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ESTIMATION OF ISOTOPIC RATIOS OF FISSION PRODUCTS BY USING THE MONTE-CARLO METHOD

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ABSTRACT

Activity ratios of fission products detected at radionuclide stations of the international monitoring system (IMS) are used to not only discriminate a nuclear test from civil nuclear releases, but also determine the explosion time under assumed scenarios. A function of the isotopic ratio with time from the explosion time up to the stop of collection can be derived, based on Bateman equations of given decay chains. Activities collected in a sample are determined by spectrum analysis. It is not a linear relationship between activities collected in the sample and activity concentrations in the air plume passing the IMS station. Non-linear relationships of isotopic ratios could also be caused by the division algorithm when the activity concentration in a denominator has a larger measurement uncertainty. Covariances between isotope concentrations might be another reason for non-linearity. Correlations of two concentrations could be caused by decay corrections of a parent-daughter chain, interference corrections between two isotopes and subtractions of the same detector background measurement. This presentation demonstrates Monte-Carlo procedures estimating the probability distributions of isotopic ratios, based on input distributions related to spectrum measurements, calibration data and decay chains. Then the isotopic ratio, its uncertainty and associated limits of the coverage interval are estimated by using numerical or analytical solution. The methods and results for three typical isotope pairs are given, e.g. ^{140}Ba and ^{140}La, ^{95}Zr and ^{95}Nb, and ^{133}Xe and ^{131m}Xe. Furthermore, the explosion time and its uncertainty can be estimated in the same way, if applicable.

INTRODUCTION

Activity ratios of paired isotopes detected in particulate and noble gas samples can be used for event screening of a nuclear explosion from releases of civil facilities (Kalinowski and Pistner, 2006; Kalinowski et al, 2010; Kalinowski, 2011). For the case that all four xenon isotopes are measured, the most discriminatory plot is four xenon relationships between ^{133m}Xe/^{131m}Xe and ^{135}Xe/^{133}Xe. A special advantage is that it is independent of the time elapsed between release and detection. This approach can only be applied to an earlier release. For all combinations of isotopes with ^{135}Xe in the numerator it takes less than 5 days before the non-fractionated release from nuclear explosions reaches the reactor equilibrium. However, the activity ratios of two radioxenon measured routinely at IMS stations, such as ^{133}Xe to ^{131m}Xe, are routinely found in a range which might come from nuclear reactors such as medical isotope production facilities or from a delayed release of a nuclear test (Kalinowski and Liu, 2020). The isotopic ratios of ^{133}Xe and ^{131m}Xe recorded in April 2013 are considered a strong evidence for the nuclear nature of the seismic event of 12 February 2013, the announced DPRK nuclear test. Both ^{133}Xe and ^{131m}Xe were detected in three samples at JPX38 and two samples at RUX58 more than 50 days after the DPRK2013. The isotopic ratios of ^{131m}Xe/^{133}Xe are consistent with simplified simulations of later releases of xenon (Ringbom et al, 2014) and also with complicated simulations of xenon accumulated in a void/tunnel due to convection-driven leakage from the explosive cavity/chimney (Carrigan et al, 2016, 2020).

On the other hand, the explosion time of the nuclear event can be estimated by using a function of the isotopic ratio with time from the explosion time up to the stop of collection based on assumed scenarios, e.g., pairs of ^{140}Ba and ^{140}La, ^{95}Zr and ^{95}Nb, and ^{133}Xe and ^{131m}Xe (Carrigan et al, 2016; Ringbom et al, 2014; Yamba et al, 2016). If the same decay chain is assumed in both before releasing and during atmospheric transport, the elapsed time since the explosion time can be estimated through numerically solving the Bateman equations. The uncertainty estimation can be performed using the Monte-Carlo method regarding non-linear functions of exponentials and logarithms (ISO/IEC, 2008; Kalinowski and Liu, 2020).

The decay features of the isotopic ratio with time might be different with respect to three quantities, e.g. activities at the release time, activity concentrations at the stop of collection and activities measured at the start of acquisition. For both discrimination of a nuclear test and estimation of the explosion time, the isotopic ratio at the stop of collection is related to activity concentrations in the air plume at an IMS station instead of activities collected in the sample (Kalinowski and Liu, 2020). One of the reasons is due to the basic assumption that activities released are linear to activity concentrations in the plume in simulations of atmospheric transport modelling (ATM). On the other hand, the ratio of activities collected in the sample is not only related to the decay constants but also the duration of sampling. Decay chains are interrupted due to activity accumulation during sampling.

The first step is to estimate the isotopic ratio of two concentrations at the stop of collection based on the measurement procedure and spectrum analysis of the sample and assumption of the concentration profile during sampling. The uncertainty estimation of the isotopic ratio could be performed using Fieller's confidence regions (Axelsson and Ringbom, 2014) or Bayesian statistics (Zahringer and Kirchner, 2008). Uncertainty estimation of the explosion time could be based on the propagation of uncertainties (Yamba et al, 2016). However, it might be very complicated, even impossible, when calculations are based on Bateman equations for a complicated decay chain.

For isotopic ratio analysis, it would be more practical to use the Monte-Carlo method (MCM) based on activities measured in the sample or associated peak counts in the spectrum analysis directly, especially for the non-linear relationship between the elapsed time and the isotopic ratio. This presentation demonstrates MCM approaches estimating probability distributions of isotopic ratios, based on input distributions related to measurements and decay chains. Then the isotopic ratio, associated uncertainty and limits of the coverage interval (LCI) can be estimated accordingly. Furthermore, the explosion time can also be estimated in the same way.

ISOTOPIC RATIO ANALYSIS

Generally, there are three isotopic ratios involved from release up to sample measurement. The first one is the ratio of activities of two isotopes at a release time. Secondly, the isotopic ratio of activity concentrations at the stop of collection could be defined in the radioactive plume at a sampling location. The third one is the ratio of activities collected in a sample, irrespective of at the stop of collection or at the start of acquisition. The activities collected in samples are measured and referred at the start of acquisition, then the ratios of the activities of two isotopes are estimated accordingly (Axelsson and Ringbom, 2014; Ringbom et al, 2014). This is different from the way the quantities are used in other investigations, such as the released activity or the activity concentration in the air plume (Kalinowski and Pistner, 2006; Kalinowski et al, 2010; Kalinowski, 2011). The procedure of the isotopic ratio analysis was described in (Kalinowski

and Liu, 2020) and below is a short summary of the related equations which are used to build MCM models.

Activities Released from a nuclear explosion

The activity evolution from the nuclear explosion up to releasing into the atmosphere is a complicated process. It is related to not only radioactive decay chains, but also the convection and diffusion of xenon gases from the explosion chimney to the ground surface. Xenon gases could be accumulating in a tunnel related to the nuclear test, resulting in a delayed release (Carrigan et al, 2016, 2020). Released activities can be estimated simply based on mass decay chains using Bateman equations (Bateman, 1910; Kalinowski, 2011). 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 activities of two isotopes at the elapsed time (t_1) since the explosion time respectively; $R(t_1)$ is the ratio of released activities.

Activity concentrations in the plume at an IMS station

After releasing, activities are diluted and transported in the atmosphere. Activity concentrations in the air plume can be expressed as the product of a spatial-temporal source and a source-receptor sensitivity (SRS) field at discrete locations and time intervals in ATM simulations by Eq. (2) (Wotawa et al, 2003; Becker et al, 2007).

$$C(t_2) = A(t_2)M(t_t) \quad (2)$$

Where, t_2 is the elapsed time since the explosion time at the stop of collection; $A(t_2)$ is the total activity released (Bq); $C(t_2)$ is the activity concentration at the stop of collection in the air plume passing through the measurement station during sampling (Bqm^{-3}); $M(t_t)$ is the SRS field (m^{-3}) associated with the sample; t_t is the transport duration of the plume from the release time to the stop of collection at the measurement station ($t_t = t_2 - t_1$).

Using Eq. (2), the isotopic ratio at the stop of collection can be estimated by Eq. (3).

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

The isotopic ratio in Eq. (3) is defined by total activities in the air plume originally. At the end of the day, it is estimated by using activity concentrations in the plume at the sampling location. That is due to the linear transport model in ATM simulations and the same dilution factor (i.e. the SRS field) for each isotope.

Activities Collected in a sample at the IMS station

Activity concentrations were assumed to be constant during sampling generally. Therefore, the activities collected in a sample at the stop of collection are estimated by Eq. (4), irrespective of an independent decay or parent-daughter decay chain.

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

Where, V_s is the air volume sampled, the subscript s indicates the activity collected in the sample; λ is the decay constant (s^{-1}); t_c is the duration of collection; $A_s(t_2)$ is the activity collected in the sample at the stop of collection. The activity concentration is referred at the stop of collection in Eq. (4), which is consistent with Eq. (3).

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

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

The isotopic ratio of activities collected in the sample is dependent on decay constants (λ_1, λ_2) as well as collection time (t_c). This differs features of the isotopic ratio of activity concentration in the air plume in Eq. (3), which is only dependent on decay parameters.

Activities Measured at the Start of Acquisition

For an independent decay, the activity $A_{1_s}(t_3)$ (Bq) in the sample at the start of acquisition (t_3) is determined by Eq. (6).

$$A_{1_s}(t_3) = \frac{x_1}{\varepsilon_1 B_{1R}}\frac{\lambda_1}{1-e^{-\lambda_1 t_a}} \quad (6)$$

Where, x_1 is the net number of counts for an isotope determined in spectrum analysis; ε_1 is the efficiency; B_{1R} is the branching ratio; t_a is the real acquisition time (s) of the spectrum.

For a decay chain of parent to daughter, the activity of the daughter isotope at the start of acquisition is estimated by Eq. (7).

$$A_{2_s}(t_3) = \frac{x_2}{\varepsilon_2 B_{2R}}\frac{\lambda_2}{1-e^{-\lambda_2 t_a}} - A_{1_s}(t_3)\frac{\lambda_2}{\lambda_2-\lambda_1}\left(\frac{\lambda_2}{\lambda_1}\frac{1-e^{-\lambda_1 t_a}}{1-e^{-\lambda_2 t_a}} - 1\right) \quad (7)$$

After the stop of collection, there is a processing period (t_p), the decay time of 24 h for particulate samples, or the processing time of a few hours for noble gas samples. Considering decay corrections, the activities at the stop of collection can be estimated by Eq. (8) and (9) for an independent decay and parent-daughter decay chain respectively.

$$A_{1_s}(t_2) = A_{1_s}(t_3)e^{\lambda_1 t_p} \quad (8)$$

$$A_{2_s}(t_2) = \left(A_{2_s}(t_3) - A_{1_s}(t_3)\frac{\lambda_2}{\lambda_2-\lambda_1}\left(1 - e^{-(\lambda_2-\lambda_1)t_p}\right)\right)e^{\lambda_2 t_p} \quad (9)$$

Sampling is completed at the stop of collection. How to measure activities collected in the sample is depended on decay chains and further processing of the sample. The collected sample might be measured by using different detection approaches, such as re-analysis in IMS radionuclide laboratories. It would be very practical to give the activities referred at the stop of collection, especially for estimation of activity concentrations.

Applying Eq. (8) and (9) into Eq. (5), the isotopic ratio of activity concentration at the stop of collection, based on the sample measurement, is estimated by Eq. (10).

$$R(t_2) = \frac{\lambda_2}{\lambda_1}\frac{1-e^{-\lambda_1 t_c}}{1-e^{-\lambda_2 t_c}}\frac{e^{-\lambda_1 t_p}}{e^{-\lambda_2 t_p}}\left(\frac{A_{2sT}(t_3)}{A_{1s}(t_3)} - \frac{\lambda_2}{\lambda_2-\lambda_1}\left(\frac{\lambda_2}{\lambda_1}\frac{1-e^{-\lambda_1 t_a}}{1-e^{-\lambda_2 t_a}} - e^{-(\lambda_2-\lambda_1)t_p}\right)\right) \quad (10)$$

Where $A_{2sT}(t_3) = \frac{x_2}{\varepsilon_2 B_{2R}}\frac{\lambda_2}{1-e^{-\lambda_2 t_a}}$ indicates the activity of the daughter isotope determined by the peak counts without the parent-daughter decay correction, which is similar to Eq. (6).

Applying Eq. (6) and (7) into Eq. (10), the isotopic ratio of activity concentrations at the stop of collection is estimated by Eq. (11), based on net numbers of peak counts.

$$R(t_2) = \frac{\lambda_2}{\lambda_1}\frac{1-e^{-\lambda_1 t_c}}{1-e^{-\lambda_2 t_c}}\frac{e^{-\lambda_1 t_p}}{e^{-\lambda_2 t_p}}\left(\frac{x_2}{x_1}\frac{\varepsilon_1 B_{1R}}{\varepsilon_2 B_{2R}}\frac{\lambda_2}{\lambda_1}\frac{1-e^{-\lambda_1 t_a}}{1-e^{-\lambda_2 t_a}} - \frac{\lambda_2}{\lambda_2-\lambda_1}\left(\frac{\lambda_2}{\lambda_1}\frac{1-e^{-\lambda_1 t_a}}{1-e^{-\lambda_2 t_a}} - e^{-(\lambda_2-\lambda_1)t_p}\right)\right) \quad (11)$$

Analysis Procedure

Activities before releasing, activity concentrations in the plume, and activities collected in the sample can be related together, based on assumptions regarding decay evolutions, ATM simulations and concentration profile during sampling. The activity concentrations in the plume serve as a key role between release and sample measurement. The activities collected in the sample are determined based on spectra analysis, and the activity concentrations are estimated under the assumption of the concentration profile during sampling. On the other hand, the activities released from a nuclear explosion can be estimated according to assumed scenarios of the nuclear explosion, and the activity concentrations are estimated based on ATM simulations.

As mentioned above, the isotopic ratio of activities collected in the sample differs from the ratio of activities in the plume as shown in Eq. (1), (3) and (5). The analysis procedure is to estimate isotopic ratios of activity concentrations at the stop of collection in the air plume, based on activities measured in the sample.

MONTE-CARLO CALCULATION OF ISOTOPIC RATIOS

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. Firstly, the values of input parameters, such as activity concentrations in the analysis report or original peak counts in the spectrum, are sampled based on associated probability distributions. Then the distributions of isotopic ratios and/or explosion times are derived. Finally, the mean values of isotopic ratios and/or explosion times, associated uncertainties and limits of the coverage interval are estimated accordingly.

Analytical estimation of isotopic ratios

The results of concentrations and associated uncertainties are given in IDC analysis reports. For linear model, the nominal value (r_0) and associated uncertainty of the isotopic ratio are estimated by Eq. (12) simply.

$$r_0 = \frac{c_2}{c_1}; u^2(r_0) = r_0^2\left(\frac{u^2(c_1)}{c_1^2} + \frac{u^2(c_2)}{c_2^2} - 2\frac{COV(c_1,c_2)}{c_1 c_2}\right) \quad (12)$$

Notice that r_0 in Eq. (12) indicates an estimate of the nominal value based on mean values of concentrations, c_1 and c_2 given in the analysis report, but $R(t_2)$ in Eq. (3) indicates a random variable, the ratio of two random variables, concentrations $C_1(t_2)$ and $C_2(t_2)$.

The division algorithm of two concentrations is a non-linear model. The ratio (r) and associated uncertainty can be estimated by high-order Taylor terms by Eq. (13).

$$r = r_0\left(1 + \frac{u^2(c_1)}{c_1^2} - \frac{COV(c_1,c_2)}{c_1 c_2}\right);$$

$$u^2(r) = r_0^2\left(\frac{u^2(c_2)}{c_2^2}\left(1 + 3\frac{u^2(c_1)}{c_1^2}\right) + \frac{u^2(c_1)}{c_1^2}\left(1 + 8\frac{u^2(c_1)}{c_1^2}\right) - 2\frac{COV(c_1,c_2)}{c_1 c_2}\right) \quad (13)$$

The biases are mainly dependent on the uncertainty of denominator, as shown in Eq. (13), especially with large uncertainties of concentrations for low level samples.

Simple MCM estimation by using concentrations

In a simplified model of the MCM approach, Gaussian distributions of concentrations were used, and their correlation was ignored. For estimation of the isotopic ratio, the probability distribution of the isotopic ratios is derived based on the model in Eq. (3). 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.

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 combining Eq. (1) and (3) and 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.

MCM estimation by using results of peak counts

As shown in Eq. (10) and (11), the isotopic ratio of activity concentrations at the stop of collection can be estimated based on the results of spectrum analysis, such as activities or net numbers of peak counts. The correlation between activities or net numbers of peak counts need to be considered, which are dependent on the analysis algorithms. For example, in the net count calculation (NCC) method for noble gas samples with beta-gamma coincidence measurements, the correlations are due to three corrections, interference corrections from higher to lower energy regions of interest, subtraction of the same detector background in memory corrections, and decay correction of Xe-133m to Xe-133 during acquisition.

Generally, gross numbers of peak counts in the spectrum follow Poisson distributions, especially for low level counts, and they are independent. It might be a very direct procedure based on the gross numbers of peak counts although it could be complicated, such as the NCC method.

RESULTS AND DISCUSSIONS

Below are preliminary case studies of applying Gaussian distributions for concentrations, resulting in distributions of isotopic ratios and elapsed times since the explosion time.

Case study 1: 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 in Eq. (12) and (13), 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 in Eq. (3) was performed by the MCM, where Gaussian distributions were used in Figure 1 (left). Both distributions of isotopic ratios and elapsed times were estimated accordingly, which differ from fitted Gaussian curves in Figure 1 (middle, right). 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, such as the model based on activities in Eq. (10) or net numbers of peak counts in Eq. (11), or even directly based on the gross numbers of peak counts in the NCC method.

Case study 2: ^{95}Zr/^{95}Nb observations at SEP63 in June 2020

The pair of ^{95}Zr/^{95}Nb was not detected at SEP63 but detected in the lab re-analysis at ITL10. In this preliminary study, Gaussian distributions of concentrations were used in Figure 2 (left). In the lab report, the concentrations are 7.02±9.87(%) and 4.02±16.4 (%) uBq/m^3 for ^{95}Nb and ^{95}Zr respectively. For the isotopic ratio of ^{95}Nb/^{95}Zr in Eq. (12) and (13), the nominal and biased values are 1.75±0.33 and 1.79±0.36, slightly different. The distribution of elapsed times was derived simply by the parent-daughter decay of ^{95}Zr to ^{95m}Nb and ^{95}Nb. The nominal value of 175.3 days is different from the mean value of 186.8 days with the LCI (89.4, 429) days. It is caused by non-Gaussian distribution of the ratios as well as the truncated distribution due to the limit of the equilibrium ratio of 2.205 for ^{95}Zr to ^{95}Nb decay.

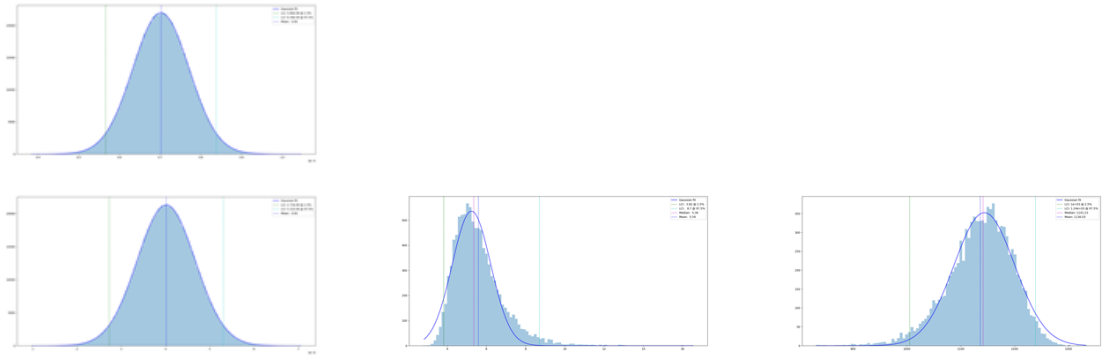

Figure 1. Distributions of inputs and calculated results under the assumption of a simplified full-ingrowth mode for ^{235}U for the DPRK2013 sample at JPX38 at 19:00 on 8 April 2013 (the stop of collection). (Left) Gaussian distribution of inputs concentrations of ^{133}Xe: 3.05±0.14 and ^{131m}Xe: 0.57±0.11 (mBq/m3); (Middle) Distribution of isotopic ratios ^{133}Xe/^{131m}Xe; (Right) distribution of lapsed times. The mean value of elapsed times is 47.3 days with the LCI (41.7, 51.7) compared with the nominal value of 47.6 days.

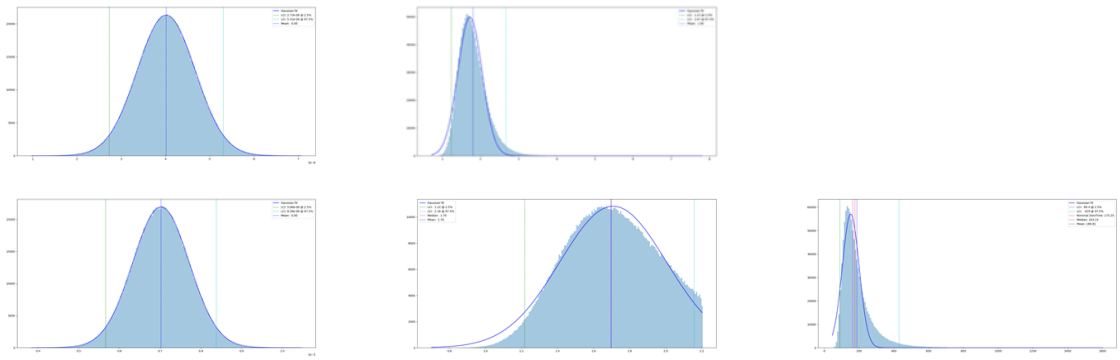

Figure 2. Distributions of inputs and calculated results for ^{95}Zr/Nb-95 observations at SEP63 in June 2020. (Left) Gaussian distribution of inputs concentrations of ^{95}Nb: 7.02±9.87(%) and ^{95}Zr: 4.02±16.4(%) uBq/m^3; (Middle) Distribution of isotopic ratios ^{95}Nb/^{95}Zr: (middle upper) the direct distribution, (middle lower) truncated distribution by the equilibrium ratio of 2.205; (Right) distribution of elapsed times. The mean value of elapsed times is 186.8 days with the LCI: (89.4, 429) days compared with the nominal value of 175.3 days.

SUMMARY

The features of isotopic ratios of activities measured at the start of acquisition are different from the ratios of activities at the release time and activity concentrations in the air plume at the stop of collection. Therefore, activity concentrations in the air plume are used in isotopic ratio

analysis, such as discrimination of nuclear event and estimation of explosion time. The division algorithm of two concentrations, random variables, is a non-linear model and the ratio and associated uncertainty can be estimated by high-order Taylor terms generally. The isotopic ratio and associated uncertainty are dependent on not only concentrations but also their uncertainties and covariances. For isotopic ratio analysis, it would be more practical to use the Monte-Carlo method based on activities measured in the sample or associated peak counts in the spectrum analysis directly, especially for uncertainty estimation related to the elapsed time since the explosion time.

In the preliminary studies, the distributions of concentrations were supposed as Gaussian and the correlations were ignored. In further studies, probability distributions of isotopic ratios and explosion times can be obtained directly based on distributions of gross or net numbers of peak counts measured, calibration data and related parameters, resulting in realistic estimates.

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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.)