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INVESTIGATING ISOTOPIC RATIO DISTRIBUTIONS AT IMS RADIONUCLIDE STATIONS USING EMISSIONS FROM NUCLEAR FACILITIES WITH DECAY CORRECTION BASED ON THE ATMOSPHERIC TRANSPORT TIME DISTRIBUTIONS FOR ONE YEAR

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ABSTRACT

The isotopic ratios of radioxenon can be useful for the discrimination between CTBT-relevant radioxenon detections related to nuclear testing and emissions from nuclear facilities. The typical isotopic ratios of radioxenon released from nuclear facilities are well known. However, due to the short half-life of some of the relevant radioxenons, the isotopic ratios have changed when reaching the International Monitoring System (IMS) radionuclide stations. For a better understanding of the expected isotopic ratios at the IMS stations, the isotopic ratio distributions of emissions from the nuclear facilities are evaluated first. Secondly, the source receptor sensitivity (SRS) fields calculated operationally with atmospheric transport modelling (ATM) are utilized to determine for these radionuclides the distributions of the travel times for one year between these facilities and the IMS stations. Then, the isotopic ratio distributions that can be expected for measurements at IMS stations can be calculated by folding these two kinds of distributions (emission and atmospheric transport time) while applying the radioactive decay equations. Finally, we compare these calculated isotopic ratio distributions of measurements at IMS stations with the real isotopic ratio distributions of measurement at IMS stations. This investigation can help to develop methods for screening by distinguishing between normal (based on known sources) and anomalous isotopic ratios. It may also be useful for discrimination between CTBT-relevant radioxenon detections and estimated observations based on emissions from known nuclear facilities as part of the effort of developing a Xenon Background Estimation Tool (XeBET).

INTRODUCTION

The verification regime of the Comprehensive Nuclear-Test-Ban Treaty (CTBT) is designed to detect any nuclear explosion conducted on Earth, whether that be underground, underwater, or in the atmosphere (CTBTO PrepCom, 2023). One of the regime's elements is the International Monitoring System (IMS), which monitors the planet for any sign of a nuclear explosion. The IMS is supported by the International Data Centre (IDC), which processes and analyses the data registered at the monitoring stations and produces data bulletins that are submitted to the Member States for their evaluation and judgement. The isotopic ratios of radioxenon can be useful for the discrimination between CTBT-relevant radioxenon detections related to nuclear testing and emissions from nuclear facilities (Kalinowski et al., 2010; Liu et al., 2022). The typical isotopic ratios of radioxenon released from nuclear facilities are well known (Kalinowski et al., 2023; Kuśmierczyk-Michulec et al., 2022). However, due to the short half-life of some of the relevant radioxenons, the isotopic ratios have changed when reaching the International Monitoring System (IMS) radionuclide stations. We investigated the isotopic ratio distributions at IMS radionuclide stations using the emission distributions at nuclear facilities and the source receptor sensitivity

(SRS) fields calculated with atmospheric transport modelling (ATM) (Kuśmierczyk-Michulec et al., 2022; Tipka et al., 2020) for *e.g.*, the development of the expert technical analysis (ETA) (Liu et al., 2022). The methods using this investigation can be applied to predict radioxenon activity concentration distribution at IMS radionuclide stations or emission distribution at nuclear facility for *e.g.*, the development of a Xenon Background Estimation Tool (XeBET) (Schoemaker et al., 2023).

METHODOLOGY

The isotopic ratio of radioxenon at IMS radionuclide station (Liu et al., 2022; Kijima et al., 2022) is given by Eq. (1):

$$r_R = \exp[-(\lambda_1 - \lambda_2)t]\, r_S\,, \tag{1}$$

where r_R, isotopic ratio of radioxenon isotope 1 to isotope 2 at IMS radionuclide station, λ_1, the decay constant of isotope 1 [s^{-1}], λ_2, the decay constant of isotope 2 [s^{-1}], t, atmospheric transport time between nuclear facility and IMS station [s], r_S, isotopic ratio of radioxenon isotope 1 to isotope 2 at nuclear facility. Then, the probability density function (PDF) of the isotopic ratio of isotope 1 to isotope 2 at an IMS station (Glen et al., 2004; ISO 11929-2:2019(E)) is given by Eq. (2) as follows:

$$P_{RR}(r_P|r_R) = kP_{RR}(r_P)\int_{-\infty}^{+\infty} P_T\left(\frac{r_R}{r_S}\right) P_{RS}(r_S)\frac{1}{|r_S|}\,dr_S\,, \tag{2}$$

where

$$P_{RR}(r_P) = \begin{cases} 1 & if\ \ r_P \geqq r_l \\ 0 & \text{otherwise} \end{cases},$$

with r_P, prediction isotopic ratio at IMS station, r_l, the minimum isotopic ratio measured at IMS station, k, the constant of proportionality, $P_T(\delta)$, the atmospheric transport time PDF of the ratio of the two ratios between nuclear facility and the IMS station, and $P_{RS}(\varepsilon)$, the isotopic ratio PDF of the ratio at the source.

The activity concentration of radioxenon at IMS radionuclide station (Wotawa et al., 2003) is given by Eq. (3):

$$c_R = m \times s_S\,, \tag{3}$$

where $c_R \equiv c_{Receptor}$, activity concentration of radioxenon at IMS station [Bq/m^3], m, source receptor sensitivity (SRS) between nuclear facility and IMS station [m^{-3}], $s_S \equiv s_{Source}$, is the source attribution, or in this case the emission amount of radioxenon at nuclear facility [Bq]. Then, the PDF of the activity concentration at an IMS station (Glen et al., 2004; ISO 11929-2:2019(E)) is given by Eq. (4):

$$P_R(c_P|c_R) = lP_R(c_P)\int_{-\infty}^{+\infty} P_m(m)P_S\left(\frac{c_R}{m}\right)\frac{1}{|m|}\,dm\,, \tag{4}$$

where

$$P_R(c_P) = \begin{cases} 1 & if\ c_P \geqq L_C \\ 0 & \text{otherwise} \end{cases},$$

with c_P, the predicted activity concentration of radioxenon at an IMS station [Bq/m^3], L_C, the critical limit [Bq/m^3], l, the constant of proportionality, $P_m(m)$, the SRS PDF of the random variable m between a nuclear facility and an IMS station, $P_S\left(\frac{c_R}{m}\right)$, the activity concentration PDF of the random variable $\frac{c_R}{m}$ at a nuclear facility. The residual (difference) between the measurement and the prediction of activity concentration at IMS station is given by:

$$c_D = c_M - c_P\ , \tag{5}$$

where c_D, the residual activity concentration [Bq/m^3], c_M, measurement activity concentration at IMS station [Bq/m^3]. Then, the residual PDF (Mallick et al., 2018) is given by Eq. (6):

$$P_D(c_D) = \int_{-\infty}^{+\infty} P_M(c_D + c_P) P_P(c_P)\, dc_P\ , \tag{6}$$

where $P_M(\kappa)$, measurement PDF of random variable κ, $P_P(\mu)$, prediction PDF of random variable μ.

The emission amount of radioxenon at nuclear facility is given by Eq. (7):

$$s_S = \frac{c_R}{m}\ . \tag{7}$$

Then, the PDF of emission at nuclear facility (Curtiss, 1941; ISO 11929-2:2019(E)) is given by Eq. (8):

$$P_S(s_P|s_S) = nP_S(s_P) \int_{-\infty}^{+\infty} |m| P_R(s_S m) P_m(m)\, dm\ , \tag{8}$$

where

$$P_S(s_P) = \begin{cases} 1 & if\ s_P \geqq 0 \\ 0 & \text{otherwise} \end{cases},$$

s_P, prediction emission amount of radioxenon at nuclear facility [Bq], n, constant of proportionality.

The Monte Carlo method (MCM) is used to solve these equations (Eq. (2), (4), (6) and (8)) numerically (JCGM 101:2008).

CASE STUDY

The emission data at Canadian Nuclear Laboratories (CNL) provided by CNL (for details see Maurer et al., 2022; Kalinowski et al., 2023) and the measurement data at an IMS radionuclide station in St. John's N.L., Canada (CAX17) in 2014 are used. The location of CNL and CAX17 are shown in Figure 1.

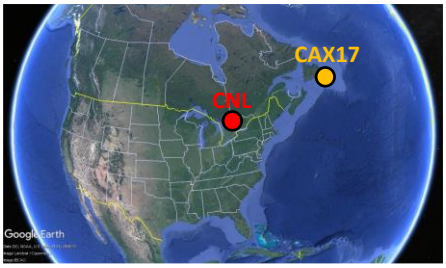

Figure 1. Location of Canadian Nuclear Laboratories (CNL) and IMS radionuclide station in St. John's N.L., Canada (CAX17).

The Monte Carlo method (MCM) is used to combine the emission distribution of Xe-135/Xe-133 at CNL in 2014 with the atmospheric transport time distribution between CNL and CAX17 in July – September 2014 using the atmospheric transport model to predict the isotopic ratio (Xe-135/Xe-133) distribution at CAX17 (See Figure 2). The transport time pertains to the first-time of arrival, or first non-zero value, in the SRS file. MCM is used in the approach as follows. [The values are sampled randomly from these two distributions. And then, they are combined (*e.g.*, multiplied or subtracted), taking into account the radioactive decay if needed.] The prediction isotopic ratio distribution was compared with the measurement isotopic ratio distribution at CAX17 (See Figure 3). The threshold using the results of prediction is 0.17 for 95% confidence level and 0.25 for 99% confidence level. It might be possible that the emission sources of samples of CAX17 with a ratio greater than 0.17 (or 0.25) are nuclear facilities other than CNL.

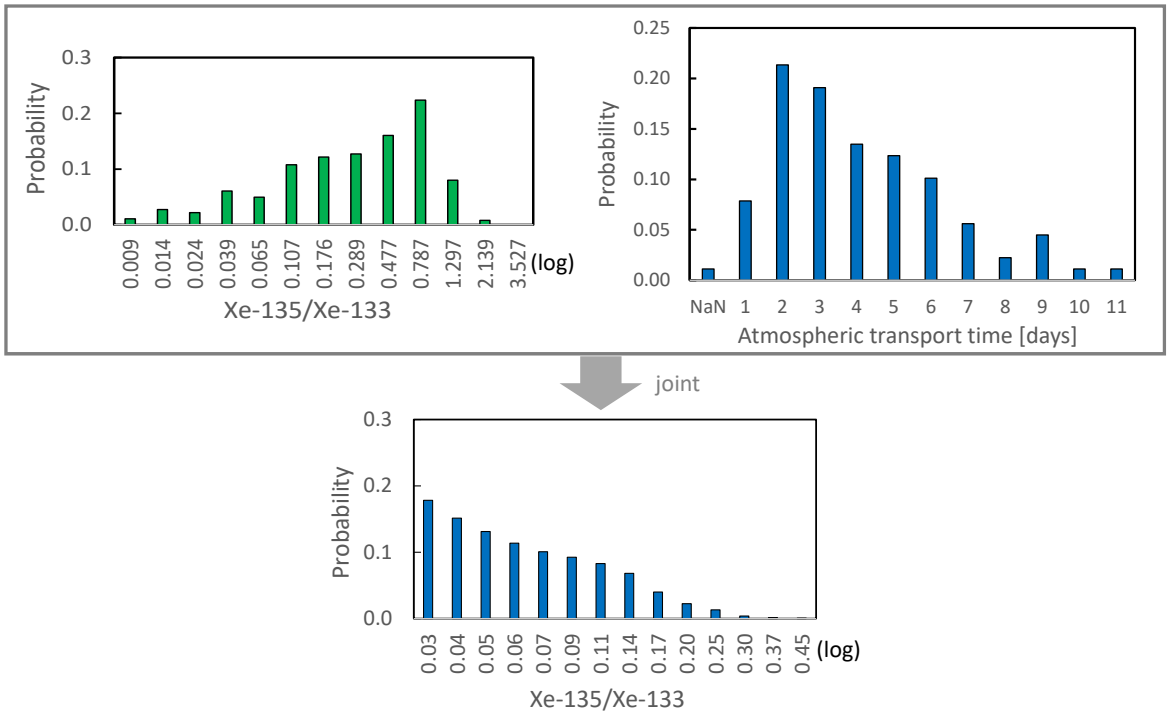

Figure 2. Prediction Xe-135/Xe-133 distribution at CAX17 (*lower panel*) using emission distribution of Xe-135/Xe-133 at CNL in 2014 (*upper left panel*) and atmospheric transport time distribution between CNL and CAX17 in July – September 2014 (*upper right panel*). The "*NaN*" in the upper right panel indicates that the radioxenon released from CNL does not pass through CAX17.

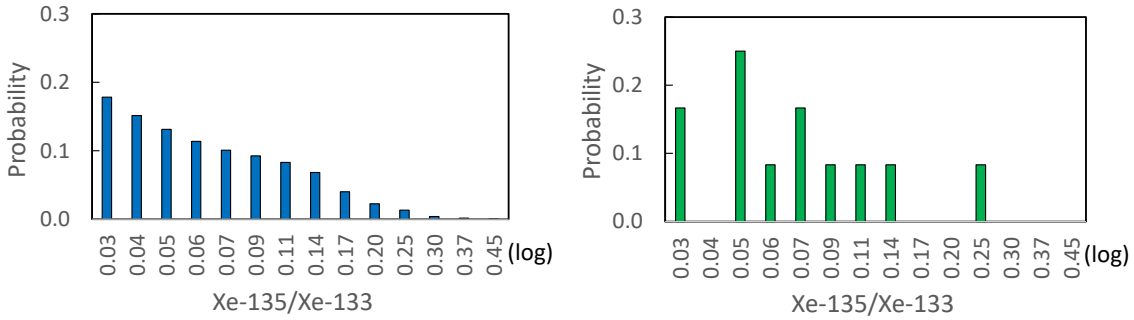

Figure 3. Comparison between the prediction (*left panel*) and the measurement (*right panel*) distributions of Xe-135/Xe-133 at CAX17.

The activity concentration distribution of Xe-133 at CNL in 2014 and the source receptor sensitivity (SRS) distribution between CNL and CAX17 in July – September 2014 using the atmospheric transport modelling are jointed to predict the activity concentration distribution of Xe-133 at CAX17 using the MCM (See Figure 4). And the predicted activity concentration distribution was compared with the measured activity concentration distribution at CAX17 (See Figure 5). The threshold using the results of prediction is 24.70 mBq/m^3 for 95% confidence level and 54.96 mBq/m^3 for 99% confidence level. It might be possible that the emission sources of samples of CAX17 with an activity concentration greater than 24.70 mBq/m^3 (or 54.96 mBq/m^3) are nuclear facilities other than CNL. Moreover, the residual distribution was also predicted (See Figure 6). The residual is a difference between the measured activity concentration and the predicted activity concentration. The expectation value and standard deviation of approximation curve (using the Gaussian fit) of the residual distribution is 0.04 and 1.39 mBq/m^3.

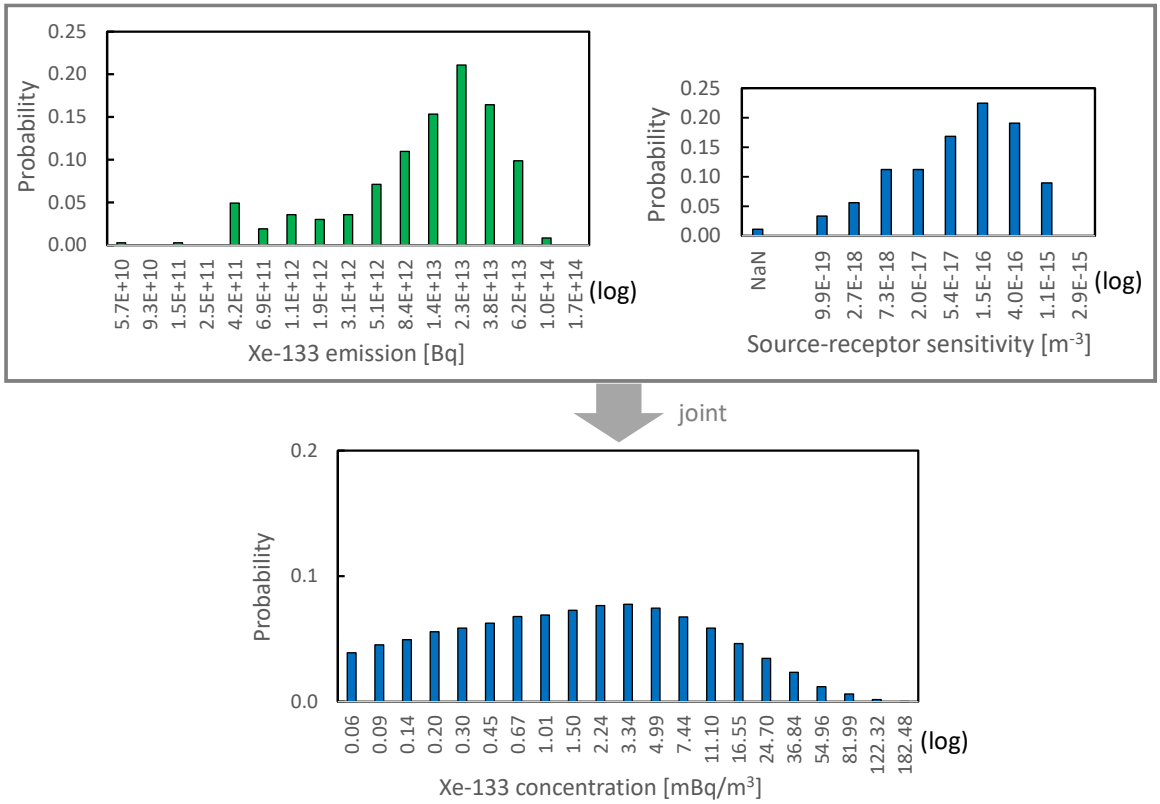

Figure 4. Prediction Xe-133 activity concentration distribution at CAX17 (*lower panel*) using emission distribution Xe-133 at CNL in 2014 (*upper left panel*) and source receptor

sensitivity (SRS) distribution between CNL and CAX17 in July – September 2014 (*upper right panel*). The "*NaN*" in the upper right panel indicates that the radioxenon released from CNL does not pass through CAX17.

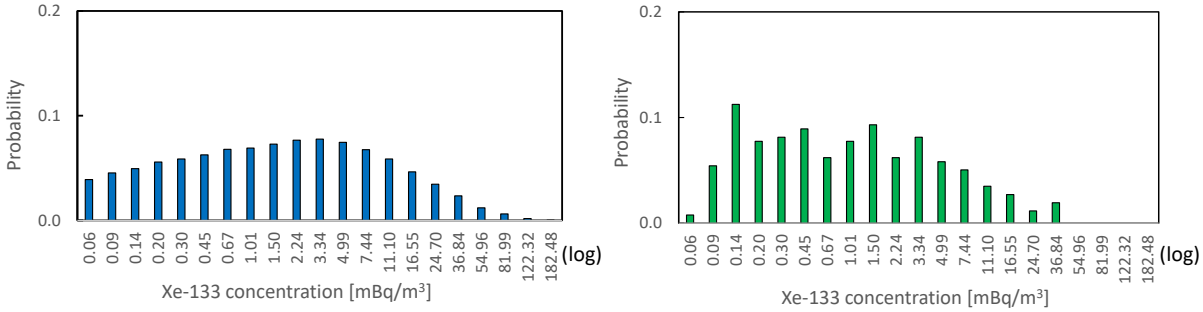

Figure 5. Comparison between the prediction (*left panel*) and the measurement (*right panel*) activity concentration distributions of Xe-133 at CAX17.

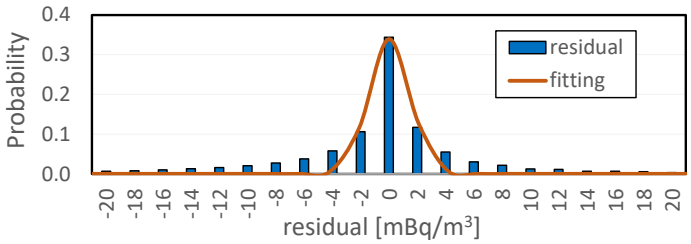

Figure 6. Residual distribution of Xe-133 at CAX17.

The activity concentration distribution of Xe-133 at CAX17 in 2014 and the source receptor sensitivity (SRS) distribution between CNL and CAX17 in July – September 2014 using the atmospheric transport modelling are jointed to predict the emission distribution of Xe-133 at CNL using the MCM (See Figure 7). And the prediction emission distribution was compared with the measurement emission distribution at CNL (See Figure 8). The expectation value (logarithmic mean) of the prediction (1.4E13 Bq) is good agreement with that of the measurement (1.2E13 Bq).

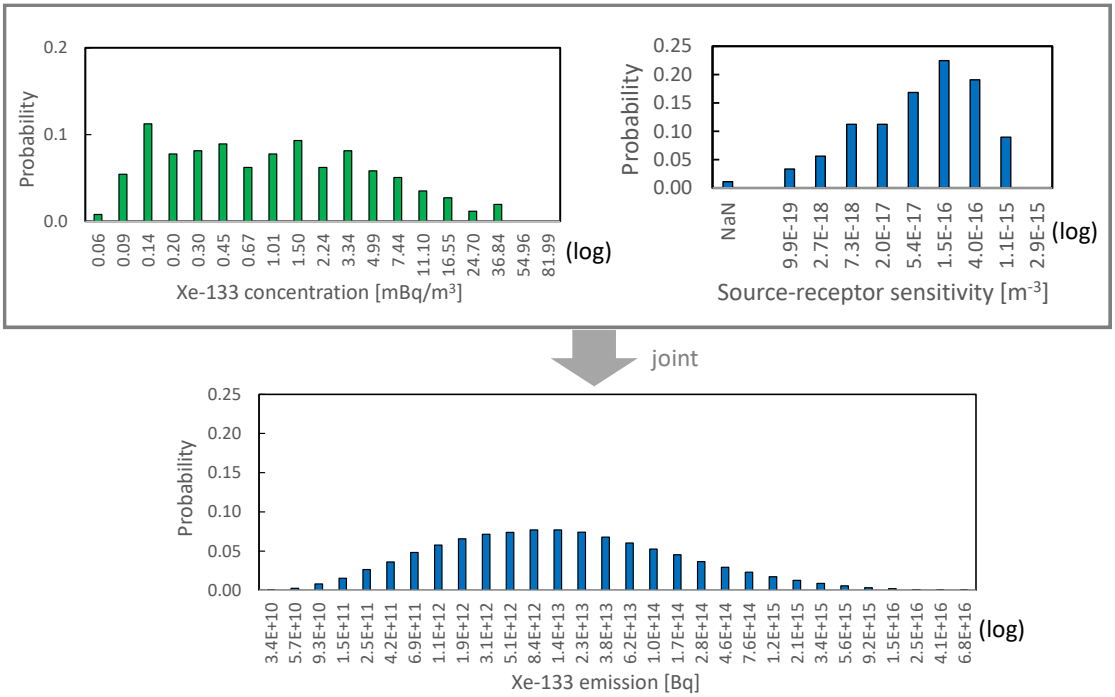

Figure 7. Prediction emission distribution of Xe-133 at CNL (*lower panel*) using Xe-133 activity concentration distribution at CAX17 in 2014 (*upper left panel*) and source receptor sensitivity (SRS) distribution between CAX17 and CNL in July – September 2014 (*upper right panel*). The "*NaN*" in the upper right panel indicates that the radioxenon released from CNL does not pass through CAX17.

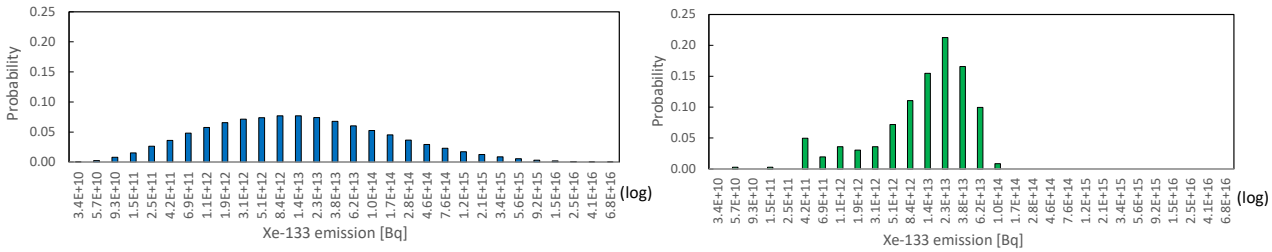

Figure 8. Comparison between the prediction (*left panel*) and the measurement (*right panel*) emission distributions of Xe-133 at CNL.

CONCLUSIONS

- The Monte Carlo method described here could be one of several prospective approaches to predict the activity concentrations and its isotopic ratios of CTBT-relevant radioxenons at IMS radionuclide stations.
- The knowledge about the radioxenon isotopic ratio distribution at the source is not sufficient for screening purposes.

DISCLAIMER

The views expressed herein are those of the authors and do not necessarily represent the views of the CTBTO Preparatory Commission.

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