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STATISTICAL HYPOTHESES AND ASSOCIATED THRESHOLDS FOR CTBT-RELEVANT NUCLEAR EVENT SCREENING BASED ON RADIOXENON ACTIVITY RATIOS MEASURED IN THE INTERNATIONAL MONITORING SYSTEM RADIONUCLIDE NETWORK

Boxue Liu*, Joshua Kunkle, Robin Schoemaker, Mark Prior, Ian Hoffman
Comprehensive Nuclear-Test-Ban Treaty Organization, Vienna, Austria
*Email address: boxue.liu@ctbto.org

ABSTRACT

Noble gas samples are routinely collected at radionuclide stations in the international monitoring system (IMS) noble gas network and the radioxenon activity concentrations in the plume of air passing over IMS stations are estimated accordingly. Anomalous radioxenon activities collected in the samples are mostly caused by emissions from nuclear facilities, but they could also indicate releases from an underground nuclear explosion (UNE). Nuclear events may be screened using the time evolution of isotopic activity ratios of CTBT-relevant radioxenon isotopes. The evolution goes from a detonation to release of an assumed UNE, through atmospheric transport, to sample collections and measurements at IMS stations. Event screening can be performed based on three kinds of plots: the four-radioxenon plot, two-dimensional plots of activity concentrations of paired radioxenon isotopes, and evolution plots of isotopic activity ratios. Based on activity evolution of assumed release scenarios of UNEs as well as distributions of isotopic activity ratios measured at the IMS stations, the thresholds for event screening are updated with respect to the early release scenarios.

INTRODUCTION

The International Monitoring System (IMS) network consists of various detection technologies, including waveform technologies (e.g., hydroacoustic, infrasound, and seismic), as well as technologies specifically engineered to measure airborne radionuclides. Radionuclide detection technologies within the IMS network are further categorized into the detection of radioactive particulates and noble gases (Auer et al, 2004; Goodwin et al, 2023).

Within the International Data Centre (IDC), particulate spectrum categorization encompasses 84 fission and activation products, while noble gas analysis focuses on four radioxenon isotopes (^{131m}Xe, ^{133m}Xe, ^{133}Xe, and ^{135}Xe) as discussed in De Geer (2001). The activities of radioisotopes collected and measured in samples are converted to activity concentrations, assuming a constant concentration profile during the sampling duration. The spectra of the samples obtained from IMS stations provide time-integrated snapshots of each collected sample, complemented by multiple preliminary spectra demonstrating the evolution of the measurement over time. Analysis results are reported in standard IDC products resulting from automatic and interactive analyses.

Activities measured at radionuclide stations within the IMS network are mostly attributed to emissions from established civil nuclear facilities, which produce some of the same non-naturally occurring radionuclides as those produced in nuclear explosions. Anomalous detections therefore can indicate either these known sources, which can mask a potential weaker signal from an actual test, whether announced or not, or a strong nuclear explosion test signal. In either scenario, isotopic activity ratios of CTBT-relevant detections serve not only to discriminate releases from a nuclear explosion source from civilian nuclear activities but also, in the event of an actual nuclear explosion, to estimate the time of detonation under an assumed release scenario (Carrigan et al., 2016, 2020; Kalinowski and Pistner, 2006; Kalinowski et al.,

2010; Kalinowski, 2011; Ringbom et al., 2014). Various methods for determining the isotopic activity ratios and associated uncertainties have been studied with applications in nuclear explosion monitoring (Zaehringer & Kirchner, 2008; Axelsson & Ringbom, 2014; Sloan et al., 2016; Yamba et al., 2016; Galan et al., 2018; Liu et al., 2023).

Isotopic ratios vary with time as can be seen by comparing the activities at the time of release with activity concentrations in the air plume over an IMS station, and finally with activities collected in the sample. It is essential to note that the isotopic ratio of activities collected in the sample is influenced by the sampling duration. Moreover, the isotopic ratio of activity concentrations in the transported air mass serves as a crucial link connecting the two ends in time, namely, from the initial release to the sample measurements (Liu et al., 2023).

The analysis of isotopic activity ratios involves a backward-in-time procedure, starting from sample measurements and tracing back to release scenarios. The first step is to estimate the isotopic ratio of two activity concentrations at the time when the air sample is collected. This estimation relies on the sample collection, spectrum acquisition and analysis, and an assumption regarding the concentration variation during the sampling duration (e.g., 12 hours). Essentially, the activity concentration in the air above a measurement station can be correlated with the released activities from a nuclear explosion through atmospheric transport modeling (ATM) (Wotawa et al., 2003; Kuśmierczyk-Michulec et al., 2021; Maurer et al., 2018, 2022). The gas-dilution factor, used to determine the activity concentration in a measurement based on an initial release of activity, applies to all radioisotopes within the volume of air (the plume) in the simulation.

Characterization of CTBT-relevant nuclear events may use the evolution of isotopic activity ratios over time, which goes from the release of an assumed UNE, through atmospheric transport, to sample collections and measurements. This can be investigated in two ways; activity concentrations at an IMS station can be estimated using an assumed release scenario regarding a UNE, and atmospheric transport modelling. On the other hand, the activities are directly determined by spectral analysis of collected samples and used to estimate activity concentrations in the air passing over an IMS station, often using an assumption of constant concentration during sampling. The isotopic ratios of activities released from the UNE can be related to the isotopic ratios of activity concentrations in the plume of air crossing the IMS station, resulting in a function of the isotopic activity ratio over the time from detonation to sample measurement. This function is used for discrimination of a nuclear test, such as a four radioxenon plot of the activity ratio relationship of $^{135}Xe/^{133}Xe$ versus $^{133m}Xe/^{131m}Xe$, and estimation of the time of detonation (Liu et al., 2023).

The statistical hypotheses behind analysis procedures are different in different stages from sample measurements to event characterization. The first hypothesis is to determine whether radioxenon is detected, H_0: the null hypothesis of detector background; H_1: the alternative of radioxenon detection. The radioxenon is assumed to be detected if the net number of counts is above the decision threshold. The second hypothesis is formulated regarding the radioxenon background at an IMS station: H_0, the null hypothesis of normal radioxenon background; H_1, the alternative of anomalous radioxenon detection. The abnormal concentration threshold is estimated based on the statistical analysis of the previous samples in a specified period, such as 365 days, resulting in two categories of B and C, while Level A is assigned to samples with no radioxenon detection. Finally, discrimination of a nuclear explosion source against releases of nuclear facilities is based on isotopic ratio analysis, e.g. relationship plots of four or three radioxenon isotopes. Both Level C and B samples in the IDC sample categorization scheme are used. The hypothesis is formulated: H_0, the null hypothesis of releases from nuclear facilities; H_1, the alternative of a nuclear explosion source. The overlap between the discrimination line and lower and upper limits of the coverage interval of isotopic ratios is tested.

This work focused on statistical hypotheses of event screening by isotopic ratio analysis. Based on activity evolution of assumed release scenarios of UNEs as well as distributions of isotopic activity ratios measured at the IMS stations, the thresholds of the isotopic activity ratios for event screening are updated.

ISOTOPIC ACTIVITY RATIO ANALYSIS

Activities Released from a Nuclear Explosion

The evolution of isotopic radioactivity from a nuclear explosion to the release of gases into the atmosphere is a complex process. It is related not only to radioactive decay chains, but also to the convection and diffusion of xenon gases from the explosion chimney to the ground surface in the case of an underground nuclear explosion (UNE) (Bateman, 1910; Kalinowski, 2011; Carrigan et al., 2016, 2020; Liu et al., 2023). The isotopic ratio of released activities is defined by Eq. (1).

$$R(t_1) = A_2(t_1)/A_1(t_1), \quad (1)$$

where $A_1(t_1)$ and $A_2(t_1)$ are the activities (Bq) of two isotopes at the release time t_1 since the time of detonation; $R(t_1)$ is the ratio of the released activities.

Activity Concentrations in the Plume at an IMS Station

For simulating the end-to-end evolution of an isotopic signature, a model must consider radionuclides released from ground or water into the air at t_1 that are transported by winds and subject to atmospheric mixing and radioactive decay. An ATM model is required to track these radionuclides, calculating their travel paths from the release location to various reception locations worldwide. To achieve this, the IDC employs the 3D Lagrangian particle dispersion model FLEXPART (Stohl et al., 1998, 2005; Stohl and Thomson, 1999), which can simulate the transport of tracers both backward and forward in time. The model run computes the global source–receptor relationship, linking the starting and ending points of the transport of a unit tracer using a source–receptor matrix. The resulting Source-Receptor Sensitivity (SRS) field is expressed as the product of a spatial-temporal source at discrete locations and time intervals: $C_{ijn} = S_{ijn} \times M_{ijn}$. In this expression, C represents the attributed concentration field of the tracer (Bq m^{-3}), S is the source attribution (Bq), and M is the source–receptor matrix acting as a dilution field (m^{-3}) and the subscripts i, j, n are the discrete space and time indices. Source attribution, such as determining the origin of radioisotopes based on factors like half-lives, is efficiently performed through matrix multiplication in a post-processing step (Wotawa et al., 2003; Stohl et al., 2005; Becker et al., 2007; Kuśmierczyk-Michulec et al., 2021).

The output of an ATM model run provides a tracer dilution matrix M, from which the dilution value in the matrix element collocating with the station can be read. The time of collection termination is denoted by t_2. This computation uses both $C(t_2)$, representing the activity concentration measured at the station, and the simulated dilution value in $M(t_2)$. The activity, $A(t_2)$, at the time of collection, t_2, is expressed as a quotient:

$$A(t_2) = \frac{C(t_2)}{M(t_2)}. \quad (2)$$

In the most simplified approach, the model is limited to one source and one release time, making the term in Eq. (2) a scalar. However, when multiple sources and numerous possible release time intervals are considered, Eq. (2) transforms into a matrix operation. Developing an analytical solution for more complex scenarios is still a challenge. Using Eq. (2), the isotopic activity ratio at the time collection ends can be calculated by

$$R(t_2) = \frac{A_2(t_2)}{A_1(t_2)} = \frac{C_2(t_2)/M(t_2)}{C_1(t_2)/M(t_2)} = \frac{C_2(t_2)}{C_1(t_2)}. \quad (3)$$

Regarding the activity evolution, the isotopic ratio in Eq. (3) is defined by the released activities of the source term. However, it is estimated based on activity concentrations in the air mass passing through the sampling location.

The relationship between the isotopic activity ratio at the release site, defined by Eq. (1), and the ratio at the IMS station, expressed in Eq. (3), is established through the ATM assumption expressed in Eq. (2). In this context, the ATM considers only the effect of dilution and that the effect of radioactive decay is calculated a posteriori.

Activities Collected in a Sample at the IMS Station

The collected activity in a sample is expressed by Eq. (4) under the assumption of independent decay and constant concentration.

$$A_s(t_2) = C(t_2)V_s\frac{1-e^{-\lambda\tau_c}}{\lambda\tau_c}, \quad (4)$$

where V_s is the air volume collected (m^3), with the subscript s indicating the activity collected in the sample, λ is the decay constant (s^{-1}), τ_c is the duration of collection (s), and $A_s(t_2)$ is the activity collected in the sample at the end of collection (t_2). $C(t_2)$ is constant in the sampling duration, which could be the average concentration from the ATM simulations.

Using Eqs. (3) and (4), the isotopic ratio of activity concentrations at the end of collection is related to the isotopic ratio of activities collected in the sample, $R_s(t_2)$, by Eq. (5)

$$R_s(t_2) = \frac{A_{2_s}(t_2)}{A_{1_s}(t_2)} = R(t_2)\frac{\lambda_1}{\lambda_2}\frac{1-e^{-\lambda_2\tau_c}}{1-e^{-\lambda_1\tau_c}}. \quad (5)$$

Evolution of Isotopic Activity Ratios from Release to Sample Measurements

The relationship between activities released from a nuclear event, activity concentrations in the plume passing over an IMS station, and activities collected in a sample at the IMS station is established through an analysis procedure. The procedure encompasses the activity evolution from an assumed UNE through ATM simulations to sample measurements. The activity concentrations in the plume play a crucial role in connecting the release and sample measurements. On one hand, activities released from a nuclear explosion can be estimated based on an assumed scenario, providing predicted activity concentrations at a station through forward ATM simulations. On the other hand, activities collected in the sample are determined through spectral analysis, and the accompanying activity concentrations are estimated under the assumption of the concentration profile during sampling. As mentioned earlier, the isotopic ratio of activities collected in the sample differs from that of activity concentrations in the plume, as shown in Eq. (5) for independent decay chains. The isotopic ratios of activity concentrations in the plume of air can be estimated based on activities measured in the sample through a backward procedure.

Event Screening Based on Isotopic Ratios

Event Screening Flags in Sample-Specific Radionuclide Reports

Xenon flags for event screening are based on Bayesian approach estimating the upper and lower limits using Gaussian distribution. Lower limits of isotopic activity ratios are used as event screening flags in the IDC radionuclide products, such as $^{135}Xe/^{133}Xe$ >5, $^{133m}Xe/^{133}Xe$ >0.3 and $^{133m}Xe/^{131m}Xe$ >2 (Zaehringer and Kirchner, 2008), and $^{133}Xe/^{131m}Xe$ >1000. The null hypothesis H_0 is nuclear explosion release when the isotopic ratio is above the threshold. This is a very simple statistical model, i.e., an overlap check with the coverage probability of 95%.

Four Radioxenon plot discriminating nuclear explosion

The characterization of a fission event can be based on isotopic activity ratio analysis of CTBT-relevant radionuclide observations at IMS stations and expected releases from nuclear explosions. The source term of the radioisotopes generated by a nuclear explosion is simulated by mathematical modelling of the activity evolution after the detonation time. Event screening of a nuclear explosion from releases of civil facilities is performed based on isotopic ratio analysis, e.g., four radioxenon plot of $^{135}Xe/^{133}Xe$ versus $^{133m}Xe/^{131m}Xe$ (Carrigan et al., 2016, 2020; Kalinowski and Pistner, 2006; Kalinowski et al., 2010; Kalinowski, 2011).

Combined Analysis Between Concentrations and Isotopic Ratios

The $^{133}Xe/^{131m}Xe$ activity ratios of the three level C samples at RN38 related to the 2013 event of the Democratic People's Republic of Korea (DPRK) between 0.1-1 are within the range of the 'normal' observed values at RN38 during the selected period, including the ratios at RN38 resulting from the Fukushima Daiichi nuclear accident. However, the activity concentrations of both ^{131m}Xe and ^{133}Xe in the three level C samples associated with the 2013 DPRK event, in a 2D plot of ^{131m}Xe and ^{133}Xe concentrations, lie outside of the domain of the general radioxenon background at RN38 as well as below the ones from the Fukushima Daiichi nuclear accident. Therefore, both values of isotopic activity ratios and associated activity concentrations may be instrumental to scrutinize with respect to assumed scenarios (Liu et al., 2023).

Thresholds of isotopic ratios for event screening

Thresholds of isotopic ratios based on UNE release scenarios

The current Bayesian limits for event screening flag in the IDC radionuclide reports are estimated with a given coverage interval of 95% under the assumption of Gaussian distribution, see thresholds in Table 1. It was found that the two thresholds of $^{135}Xe/^{133}Xe$ and $^{133m}Xe/^{133}Xe$ do not work. As shown in Figure 1 and Table 2, the ratio of $^{133m}Xe/^{133}Xe$ is decreasing to the threshold of 0.3 less than 1 day and while the other one of $^{135}Xe/^{133}Xe$ reach the threshold of 5 in between 1 and 2 days. However, a plume of air transporting from a release site to a IMS radionuclide station might take one day to a few days (Ringbom et al., 2014; Carrigan et al., 2016, 2020; Liu et al., 2023).

Let's take 5 half-lives to estimate thresholds of isotopic ratios. The thresholds are estimated approximately based on the shorter half-life of the pair isotopes in numerator. For ^{135}Xe, the threshold is selected as 1 by ca. 2 days due to the half-life of 9.14 hours, as shown in Table 2. The new thresholds for the other three pairs in Table 2 are estimated based on ^{133m}Xe and ^{133}Xe on ca. 10 and 20 days respectively.

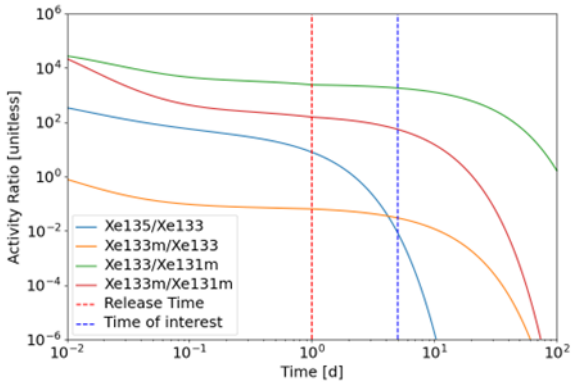

Figure 1 Time evolution of isotopic ratios after a UNE (full ingrowth of U235f)

Table 1 Suggested thresholds of Isotopic activity ratios based on the release scenarios of full ingrowth U235f

	$^{133m}Xe/^{131m}Xe$	$^{133}Xe/^{131m}Xe$	$^{133m}Xe/^{133}Xe$	$^{135}Xe/^{133}Xe$
Current	2	1000	0.3	5
Suggestion	10	500	0.01	1

Table 2 Isotopic ratios on given days after a UNE (full ingrowth U235f)

Days	$^{133m}Xe/^{131m}Xe$	$^{133}Xe/^{131m}Xe$	$^{133m}Xe/^{133}Xe$	$^{135}Xe/^{133}Xe$
1	155	2410	0.0645	7.94
2	118	2250	0.0525	1.29
5	55.9	1840	0.0303	0.00937
10	13.3	1240	0.0108	7.53E-7
20	1.26	634	0.00198	1.38E-13

The abnormal thresholds of isotopic ratios based on radioxenon background

$^{133}Xe/^{131m}Xe$

The isotopic ratios of $^{133}Xe/^{131m}Xe$ were measured in the samples from two IMS stations of JPX38 and RUX58 related to DPRK2013 event (Ringbom et al., 2014; Carrigan et al., 2016, 2020; Liu et al., 2023). As a case study, a distribution of isotopic ratios of $^{133}Xe/^{131m}Xe$ measured at the IMS station USX75 in 2014 to 2023 is given in Figure 2. The ratio of a 95% quantile is ca. 30, much lower than the threshold of either the old value of 1000 or the new suggestion of 500.

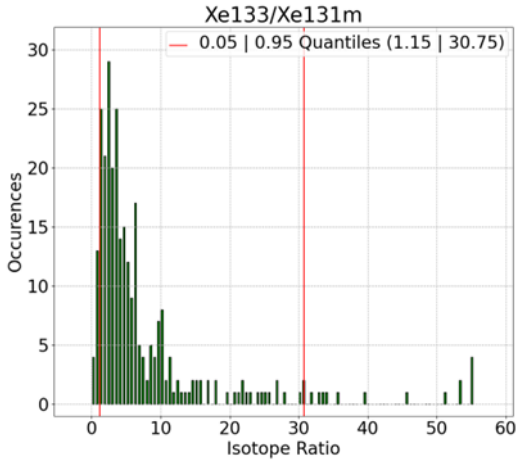

Figure 2 Distributions of isotopic ratios $^{133}Xe/^{131m}Xe$ measured at USX75 in 2014 – 2024.

4Xe-Plot of $^{135}Xe/^{133}Xe$ versus $^{133m}Xe/^{131m}Xe$

A four-xenon plot of the distributions of routine IMS xenon samples at USX75 in 2014 to 2023 is given in Figure 3. The distributions of isotopic ratios of two pairs are given in Figure 4. For $^{135}Xe/^{133}Xe$, more than 5% is above the threshold of 5 (old) or 1 (new). For $^{133m}Xe/^{131m}Xe$, more than 5% is above the old threshold of 2 but less than 5% for the new suggestion of 10. However, all the detected ratios are in the civil domain, i.e., the radioxenon background at the IMS station USX75.

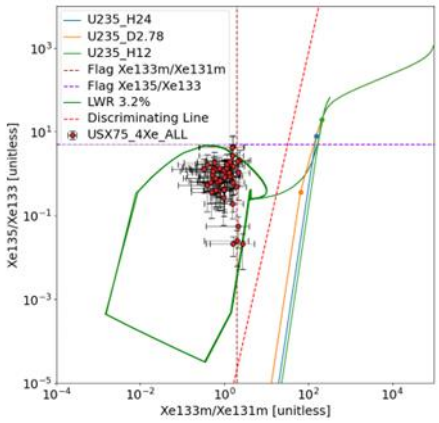

Figure 3 Four xenon plot of isotopic ratios measured at USX75 in 2014 – 2024

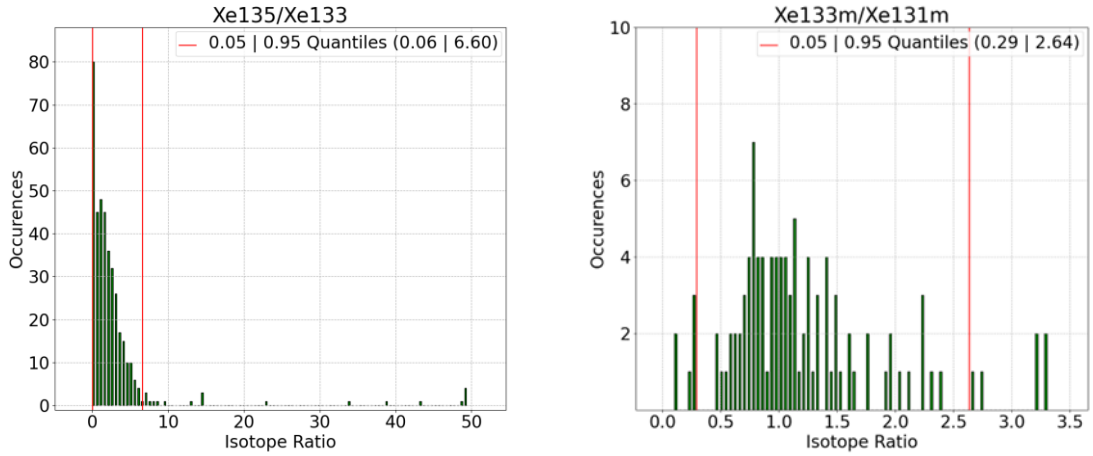

Figure 4 Distributions of isotopic ratios measured at USX75 in 2014 – 2024. Left: $^{135}Xe/^{133}Xe$; Right: $^{133m}Xe/^{131m}Xe$.

SUMMARY

Isotopic activity ratios of fission products detected at IMS radionuclide stations are used for characterization of a CTBT-relevant fission event. For both discrimination of a nuclear test and estimation of the detonation time, the isotopic ratio at the time of collection stop is related to activity concentrations in the plume of air. The function of the isotopic activity ratio over the time from detonation to sample measurement is used for discrimination of a nuclear test, such as a four radioxenon plot of the activity ratio relationship of $^{135}Xe/^{133}Xe$ versus $^{133m}Xe/^{131m}Xe$, and estimation of the time of detonation.

The thresholds of the isotopic activity ratios for event screening are updated with respect to UNE scenarios related to early releases after given days, ca. 5 half-lives of the isotopes in numerators. On the other hand, the abnormal thresholds of radioxenon background are based on the distributions of isotopic ratios measured at CTBT measurement sites with given false positive of 5%. This approach needs to be investigated further.

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