chain curve. This may be different for rapid uncontrolled venting but cannot be demonstrated here due to the lack of data.

2.7. Short-time Irradiated HEU Targets

Raith (2006) reports samples containing 1 µg of 90% enriched ^{235}U were irradiated at the Training, Research, Isotopes, General Atomic (TRIGA) Mark II research reactor at the Atomic Institute of the Austrian Universities in Vienna. Figure 5 shows short-time irradiated 90% enriched ^{235}U measurements. The measurements lie near the unfractionated decay chain curves and are consistent with the results of Sect. 2.5 (compare with Fig. 4).

2.8. Observations of Atmospheric Xenon

The first reported regular global xenon observations of all four relevant xenon isotopes began with the International Noble Gas Experiment (INGE), initiated
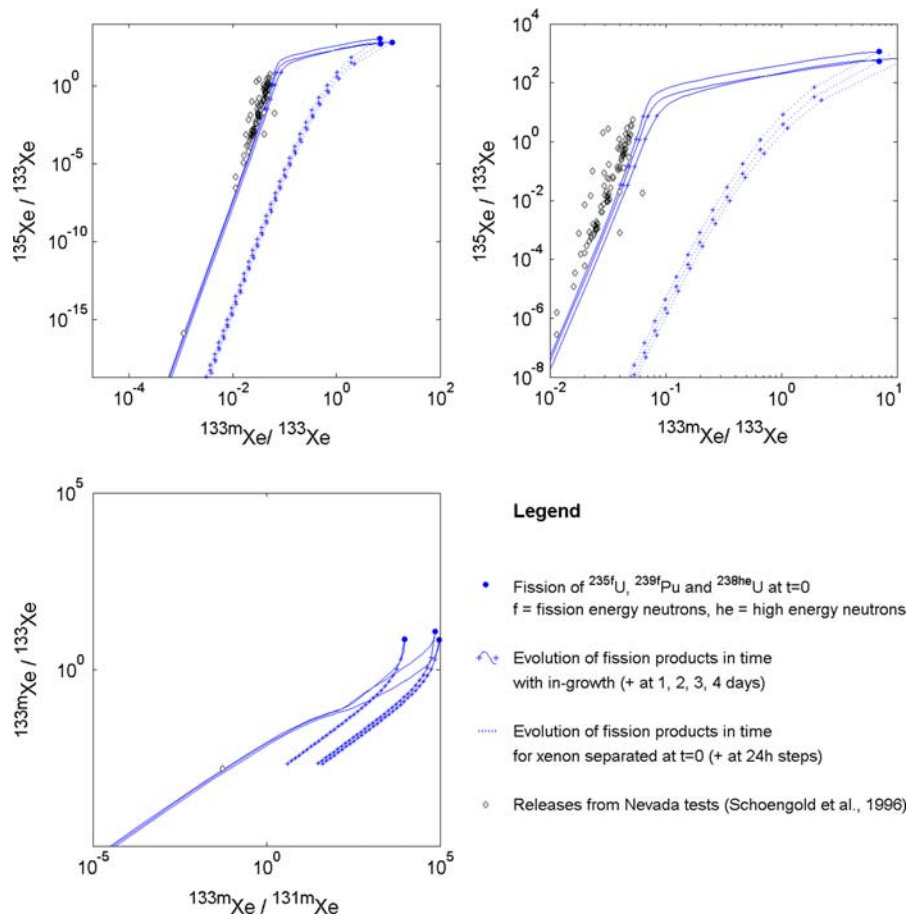
Figure 4
Comparison of simulated and reported xenon isotopic activity ratios released from nuclear tests (102 selected). The expected isotopic ratios present at $t = 0$ are denoted with the *filled circles*, and *lines* and "+" symbols are used to show the change in the isotopic ratios in time steps of full days

and headed by the CTBTO PrepCom Preparatory Commission (BOWYER *et al.*, 2002; AUER *et al.*, 2004, 2010; and SAEY and DE GEER, 2005). The study reported here uses the concentrations measured in more than 1,800 xenon samples taken at five different locations. Data analyzed with two different measurement systems are used here. These are the Système de Prélèvement d'Air Automatique en Ligne avec l'Analyse des radioXénons atmosphériques (SPALAX) from the French Atomic Energy Commission (CEA), France (FONTAINE *et al.*, 2004), and the Swedish Automatic Unit for Noble Gas Acquisition (SAUNA) developed by the Swedish Defence Research Agency (FOI), Sweden (RINGBOM *et al.*, 2003).

SPALAX measurements have collection times of 24 h and were taken at Ottawa and at Yellowknife, both in Canada (STOCKI *et al.*, 2004, 2005). Other sets of SPALAX data have been taken on Schauinsland, a mountain in nearby Freiburg (AUER et al.; 2010), Germany, as well as at Bruyères-le-Châtel, France. Samples taken in Stockholm have collection times of 12 h and were measured with the SAUNA system.

Apart from the samples taken in Ottawa, most of the daily samples have no radioxenon or solely ^{133}Xe, only a few of these samples have high ^{133}Xe concentration and also include ^{135}Xe and/or ^{133m}Xe. ^{131m}Xe is detected with the lowest frequency.
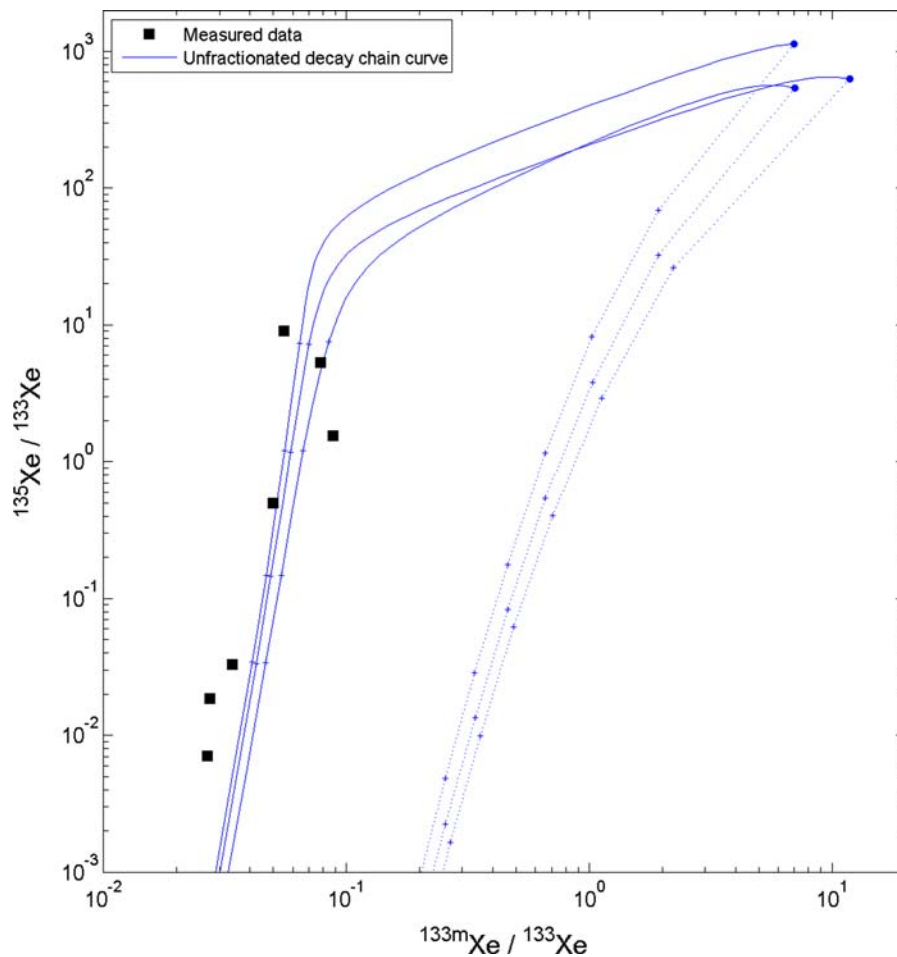
Figure 5
Measured data for very short-term irradiation of HEU targets (RAITH, 2006)

Figure 6 shows the distribution of all measured xenon isotopic activity ratios. They are spread over several orders of magnitude.

The Indian and Pakistani nuclear tests in May 1998 could not be detected by INGE stations because they were not operative. Regarding the claimed nuclear test in North Korea in October 2006, measurements were taken in South Korea by RINGBOM et al. (2007), RINGBOM et al. (2009) and backtracked to the test location by BECKER et al. (2010). Earlier SAEY et al. (2007) presented a study relating a radioxenon detection at the IMS station in Yellow-knife, Canada to the same event.

3. Source Discrimination Based on Single Xenon Isotopic Activity Ratio Relationship

3.1. Isotopic Activity Ratios as a Function of Time

The iodine activity released by nuclear reactors is by at least three orders of magnitude smaller than the discharged xenon activity (see e.g., VAN DER STRICHT and JANSSENS, 2001). Therefore, the xenon isotope concentrations are governed by decay with negligible in-growth.

Figure 7 depicts the change over time of xenon isotopic activity ratios simulated for LWRs. The reactor equilibrium and the simulation maximum are
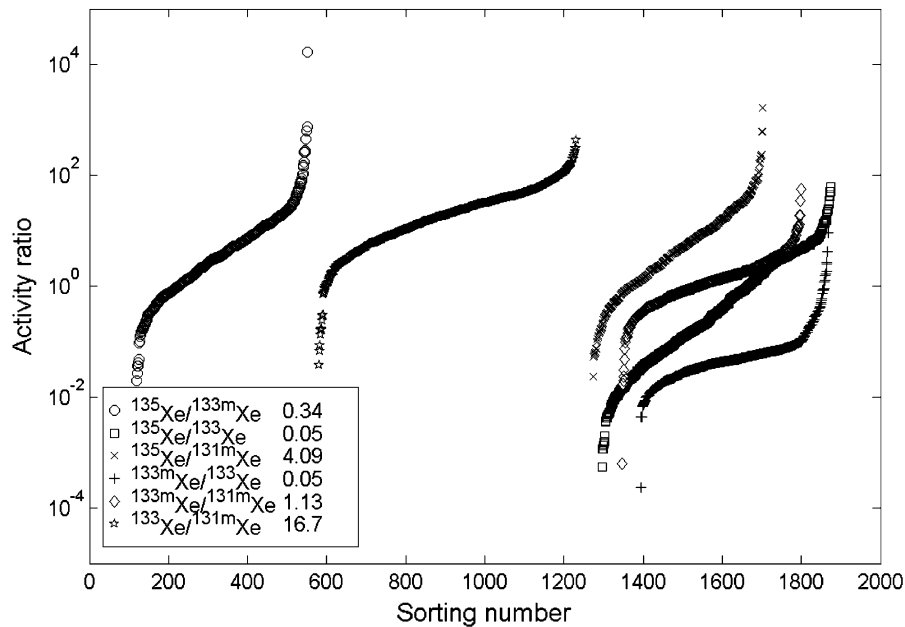
Figure 6
Distribution of atmospheric xenon isotopic activity ratios as measured at different sites of the International Noble Gas Experiment (INGE) in Europe and North America. The logarithmic mean ratios are given in the text box

displayed to mark the band of ratios that can be expected as a result of normal operational releases. This has to be compared to the isotopic ratios resulting from a nuclear explosion.

For fully fractionated releases from underground explosions the evolution in time follows an exponential decrease and will show as a straight line unless ^{133}Xe is involved. For that one of the precursor ^{133m}Xe is present. In this case a bent curve can be seen in the semi-logarithmic graph (see Fig. 7).

3.2. Window of Opportunity for Source Discrimination

For comparison, the change over time of xenon isotopic activity ratios is shown in Fig. 7 for both reactor simulations and releases of underground nuclear explosions. In addition, measured releases at the Nevada Test Site are marked with the reported start time of the release. In general, the largest amount of activity is released shortly after the emission begins. The emission rate typically declines steeply and remains at a low level before it is reported to end.

All isotopic ratios used here have the shorter-lived isotope in the numerator so as to have them decreasing with progressing time with their maximum at time zero. Discrimination of nuclear test releases against reactor discharges in the sense of a test being the sole explanation requires that the isotopic ratio remains higher than the maximum of the reactor discharge band at time zero, because in general the age of the sampled plume is not known. This requirement can be relaxed if the suspected detonation time is known from seismic analysis.

Therefore, a margin for source discrimination is defined by the gap between the lowest possible activity ratio for nuclear explosions and the maximum of the reactor emission simulation. If it exists at all, it not only has a certain width (height of the gap) but also a limited duration, since it takes a certain time until this gap vanishes due to the radioactive decay.

Besides isotopic activity ratios, there are other screening methods that may be based on absolute concentrations and may involve an outlier analysis to classify a measurement as anomalous in comparison to typical atmospheric background at that detector
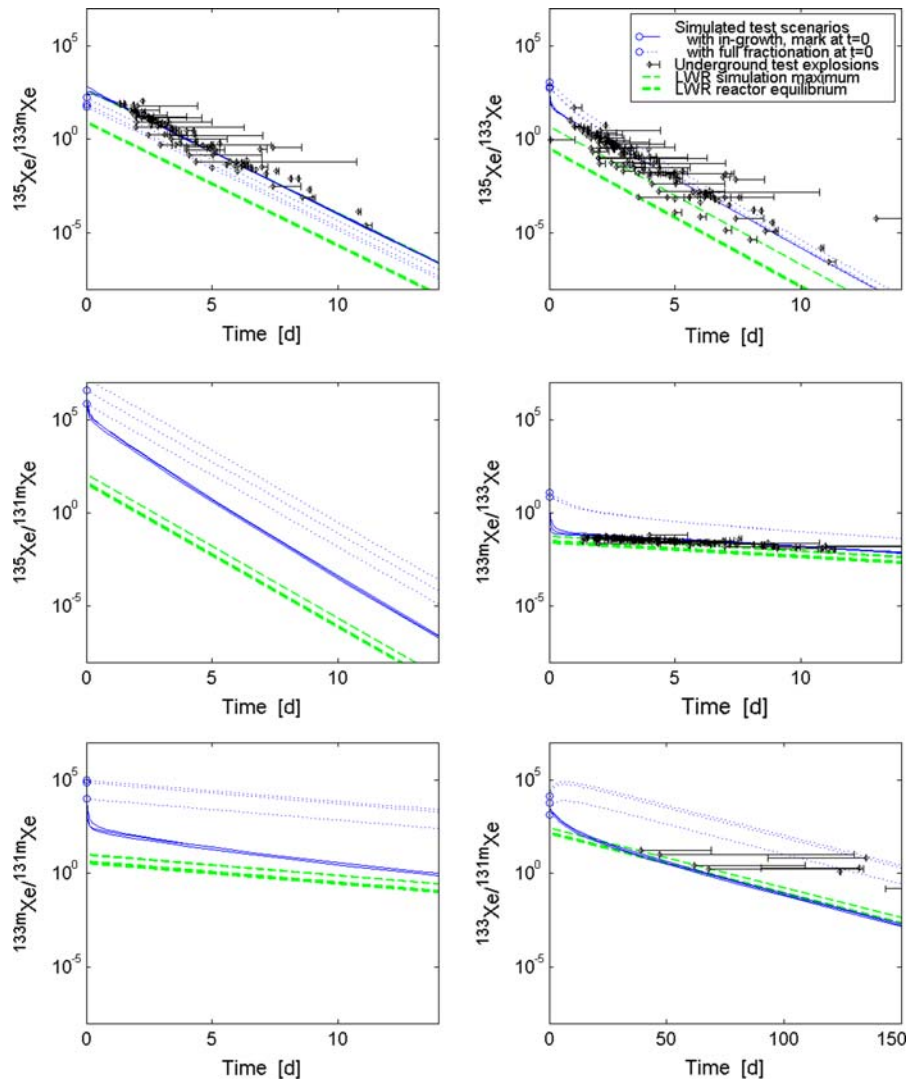
Figure 7

Change over time of isotopic activity ratios of reactor emission simulations compared with nuclear explosion scenarios and Nevada test release data. Note different time scale in the last subplot. The *horizontal bars* indicate the reported duration of the release

site. This is of particular relevance on the southern and in remote areas of the Northern Hemisphere. However, source discrimination may be less complicated and more robust if three or four isotopes are measured and used for source discrimination as described in the following section.

3.3. Single Isotopic Ratios

At the time of a nuclear explosion all combinations of isotopes as shown in Fig. 7 have ratios

that are above typical reactor emissions as indicated by the equilibrium of light water reactors (LWRs). Information relative to the time evolution of the four xenon isotope activities for a 1 kt nuclear test can be found in KALINOWSKI (2010a). For all combinations of isotopes with ^{135}Xe in the numerator it takes less than 5 days and for the activity ratio ^{133m}Xe/^{133}Xe 6 days, before the non-fractionated release from nuclear explosions reaches the reactor equilibrium. For the remaining two plots shown in Fig. 7 it takes significantly longer. The

margin of discrimination would be even better, if fractionation would occur, because in general this results in activity ratios that decrease more slowly. However, reactor emissions may have xenon isotopic activity ratios that are found above the equilibrium. If the reactor simulation maximum is considered, the discrimination method based on single isotopic ratios is less powerful.

Since the ratios decrease with time, any observed isotopic activity ratio could indicate a nuclear explosion. Once a ratio that is specific for a nuclear explosion has declined below the maximum possible for reactor emissions, it loses its unambiguous character. A nuclear reactor emission is never unambiguous. It can always be suspected to be an aged nuclear explosion signature. Therefore, the ratios do not strictly allow for screening out irrelevant cases in general.

4. Source Discrimination Based on Xenon Isotopic Activity Ratio Relationship

4.1. Multiple Isotopic Ratios

To overcome these shortcomings of combinations of two xenon isotopes regarding source discrimination, more information is necessary. The xenon activity concentrations themselves are not widely applicable parameters for this purpose, since they change by dilution during the transport through the atmosphere. However, all isotopic ratios are independent of this effect. They change only by radioactive decay, unless precursors are carried along in the same plume at relevant concentration. Therefore, a third or fourth measured xenon isotope could carry in its ratio to another measured isotope the additional information required for source discrimination. The analysis method proposed here is to use a
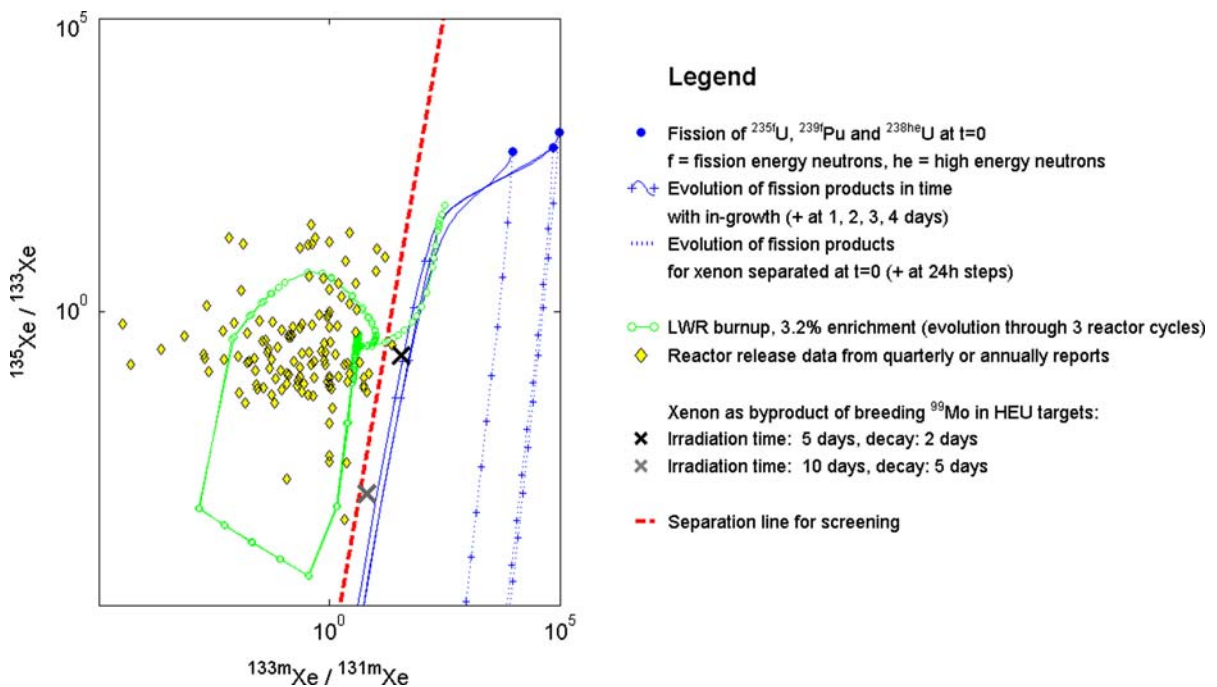

Figure 8

Time-invariant source discrimination based on xenon isotopic activity ratio relationship with reactor emission data for the case in which all four isotopes are measured. The *dashed line* marks the time-invariant screening separation. All isotope ratio relations found above (i.e., left to) this line can be screened out, i.e. the related samples are definitely irrelevant for CTBT verification purposes, because they cannot be explained by a nuclear explosion. All samples that have ratio dependencies found below (i.e., right to) the line might be relevant for CTBT monitoring purposes. The exact location of the separation line is subject to further studies

set of plots that presents the relationship of one isotopic activity ratio to another one. The activity ratios change due to decay and with time they move along a defined line that is straight on a log–log plot in case no precursors are present in the plume. If ^{133}Xe is involved the presence of its precursor ^{133m}Xe causes the line to be slightly bent. It should be noted that this paper assumes that pure air masses from single sources, and in particular no mixing between releases from reactors and with nuclear weapons test emissions.

In Fig. 8, the proposed method is shown for the case that all four xenon isotopes are measured and made use of. Two isotopes are used on the abscissa; the other two are taken for the activity ratio on the ordinate. The legend shown in Fig. 8 also applies to
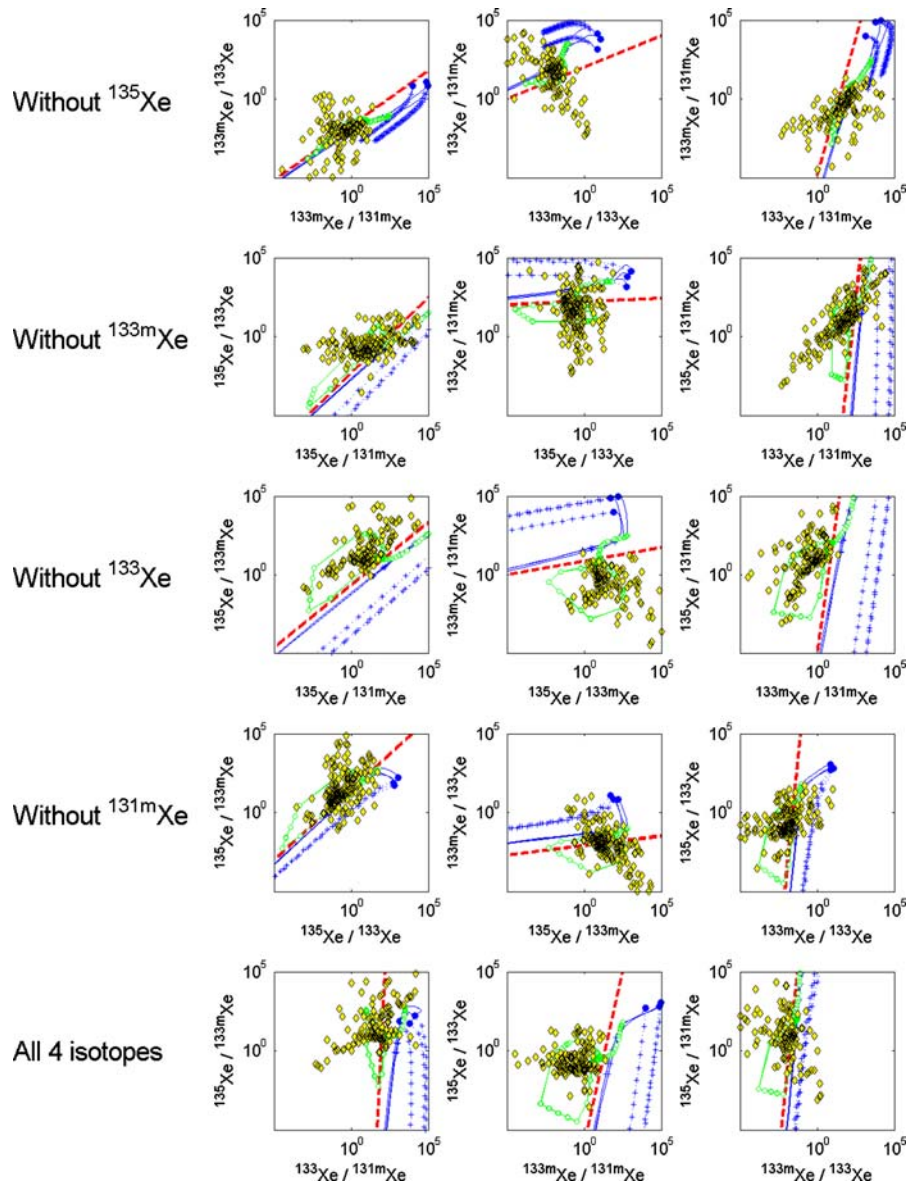

Figure 9
All 15 possible combinations of two xenon isotopic activity ratios without exchange of axes and restricted to those that have the shorter-lived isotope in the numerator. Reported annual reactor emission data are shown together with simulated curves
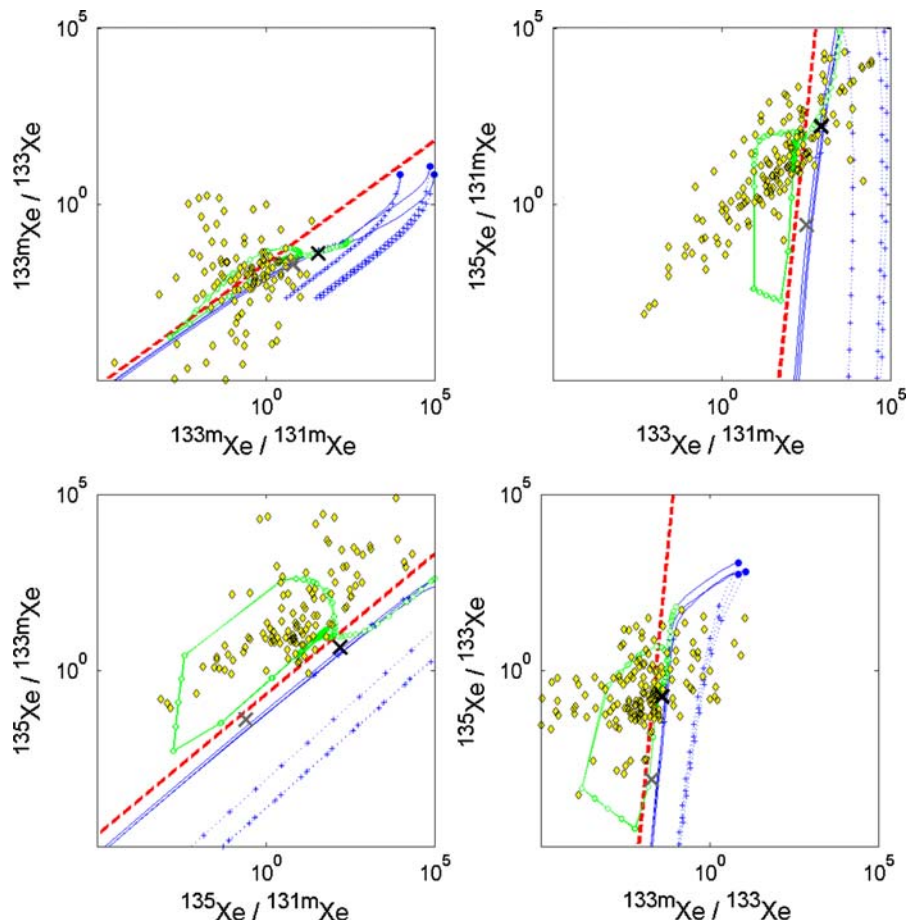
Figure 10
Time-invariant source discrimination based on xenon isotopic activity ratio relationship with reactor emission data for the case in which three out of four isotopes are measured

the following figures. The simulation curves entered here separate the plot area into two distinctive domains. The trajectories for three operational cycles of a nuclear reactor follow a circular pattern in the left half of the plot (KALINOWSKI and PISTNER, 2006). The simulation curves for nuclear explosion scenarios remain in the right half of the plane.

Figure 9 shows all 15 possible xenon isotopic activity ratio combinations without permutation of axes and is restricted to those that have the shorter-lived isotope in the numerator. Each row of sub-plots applies to a different set of xenon isotopes. Most of the 15 plots exhibit overlapping regions for nuclear reactor and explosion scenarios.

Since each row of three plots in Fig. 9 is a different projection of the same three-dimensional space, the plots in one row are not independent from each other. Therefore, it is sufficient to consider only one representative plot for each set of three or four isotopes. The most discriminatory plot with all four isotopes is the one shown in Fig. 8. The selection of best plots for each of the four combinations of three different isotopes is shown in Fig. 10. All further discussions refer to these one plus four plots.

The method for source discrimination proposed here is to define a screening condition as a function of three or four xenon isotopes and to keep it independent from radioactive decay. Two different isotopic ratios are shown on the abscissa and ordinate. Since the axes are on the logarithmic scale, the decay causes the point representing a certain isotopic starting mixture to move on a straight diagonal line

towards minus infinity in both dimensions. In the case of xenon precursors being present in the plume, this would be a bent curve that converges with the straight line defined by decay. The direction of this straight line is determined by the decay constant of the three or four isotopes under consideration.

In each plot a separation line is drawn between the reactor and the explosion domain. Any isotopic activity ratio combination on the separation line moves down along that line with the radioactive decay. Any isotopic activity ratio combination off that line remains on the same side of the line forever. The only exception is ^{133m}Xe/^{133}Xe that could cross the line from a starting point on the nuclear test domain close to the separation line. Radioactive decay changes the isotopic activity ratios along a line that is parallel to the separation line.

This decay invariance offers a new and important quality for source discrimination. It allows for screening out all irrelevant detections because the separation between explosion and non-explosion scenarios is independent from the age of the detected release, i.e., neither the delay before release nor the time for transport through the atmosphere needs to be known.

4.2. Validation of the Multiple Isotopic Relationship Screening Method with Atmospheric Observations Data

The isotopic activity ratio relationship screening method can be validated with atmospheric observation data described in Sect. 2.8. Figure 11 presents the isotopic ratio relations for all air samples in which all four xenon isotopes were measured. Figure 12 shows the same data in the four plots with three out of four isotopes measured, i.e., all samples in which one isotope was not detected are included as well. The error bars shown are statistical contributions only. According to AUER et al. (2004), the systematic error is estimated to be on the order of ±10%. Table 2 shows the screening statistics for all air samples. The screening method works exceptionally well if all four isotopes are measured. Among the cases where three out of four isotopes are measured the case without ^{131m}Xe is the worst. If this isotope is not present or not considered because of a possible background due to its long half-life, the screening method no longer works. If only ^{135}Xe is missing three quarters can be screened out. The screening method is almost 100% successful in the cases without ^{133m}Xe and without ^{133}Xe.
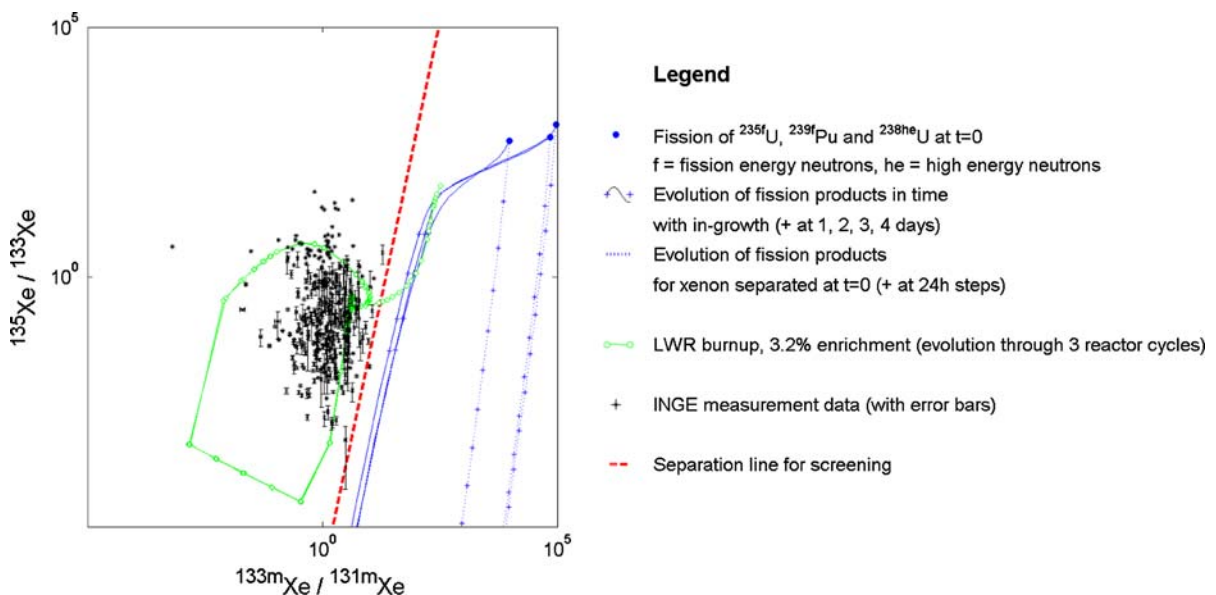

Figure 11
Time-invariant source discrimination for atmospheric measurements with all four isotopes, based on xenon isotopic activity ratio relationship

The station CAX05 in Ottawa is close to one of the isotope production facilities (Chalk River Nuclear Laboratory). Emissions after short-time irradiation of HEU targets have ratios below the screening line are illustrated in Figs. 8 and 10. Therefore one could expect at CAX05 to detect isotopic ratios below the screening line. However, as it can be seen in Fig. 12 this does not happen. The reason for this is twofold. The ^{131m}Xe, due to its long half-life and on-site storage of fission wastes tends to remain in the atmosphere as background from the waste silos and from past emissions. Furthermore, ^{131m}Xe is formed independently at CRL as a by-product of ^{131}I production via irradiation of ^{130}Te targets. Mixing this background into a fresh released plume from isotope production shifts all ratios with ^{131m}Xe in the denominator to lower

values and presumably above the screening line. Therefore it is vital to know the typical local background of ^{131m}Xe at the given station when using the multiple isotopic ratio plot method for screening.

4.3. Quantifying the Separation Line

The optimum position of the separation line for operational use is subject to further studies and depends on user-specific decision criteria. Therefore, an indicative line is constructed which has an optimized slope and a free parameter that allows it to be moved closer or further away from the nuclear test domain.

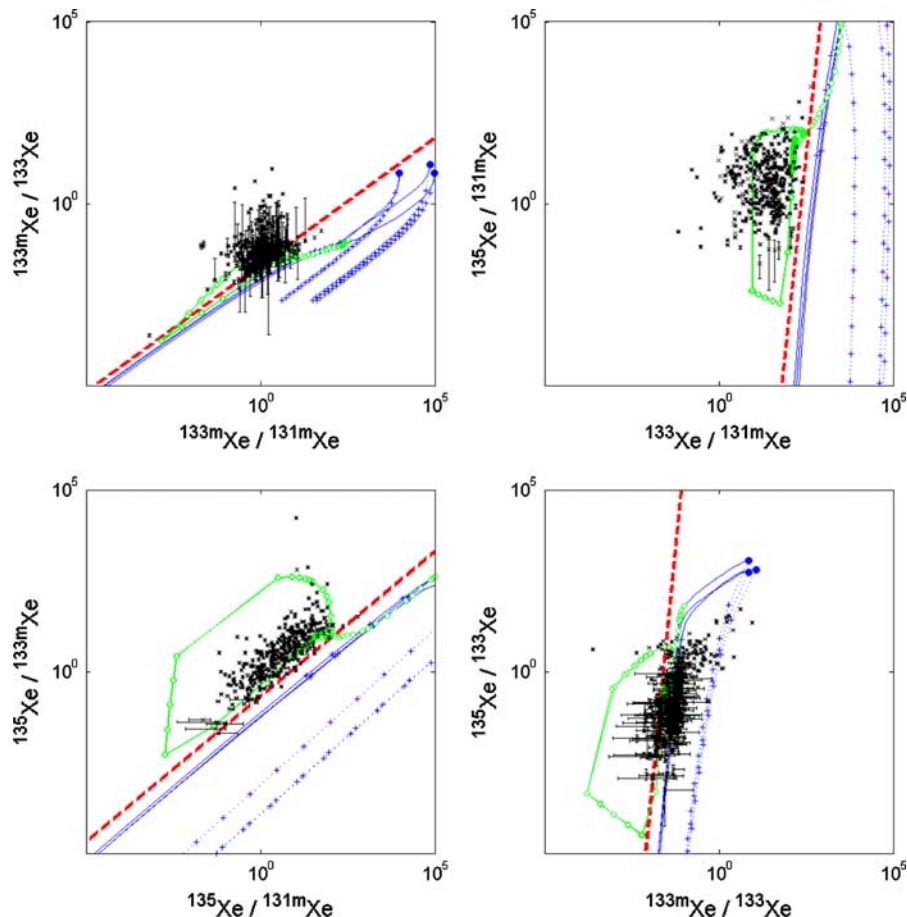The straight lines marking the separation between explosive and power reactor scenarios in the plots of

Figure 12
Time-invariant source discrimination for atmospheric measurement data with three out of four isotopes measured based on xenon isotopic activity ratio relationship

Table 2

Screening statistics for atmospheric observation data

Measured isotope	Total	Above (%) (reactor domain)	Below (%) (test domain)
All four isotopes	286	285 (99.7)	1 (0.3)
All or without ^{135}Xe	361	270 (74.8)	91 (25.2)
All or without ^{133m}Xe	305	305 (100)	0 (0)
All or without ^{133}Xe	288	281 (97.6)	7 (2.4)
All or without ^{131m}Xe	310	39 (12.6)	271 (87.4)

1,875 measurements were made. The column "total" shows how many measurements reported all four or three out of four isotopes. The columns "above" and "below" show how many of them lie above or below the screening line

Figs. 8, 9, 10, 11 and 12 are parallel to a linear least-squares fit to a lower part of the unfractionated decay chain curve. The slope of the separation line is denoted by $m_{a,b,c,d}$ where a, b, c and d indicate xenon isotopes (any of ^{135}Xe, ^{133m}Xe, ^{133}Xe and ^{131m}Xe), a stands for the numerator and b for the denominator of the ratio along the ordinate, c and d for the ratio along the abscissa.

A sample can be screened out, if the entry is found above (or to the left of) this screening line. This can be expressed as the following condition:

$$\log\left(R_{a,b} - \sigma_{a,b}\right) > m_{a,b,c,d} \cdot \log\left(R_{c,d} + \sigma_{c,d}\right) + B_{a,b,c,d}, \tag{1}$$

where the isotopic activity ratio for the xenon isotopes a and b is

$$R_{a,b} = \frac{C_a}{C_b} \tag{2}$$

and the variance of the isotopic activity ratio for the xenon isotopes a and b is

$$\sigma_{a,b}^2 = \left(\frac{\sigma_a}{C_a}\right) + \left(\frac{\sigma_b}{C_b}\right) - 2\frac{\sigma_a \sigma_b}{C_a C_b}\rho \tag{3}$$

with C_a and C_b being the measured atmospheric concentrations, σ_a and σ_b are their measurement uncertainties and ρ_{ab} is the correlation coefficient. Most data considered here satisfy $\rho_{ab} = 0$ as they are analyzed by gamma-spectrometry. The error is overestimated in the few cases where the X-ray lines are involved. In certain cases, one or two concentrations might be replaced by the Minimum Detectable Concentration (MDC, see below). The constant $B_{a,b,c,d}$ is a discrimination margin that can be used to position the screening line at the desired distance to the two separate clusters. The exact position has yet to be determined. Moving the screening line away from the nuclear test domain would render the discrimination more conservative.

Raising both sides of the screening condition to the power of 10 leads to the following relation:

$$R_{a,b} - \sigma_{a,b} > K_{a,b,c,d} \cdot \left(R_{c,d} + \sigma_{c,d}\right)^{m_{a,b,c,d}} \tag{4}$$

with the constant $K_{a,b,c,d}$ being derived from one selected point on the screening line denoted by the index 0:

Table 3

Screening parameters K and m for all cases with three or four xenon isotopes

Measured isotope	Ordinate: a/b abscissa: c/d	Separation parameter K (tentative)	Slope m of the decay line	Number of cases in atmospheric observation data
All four isotopes	^{135}Xe/^{133}Xe ^{133m}Xe/^{131m}Xe	1.00×10^{-6}	4.4388	286
All but ^{135}Xe	^{133m}Xe/^{133}Xe ^{133m}Xe/^{131m}Xe	2.00×10^{-2}	0.6972	75
All but ^{133m}Xe	^{135}Xe/^{131m}Xe ^{133}Xe/^{131m}Xe	3.78×10^{-22}	9.0774	19
All but ^{133}Xe	^{135}Xe/^{133m}Xe ^{135}Xe/^{131m}Xe	2.00×10^{-1}	0.7996	2
All but ^{131m}Xe	^{135}Xe/^{133}Xe ^{133m}Xe/^{133}Xe	3.27×10^{15}	9.7847	24

$$K_{a,b,c,d} = 10^{B_{a,b,c,d}} = \frac{R_{a,b,0}}{R_{c,d,0}^{m_{a,b,c,d}}}. \qquad (5)$$

The separation parameter K is related to the constant B and defines where the screening line is positioned. It is placed above the nuclear test domain in the ratio versus ratio relationship plot. Increasing K moves the screening line up and renders the source discrimination to become more conservative, i.e. it increases the detection probability at the cost of raising the false alarm rate at the same time.

The screening criteria for all cases with three or four different xenon isotopes are given in Table 3. Thick dashed lines mark them for a tentatively selected discrimination margin B in Figs. 8, 9, 10, 11 and 12.

For example, the following criterion for screening out is found to be applicable, if the atmospheric concentrations of all four isotopes are quantified (see dashed line in Fig. 9, bottom middle sub-plot as well as in Figs. 8 and 11):

$$\frac{C_{Xe-135}}{C_{Xe-133}} - \sigma_{Xe-135,Xe-133} > 1$$
$$\times 10^{-6} \left(\frac{C_{Xe-133m}}{C_{Xe-131m}} + \sigma_{Xe133m,Xe-133} \right)^{4.4388}. \qquad (6)$$

As mentioned above, the exact locations of the screening lines are subject to further studies. Especially, further nuclear reactor source and release scenarios as well as the effect of mixing a weak relevant signal with ambient background have to be studied. The operational screening parameters will be selected according to the wanted detection probability and the acceptable false alarm probability.

The main lesson of these plots is that with the exception of fresh fuel being exposed for only a very short time (less than 20–30 days), there are no conditions during reactor operation that generate xenon isotopic activity ratios in the domain of nuclear test explosions in three out of the five different plots of isotopic activity ratio relationships. The combination of isotopes that provides the least opportunity for discrimination between explosions and reactor sources is the one without ^{131m}Xe. The parent of this isomer, ^{131}I, has a half-life of 8.02 days. The other isotopes have shorter-lived precursors with a maximum of 2.19 days (^{133m}Xe) for ^{133}Xe, 20.8 h (^{133}I)

for ^{133m}Xe and 6.61 hours (^{135}I) for ^{135}Xe. Due to the long half-life of its precursor, the presence of ^{131m}Xe in the set of quantified isotopes provides for good separation capability.

4.4. Detection Limit to Substitute Missing Concentration Measurements

Even in sensitivity measurements of atmospheric xenon activity concentrations in ground level air (AUER et al., 2004; SAEY and DE GEER, 2005; STOCKI et al., 2004), the instrumentation used cannot always detect all of the xenon isotopes because two or more of them are at very low concentrations and thus below the detection limits. Therefore, a robust screening method using xenon ratios should be able to handle the circumstances in which only 2 or 3 of the isotopes are measured, and the others are presumed to be either at or below the detection limit.

In the event that only two or three of the xenon isotopes are detected, the isotopic ratio relationship method might still be applicable. If the isotope in the numerator of an isotopic activity ratio is not quantified, the real ratio is to a high probability lower than one gets by replacing the activity with the minimum detectable concentration (MDC). If the isotope concentration in the denominator is replaced by its MDC, the real ratio is larger than the calculated value. The goal for the MDC set by CTBTO guidelines is 1 mBq/m^3 for ^{133}Xe. SPALAX data analyzed with Aatami achieved an MDC of 0.9 mBq/m^3 for ^{133m}Xe and 0.7 mBq/m^3 for ^{131m}Xe (STOCKI et al., 2004). For ^{135}Xe, an MDC of 0.4 mBq/m^3 can be achieved and 0.2 mBq/m^3 for ^{133}Xe. The application of the Bayesian method (ZÄHRINGER and KIRCHNER, 2008) allows extraction an optimum information for the isomers (VIVIER et al., 2009), especially at very low activities.

If only two xenon isotopes are measurable, the ratio relationship method still appears to work by applying the MDC for a third one with the exception of ^{135}Xe and ^{133m}Xe being the measured isotopes. That case should be rare since these are the two shorter-lived isotopes that are most likely those having decayed below the detection limit at the time of the measurement. Table 4 presents the applicability of screening based on two measured xenon

Table 4

Applicability of isotopic activity ratio relationship method for all cases with two measured xenon isotopes and with the MDC being used for one of the non-detected isotopes

Measured isotope	Using MDC for third isotope	Comment: Screening possible Yes/no	Best option	Number of cases in atmospheric observation data
^{135}Xe, ^{133m}Xe	^{133}Xe	No	None	0
	^{131m}Xe	No		
^{135}Xe, ^{133}Xe	^{133m}Xe	Yes	Without ^{131m}Xe	66
	^{131m}Xe	No		
^{135}Xe, ^{131m}Xe	^{133m}Xe	Yes	Without ^{133}Xe or without ^{133m}Xe	0
	^{133}Xe	Yes		
^{133m}Xe, ^{133}Xe	^{135}Xe	Yes (practically)[a]	Without ^{131m}Xe	41
	^{131m}Xe	No		
^{133m}Xe, ^{131m}Xe	^{135}Xe	Yes (practically)[a]	Without ^{135}Xe	0
	^{133}Xe	Yes		
^{133}Xe, ^{131m}Xe	^{135}Xe	Yes (practically)[a]	Without ^{133m}Xe	127
	^{133m}Xe	Yes (practically)[a]		

[a] Theoretically, time independent screening is not possible. However, if from practical considerations the age of the sample can be limited to a reasonable maximum (e.g., 2 weeks), screening is possible

Table 5

Applicability of isotopic activity ratio relationship method for all cases with one measured xenon isotope only and with the MDC being used for two of the non-detected isotopes

Measured isotope	Using MDC for two isotopes	Comment: Screening possible Yes/no	Best option	Number of cases in atmospheric observation data
^{135}Xe	^{133m}Xe, ^{133}Xe	No	None	28
	^{133m}Xe, ^{131m}Xe	No		
	^{133}Xe, ^{131m}Xe	No		
^{133m}Xe	^{135}Xe, ^{133}Xe	No	None	3
	^{135}Xe, ^{131m}Xe	No		
	^{133}Xe, ^{131m}Xe	No		
^{133}Xe	^{135}Xe, ^{133m}Xe	Yes (practically)[a]	Without ^{131m}Xe	875
	^{135}Xe, ^{131m}Xe	No		
	^{133m}Xe, ^{131m}Xe	No		
^{131m}Xe	^{135}Xe, ^{133m}Xe	Yes (practically)[a]	Without ^{133}Xe	3
	^{135}Xe, ^{133}Xe	Yes (practically)[a]		
	^{133m}Xe, ^{133}Xe	Yes (practically)[a]		

[a] Theoretically, time independent screening is not possible. However, if from practical considerations the age of the sample can be limited to a reasonable maximum (e.g. 2 weeks), screening is possible

isotopes with the MDC being used for one of the non-detected isotopes.

Table 5 lists the applicability of the screening method based on only a single measured xenon isotope and with the MDC being used for two of the non-detected isotopes. This still works, if only ^{133}Xe or only ^{131m}Xe are measured. The first is in fact one of the isotopes being most frequently detected alone, because its share in nuclear reactor releases is the highest and also it has the second longest half-life. Giving a sufficiently large source term, ^{131m}Xe might be the last isotope moving below the detection threshold during the transport through the atmosphere, since it has the longest half-life.

5. Conclusions and Implications for Monitoring Nuclear Explosions

Single xenon isotopic activity ratios may not be as well suited for source discrimination as previously thought, unless the explosions are highly fractionated or no reactor sources are present in the region. Their applicability has been overestimated mainly because the large range of these single ratios in reactor emissions was not adequately considered. There is only one single xenon isotopic activity ratio that can be used alone, namely, $^{133m}Xe/^{131m}Xe$. It can be used with confidence for source discrimination for all nuclear testing scenarios if a single release source is assumed. There is some limited potential for the activity ratios without ^{135}Xe. $^{133m}Xe/^{133}Xe$ is the most relevant example, particularly if early fractionation takes place.

Even if a single xenon isotopic activity ratio is not suitable for source discrimination, it may still be valuable for determining the plume age (KALINOWSKI, 2010a) and tracking it back with atmospheric transport simulation in order to investigate a spatial overlap of the backward plume with waveform location predictions related to a suspected nuclear test. This has to be considered when drawing conclusions on the usability of xenon isotopic ratios for nuclear explosion monitoring.

A new method for source discrimination has been proposed here based on the relationship of two different isotopic activity ratios. This requires three or four xenon isotopes to be quantified. In some cases the method works with the detection limit by substituting a missing concentration value, if certain isotopes are not detected. Hence, it is possible to use this method if only two isotopes are detected. A special advantage of this new method is its independence on the time periods elapsed between generation and release as well as between release and detection, i.e., it is dilution and decay invariant. Quantitative screening conditions are derived for all combinations of three or four xenon isotopes.

A proof of concept for this new screening method is given here by validating it against a comprehensive data set. These data include simulations as well as empirical data of nuclear reactors and explosions as well as observations of atmospheric xenon at different locations in Europe and North America.

In particular the conditions and limitations are:

- Suitable combination of isotopes need to be detected
- Background must be precisely known, in particular ^{131m}Xe
- No mixing in of fresh releases from other sources
- Dependent on decision procedure whether or not a nuclear explosion is likely indicated by the ratio (in this paper simply indicated by separation line)
- Excessively large xenon isotopic activity ratio uncertainties

Further work is required in order to put this method in operation. In particular in the light of the detection goals, various uncertainties and possible mixing of a weak relevant signal with ambient background have to be considered. The limitations posed by the need to use three or more xenon activities calls for additional screening methods for the cases of fewer isotopes being quantified. These may be based on a combination of single ratios as well as absolute concentrations and may involve an outlier analysis to classify a measurement as anomalous in comparison to typical atmospheric background at that detector site. This can be supported by atmospheric transport simulations using information about known civilian sources.

Preparatory Commission or any other organisation one of the authors is affiliated with.

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