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ALGORITHMS AND ASSOCIATED HYPOTHESIS TESTING APPLIED TO NOBLE GAS SPECTRUM ANALYSIS RESULTS AND EVENT CHARACTERIZATION REGARDING CTBT-RELEVANT NUCLEAR EVENTS

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ABSTRACT

Radionuclide stations in the international monitoring system (IMS) network of the Comprehensive Nuclear-Test-Ban Treaty (CTBT) Organization (CTBTO) routinely collect air samples and assess activity concentrations. Activities collected in samples are often caused by emissions from nuclear facilities, but they could also indicate a noble gas release from a nuclear explosion. Characterization of CTBT-relevant nuclear events may use the evolution of isotopic ratios over time, which goes from the release of an assumed nuclear explosion, through atmospheric transport, to sample collection and measurements. This work outlines the statistical hypotheses behind analysis procedures 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 nuclear explosion source. The overlap between the discrimination line and lower and upper limits of the coverage interval of isotopic ratios is then tested.

INTRODUCTION

The IMS network is comprised of several types of detection technologies including waveform (hydroacoustic, infrasound, seismic) and radionuclide. Measurement of radionuclides in the IMS is used to confirm whether an event detected by the waveform technologies was of nuclear origin, and as a primary detection technology if there are events that have no waveform signature measured by the IMS (Kalinowski et al, 2020).

The radionuclide detection technologies are further broken down into the detection of radioactive particulates and noble gases. There are 80 planned particulate systems and 40 noble gas systems. Of the Noble gas systems, there are two main types; pure gamma spectra are acquired by SPALAX systems with HPGe detectors, and beta/gamma coincidence spectra taken by SAUNA II systems with NaI(Tl) and plastic scintillation detectors. There are 84 fission and activation products used for particulate spectrum categorisation within the International Data Centre (IDC), and four radioxenon isotopes (^{131m}Xe, ^{133m}Xe, ^{133}Xe and ^{135}Xe) for noble gas. The spectra of the samples from IMS stations are a time-integrated snapshot of each collected sample (although there are also multiple preliminary spectra, showing the evolution of the measurement). Radioisotope activities collected and measured in samples are converted to activity concentrations under an assumption of constant concentration profile in the sampling duration. Analysis results are reported in standard IDC products as output from automatic and interactive analyses. Some samples, such as those that contain potentially abnormal activity concentrations of CTBT relevant radionuclides, can be sent to IMS laboratories for re-analysis (CTBTO, 2020).

Radioxenon isotopes measured at radionuclide stations of the IMS may indicate releases from underground nuclear explosions (UNEs) but are often caused by emissions from nuclear facilities. 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).

Basically, there are two kinds of testing decisions when interpreting measurement results at IMS stations. One is the decision whether a radioxenon isotope(s) is(are) detected in IMS samples, i.e., radioxenon detection. The other one is the decision whether the detected radioxenon isotope(s) is(are) caused by a nuclear explosion, i.e., characterization of the CTBT relevant nuclear event. This work outlines statistical hypotheses behind analysis procedures from sample measurements to event characterization.

PROCEDURES FROM IMS MEASUREMENTS TO IDC SPECTRUM ANALYSES AND EVENT CHARACTERIZATION

Radionuclide Measurements in the IMS Network

Eighty radionuclide monitoring stations within the IMS host particulate samplers. Currently, there are two types of particulate systems on the network, manual and automatic. These both currently operate on the same collection, decay and measurement cycle (Goodwin et al., 2023). Forty of these stations, in addition to particulate monitoring, will also host the equipment and means to measure Noble gasses, specifically Xenon. Currently on the IMS, there are two types of Noble Gas systems, e.g., SAUNA II and III and SPALAX. The current version of the SPALAX on the network has a 24-hour collection cycle. The SAUNA II uses a plastic scintillator / Sodium Iodide beta-gamma coincidence detector, with the air sampled for 12 hours. The SAUNA III system operates with two parallel 6-hour cycles and can therefore achieve 4 samples per day.

IDC spectrum analyses

The spectrum analysis method of single channel analyser curve (SCAC) was developed for gamma spectrum analysis of particulate samples. It is a unique algorithm for automatic processing of IMS gamma spectra. The concept of the SCAC method is based on Currie's approach regarding the estimation of critical limit, in addition of the critical level curve (LCC) and baseline (B). However, the uncertainty estimation is based on Bayesian statistics.

The net count calculation (NCC) method is used in analysis of 2D beta/gamma coincidence spectra, quantifying the presence of ^{131m}Xe, ^{133m}Xe, ^{133}Xe and ^{135}Xe in noble gas samples from beta-gamma coincidence measurement systems. Along with new generation systems, different variations of the NCC method were developed. Net numbers of counts are the same in between, but the associated uncertainties might be different with respect to analysis algorithms (Liu et al., 2023).

Atmospheric transport modelling

Atmospheric transport modelling (ATM) simulations add value to the radionuclide analysis by estimating the path of particulates or noble gases through the atmosphere, back to their possible source regions. The IDC aims to establish a world-class ATM system to meet the needs of an integral CTBT verification system. IDC's ATM pipeline includes four major elements: acquisition of meteorological data, modelling, post-processing, and visualisation (Kuśmierczyk-Michulec et al., 2021). The current operational ATM pipeline is based on a Lagrangian particle dispersion model driven by global meteorological fields provided by the European Centre for Medium-Range Weather Forecasts (ECMWF) and the National Centres for Environmental Prediction (NCEP) at a resolution of 0.5 degrees. For each sample at each radionuclide measurement station, the pipeline computes global source-receptor sensitivity (SRS) data for a two-week timeframe (back in time). This backward mode yields daily files that include information about geocoordinates (latitude, longitude), time step, and dilution values, pointing the receptor to probable regions of interest for source localisation. However, in the case a source location is known beforehand, say for an announced nuclear test by the DPRK or the Fukushima nuclear accident, both historical forward modelling and near real-time forecasting are possible in ATM's forward mode, enabling predictions as to which IMS radionuclide stations are likely to be affected by a potential radioactive release in the transported plume. A final step utilises both ATM's backward and forward output by visualising several products in WebGrape (Web-connected Graphics Engine.

Characterization of CTBTO-Relevant Nuclear Events

Activity ratios of CTBT-relevant radionuclides detected in particulate and noble gas samples can be used to discriminate a nuclear explosion source against the releases originating from other nuclear facilities. In case all four radioxenon isotopes are detected, the most discriminating plot is the activity ratio relationship between ^{133m}Xe/^{131m}Xe and ^{135}Xe/^{133}Xe. An important feature is that observation data can be mapped onto the chart for distinguishing underground nuclear explosions (UNEs) from civilian applications without knowing the detonation time. This approach can only be applied to an early release, e.g., less than a few days, due to a short half-life of ^{135}Xe (9.14 h). For all combinations of isotopes with ^{135}Xe in the numerator it takes less than five days before the non-fractionated release from a nuclear explosion reaches chemical equilibrium (Kalinowski et al., 2010; Kalinowski, 2011).

DISTRIBUTION OF ACTIVITY CONCENTRATION FOR EACH SAMPLE

Statistical models between conventional and Bayesian statistics

For repeated measurements, the mean and deviation are estimated, see Figure 1. For a single measurement of each sample, the PDF of the Poisson distribution, which was derived from conventional statistics, is used for uncertainty estimation or Monte Carlo calculation. The variance of the gross number of counts of a ROI, such as ROI-3 and ROI-5, for a given sample spectrum, will be estimated as the gross number of counts itself.

Detector Background

The hypothesis testing is to determine whether a radioxenon detection is caused by the detector background. The two hypotheses in Table 1 are formulated accordingly:

- H_0: the null hypothesis of detector background.
- H_1: the alternate hypothesis of radioxenon detection.

Here, the decision threshold, i.e., critical limit (LC), is used. For example, the net number of counts for the ROI-5 can be expressed as $x_5 = C_5 - B_5$, in which C_5, B_5 and x_5 are the gross number of counts, detector background counts and net number of counts in the ROI-5 (associated to ^{131m}Xe) respectively, resulting in a Skellam distribution, see Figure 2. Applying Bayesian statistics, a distribution of non-negative net numbers of counts can be derived, resulting a non-zero net number of counts when $C_5 = B_5$, see Figure 3.

The net number of counts in the ROI-5 can be interference by the ROI-3, e.g., $x_5 = C_5 - B_5 - R_{35}x_3$, where $x_3 = C_3 - B_3$ is the net number of counts in the ROI-3 (associated to ^{133}Xe). In the conventional statistics, when $C_5 = B_5$ and $C_3 = B_3$, the mean values of x_3 and x_5 should be zero. However, in Bayesian statistics, a non-zero mean value of x_3 will result in a negative offset for x_5, see Figure 4 (left). Consequently, the decision threshold will be increased, see Figure 4 (right).

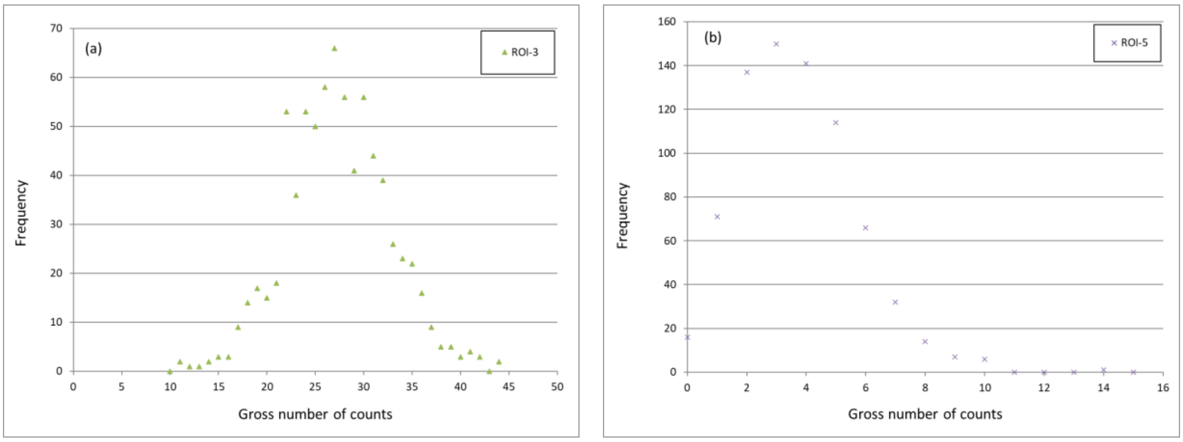

Figure 1 The distribution of gross numbers of counts for ROI-3 and ROI-5 for JPX38_004 in the NCC method in repeated gas background spectra, which follows the Poisson distribution.

Table 1 The null hypothesis and alternative one on the detector background against the radioxenon signal

	True detector background	True radioxenon detection
Do not reject H_0	Correct detector background	False negative
Reject H_0	False positive	Correct radioxenon detection

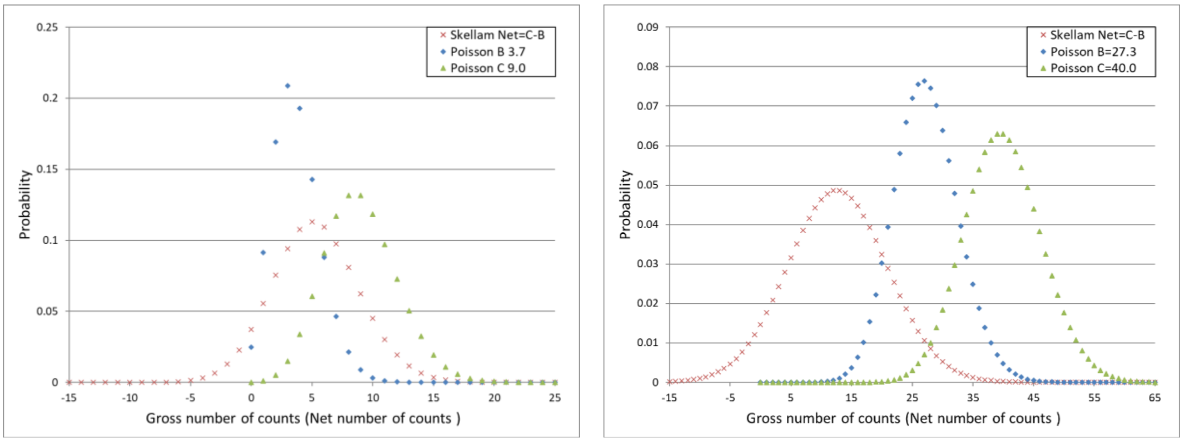

Figure 2 Skellam distribution of the difference of two Poisson distributions (the gross numbers of counts of detector background and a sample above the LC for JPX38_004: Left for ROI5; Right for ROI-3.

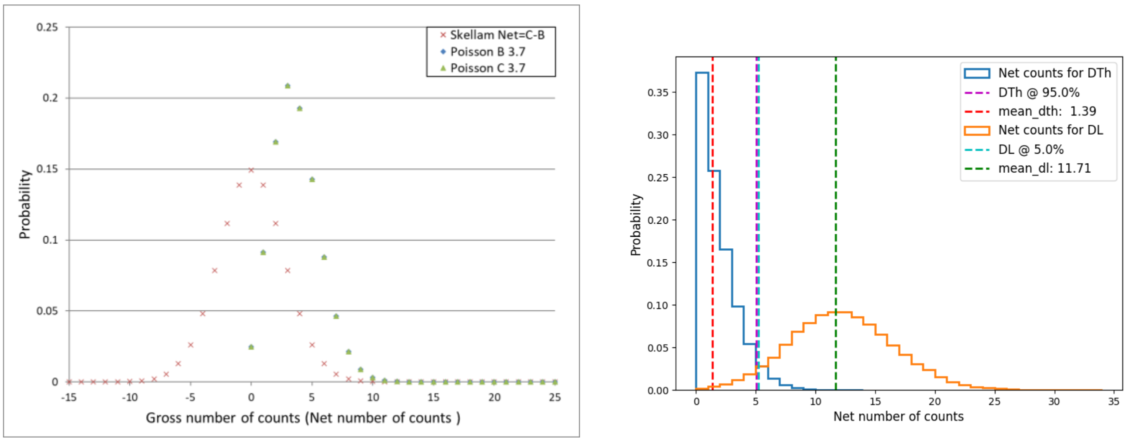

Figure 3 Distribution of net numbers of counts for ROI-3 for the detector background and the distribution for estimating detection limits based on Bayesian statistics (no interference from ROI-3). Left: Conventional statistics: Right: non-negative net number of counts.

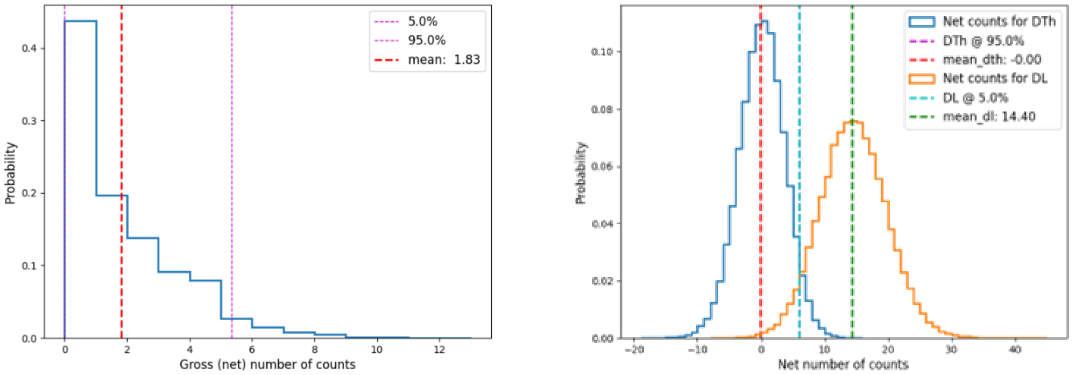

Figure 4 Distribution of net numbers of counts in ROI-5 by applying the non-negative to ROI-3, resulting in a negative offset (left). Distribution of the net numbers of counts in ROI-5 for estimating decision threshold and detection limit, resulting in a larger decision threshold (right).

Subtraction of memory effect

In case gas background measurements are available, such as for SAUNA systems in the IMS radionuclide network, the associated gas background spectrum can be analyzed in the same way as the spectrum of the sample itself. And the same null hypothesis regarding the detector background above is applied. The subtraction of memory effect should be performed only if radioxenon is detected in the gas background spectrum. Then the corrected activity concentrations will be used in the subsequent analysis.

The gas background measurements are somehow repeated measurements for the detector background in case the memory is not present. The correction of memory effect can be performed based on the trend analysis of the numbers of ROI counts in the gas background spectra at IMS stations in a specific measurement period, see Figure 5. Based on the fitted decay curve, the gross number of counts for the gas background can be estimated, along with the uncertainty of the fitted error. It is assumed the gross numbers of counts follow a Poisson distribution based on the priori knowledge about the gas background for memory-free measurements. Therefore, for the uncertainty of the fitted gross number of counts, there will be two components, the fitted error and uncertainty related to the Poisson distribution.

DISTRIBUTION OF DAILY ACTIVITY CONCENTRATIONS

The noble gas systems in the IMS radionuclide network are operated routinely. The xenon detections in daily samples originate from routine and non-routine releases from nuclear facilities, which includes the release sources from nuclear accidents. The hypothesis testing is to determine whether the radioxenon detection is caused by the normal radioxenon background. A criterion discriminating a nuclear explosion source with respect to the radioxenon background at the IMS station needs to be established. The question is which quantity should be used, activity concentration or isotopic ratio of activity concentrations, resulting in two different approaches, anomaly radioxenon detections using concentrations and discrimination using isotopic ratios, respectively.

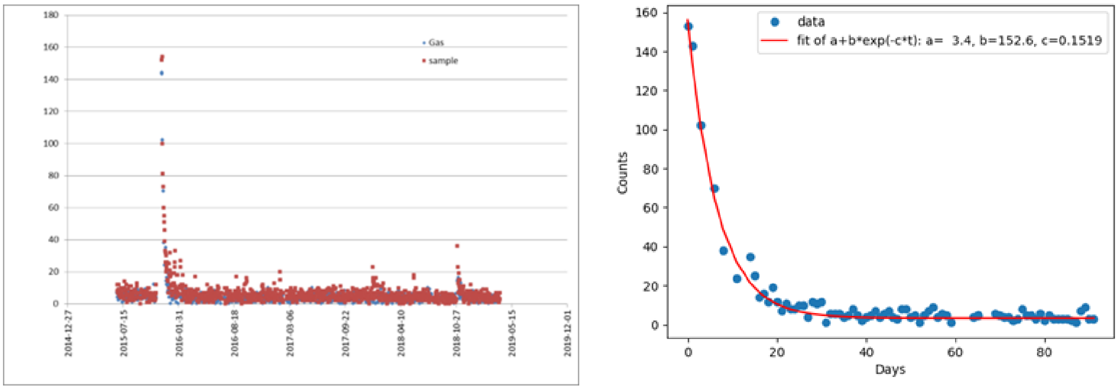

Figure 5 Memory effect in the gas background spectra. It can be fitted as $D_i(G) = a + be^{-ct}$.

Abnormal Radioxenon Background at IMS Stations

Regarding the radioxenon background at IMS stations, the two hypotheses in Table 2 are formulated:

- H_0: the null hypothesis of normal radioxenon background.
- H_1: the alternate hypothesis of anomaly radioxenon detection.

Table 2 The null hypothesis and alternative one on the normal radioxenon background from releases of nuclear facilities against an abnormal radioxenon detection

	True normal radioxenon background	True abnormal radioxenon detection
Do not reject H_0	Correct radioxenon background	False negative
Reject H_0	False positive	Correct abnormal radioxenon detection

The abnormal concentration threshold is based on the statistical analysis of a set of previous samples in a specified period, such as the long-term period of 365 days. The abnormal threshold c_{abn} is defined by:

$$c_{abn} = c(50) + n(c(75) - c(25)), \qquad (1)$$

where $c(x)$ is the concentrations at given probabilities of 25, 50 and 75, respectively, and $n = 3$. Noble gas samples are categorized into three levels, A/B/C in the categorization mechanism at the IDC. The decision threshold, i.e., LC, is used to discriminate whether a radioxenon is detected with respect to the distribution of the detector background. A sample is identified as the Level A in which the concentration is below the LC, otherwise, the level B or C.

Once a radioxenon is detected, the abnormal threshold is used to screen whether the radioxenon detection is caused by the normal radioxenon background at the IMS station. A sample is identified as the Level B once the concentration is below the abnormal threshold, otherwise, as the Level C. Notice that the purpose of the abnormal threshold is not used directly to discriminate a signal of nuclear explosion from radioxenon background, i.e., a Level C does not mean a detection of nuclear explosion.

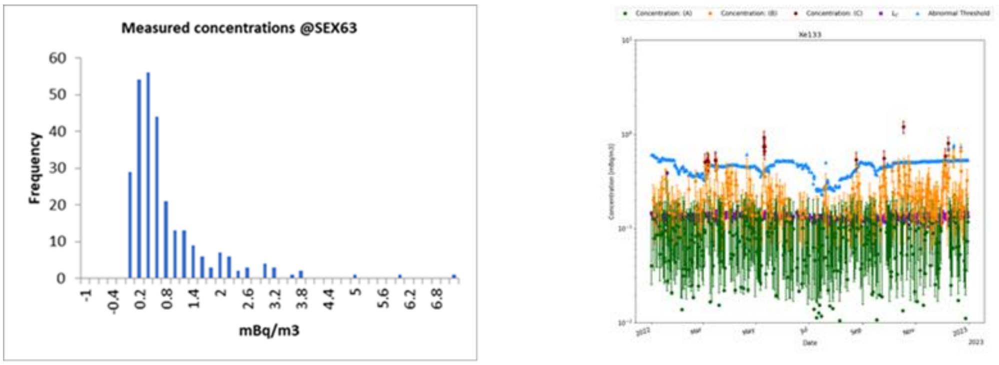

Figure 6 Distribution of daily activity concentrations in 2014 at SEX63

DISTRIBUTION OF ISOTOPIC RATIOS

Discrimination of a Nuclear Explosion Source

Discrimination of a nuclear explosion source against releases of nuclear facilities is based on isotopic ratio analysis basically, i.e., the relationship plots of four or three xenon isotopes. For the event discrimination using isotopic ratio analysis, both Level C and Level B samples in the IDC sample categorization scheme are used. The two hypotheses in Table 3 are formulated:

- H_0: the null hypothesis of nuclear facility release.
- H_1: the alternate hypothesis of nuclear explosion release.

Table 3 The null hypothesis and alternative one on the radioxenon background from releases of nuclear facilities against a nuclear explosion source

	True nuclear facility releases	True nuclear explosion release
Do not reject H_0	Correct nuclear facility releases	False negative
Reject H_0	False positive	Correct nuclear explosion release

MCM estimation by using concentrations

Instead of the analytical procedure, the MCM approach is based on the propagation of probability distributions (ISO/IEC, 2008). The analysis model is dependent on the measurement procedure and spectrum analysis of the sample.

In a simplified model of the MCM approach, Gaussian distributions of activity concentrations were used, and their correlation was ignored. The probability distribution of the isotopic ratios is derived first, see Figure 8 (a). Then the mean value and uncertainty are estimated accordingly. Furthermore, the limits of the coverage interval with the given probability, such as 95%, are estimated as well.

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%. Using the lower limit is to get a more conservative decision.

Four Radioxenon plot discriminating nuclear explosion

The discrimination between the highly variable radioxenon background caused by nuclear facilities and CTBT-relevant events is a challenging but crucial task. The characterization of a fission event is 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 are performed based on isotopic ratio analysis, e.g., four radioxenon plot of ^{135}Xe/^{133}Xe versus ^{133m}Xe/^{131m}Xe, see Figure 7. The discrimination line is given by:

$$r_{135/133} = 10^{-6} r_{133m/131m}^{4.4388}. \qquad (2)$$

For the release from nuclear facilities, the ellipse of the coverage interval of the IMS observations will be located in the left side of the discrimination line. Otherwise, it locates in the right side for the release source from a nuclear explosion. Statistical hypothesis testing is used to determine if the measured ratios result from releases from nuclear facilities or from a nuclear explosion. The null hypothesis is that a xenon detection is caused by a release(s) from nuclear facilities. The probability distribution for the null hypothesis is modelled with a two-dimensional normal distribution. Equal-probability contours of the probability distribution are ellipses in the two-ratio relationship plot. The mean values and covariance matrix of the probability distribution are iteratively determined so that 95% of sample measurements from nuclear facilities are contained by the ellipse contour at 2 standard deviations from the mean. Example measurements are provided through known measurements of releases from nuclear facilities, see Figure 7. Ratio measurements which fall outside of the 95% confidence ellipse are considered to be inconsistent with the null hypothesis and the source of a nuclear explosion cannot be ruled out. Therefore, the lower and right limits of the ellipse of the coverage interval needs to be applied, $(r_{135/133} - r^{l}, r_{133m/131m} + r^{u})$, $r^{l}$and r^{u} the lower and upper limits of the related isotopic ratios, respectively. Once the end point of lower and right limits crosses the discrimination line, a nuclear explosion cannot be ruled out.

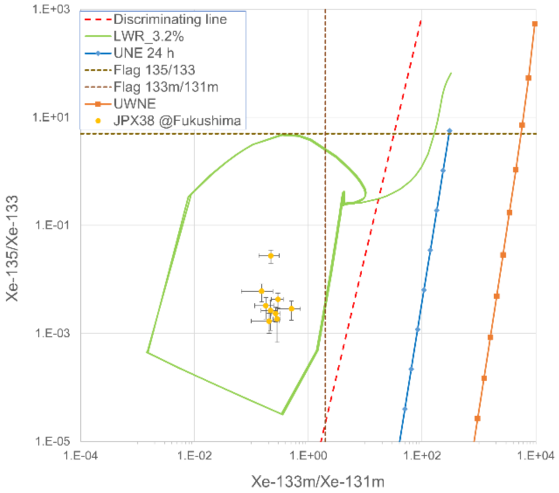

Fig. 7 Four radioxenon plot of ^{135}Xe/^{133}Xe versus ^{133m}Xe/^{131m}Xe. The evolution curves of the isotopic activity ratios for the two scenarios of ^{239f}Pu are included: UNE release at 24 h after detonation and underwater nuclear explosion (UWNE) immediate release (Burnett et al., 2019). The uppermost point indicates the release at 24 hours after the detonation time for UNE and zero hour for UWNE. The trajectory of LWR burn-up for 3.2% ^{235}U enrichment (evolution through three reactor circles) was replotted from Fig. 8 of Kalinowski et al. (2010). Four radioxenon isotopes were detected in a few samples at JPX38 after the Fukushima nuclear disaster in 2011 (Uncertainties of the isotopic activity ratios with two standard uncertainty). Replotted from Fig. 12 of Liu et al. (2023).

Event timing of nuclear explosion

Estimation of the explosion time of a nuclear event is based on an assumed scenario and a function of isotopic ratios over time, such as pairs of parent-daughter decay chains of ^{95}Zr/^{95}Nb, ^{140}Ba/^{140}La and ^{133m}Xe/^{133}Xe or pairs of independent fission products of ^{133}Xe/^{131m}Xe, ^{103}Ru/^{106}Ru (Yamba et al., 2016; Liu et al., 2023). The probability distribution of the elapsed times since the explosion time is derived based on the function of the isotopic ratio with time, a model based on Bateman equations related to the decay chain (Kalinowski and Liu, 2020). In a similar way, the mean value and associated uncertainty, including the limits of the coverage interval, are estimated by using the derived probability distribution.

Case study: Detections of the DPRK 2013 event

In this case study, the isotopic ratio and elapsed time for one of samples related to the DPRK2013 test were estimated using the Monte-Carlo method. For the sample at JPX38 at 19:00 on 8 April 2013 (the stop of collection), the concentrations are ^{133}Xe 3.05±0.14 (one standard deviation unless otherwise stated) and ^{131m}Xe 0.57±0.11 (mBq/m^3). For the isotopic ratio of ^{133}Xe to ^{131m}Xe, the nominal and biased values are 5.35±1.06 and 5.55±1.20 respectively. The bias is due to the relative uncertainty of 19% for ^{131m}Xe in the denominator.

The isotopic analysis in the concentration model was performed by the MCM, where Gaussian distributions were used. Both distributions of isotopic ratios and elapsed times were estimated accordingly, which differ from fitted Gaussian curves in Figure 9. This is due to exponentials and logarithms involved as well as the relative uncertainty of 19% for ^{131m}Xe. The ratio of activity concentrations of ^{133}Xe/^{131m}Xe is 5.59±1.29, different from the nominal value of 5.35±1.06 but within one standard uncertainty. Under a simplified full-ingrowth model, the mean values of elapsed times since the explosion time are 47.3±2.5 and 45.5±2.4 days for ^{235}U and ^{239}Pu respectively, which are close to the actual 54.5 days. The parameters and assumptions involved need to be further investigated, even directly based on the gross numbers of peak counts in the NCC method.

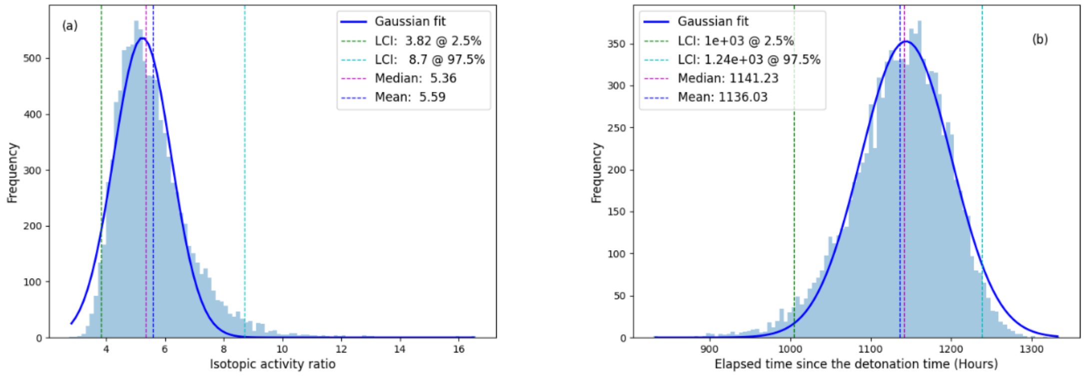

Fig. 8 Distributions of isotopic activity ratios and elapsed times derived by MCM under the assumption of a simplified full-ingrowth model for ^{235}U for the DPRK2013 sample at JPX38 at 19:00 on 8 April 2013 (collection stop). (a) Distribution of isotopic activity ratios ^{133}Xe/^{131m}Xe; (b) Distribution of elapsed times (hours). Replotted from Fig. 11 of Liu et al. (2023).

SUMMARY

The IDC data analysis and categorisation are applied on each measurement, for each sample at IMS stations. No repeated measurement is available, except for re-analysis at CTBT laboratories for certain selected samples. The numbers of peak counts in the single measurement are estimated by using the likelihood function, which has the same formula with the probability density function (PDF) of a priori distribution. The associated uncertainties are systematic uncertainties. The detector background is estimated based on a priori distribution and related measurements. Developments and enhancements of analysis algorithms should be consistent with estimation of measurement uncertainty and characterization limits based on Bayesian statistics.

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 nominal value of the isotopic activity ratio is estimated directly by the division of two activity concentrations and associated uncertainties given in IDC analysis reports. For a non-linear model of division operation, the biased value of an isotopic activity ratio and associated uncertainty needs to be estimated by high-order Taylor terms, e.g., the second-order polynomial, and they are dependent mainly on uncertainties of denominators, especially with large uncertainties of concentrations for low level samples. It is better to use the Monte-Carlo method, estimating isotopic ratios and their uncertainties based on activities measured in the sample or associated peak counts directly. 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.

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(The views expressed in this paper are those of the authors and do not necessarily reflect the views of the CTBTO that the authors represent.)