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New evaluated radioxenon decay data and its implications in nuclear explosion monitoring

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ABSTRACT

This work presents the last updated evaluations of the nuclear and decay data of the four radioxenon isotopes of interest for the Comprehensive Nuclear-Test-Ban Treaty (CTBT): Xe-131 m, Xe-133, Xe-133 m and Xe-135. This includes the most recent measured values on the half-lives, gamma emission probabilities (Pγ) and internal conversion coefficients (ICC). The evaluation procedure has been made within the Decay Data Evaluation Project (DDEP) framework and using the latest available versions of nuclear and atomic data evaluation software tools and compilations. The consistency of the evaluations was confirmed by the very close result between the total available energy calculated with the present evaluated data and the tabulated Q-value. The article also analyzes the implications on the variation of the activity ratio calculations from radioxenon monitoring facilities depending on the nuclear database of reference.

1. Introduction

Due to their nuclear properties and their high fission yield, four radioxenon isotopes are of special interest for the CTBT verification regime: Xe-131 m, Xe-133, Xe-133 m and Xe-135 (De Geer, 2001). These radioactive fission products are used as gaseous indicators of nuclear explosions, with the advantage over the particle bound fission and activation products that they are released from underground nuclear tests with fairly high probability. Being noble gases, they do not interact with material in the soil and have very low solubility in water, so they escape through rock fissures to the atmosphere. The International Monitoring System (IMS) being set-up by the CTBTO Preparatory Commission has a worldwide network of radioxenon gas systems continuously monitoring radioxenon activity concentrations in air. The CTBTO also operates 16 radionuclide laboratories, some of which are (or will be) certified for noble gas capabilities. After Entry-into-Force of the CTBT, on-site inspections (OSI) can be performed to investigate suspect events.

The IMS has been designed with the goal of a 90% detection probability within approximately 14 days after an explosion with 1-kt TNT-equivalent has occurred in the atmosphere or from venting by an underground or underwater explosion assuming a detection limit of 10 $\mu Bq/m^3$ for Ba-140 in particulate filter samples, while the detection limit for Xe-133 has been set at 1 mBq/m^3 in noble gas samples (Schulze et al., 2000).

Within the context of the CTBT event screening process, it is highly important to distinguish the method of origin of radioxenon detections. Radioxenon isotopes are released from some civilian sources such as nuclear reactors and medical isotope production facilities. The most frequently recommended method to differentiate the origin currently employs plots of the isotope ratios $^{135}Xe/^{133}Xe$ versus $^{133m}Xe/^{133}Xe$ (Matthews et al., 2012). This approach was first proposed by Kalinowski and Pistner (2006). The three isotopes are rarely detected in one sample, therefore, simple activity ratios based on two isotopes are applied instead. With regard to the two axes of the isotope plots, high ratios (> 5 and > 2 respectively) are rarely detected from civilian sources and may provide evidence of a nuclear explosion but the assignation must be done carefully (Kalinowski and Tuma, 2009). All possible approaches for discriminating between reactor and explosion sources were systematically examined by Kalinowski et al. (2010). This study revealed that certain combinations of three or even only two isotopes can be used for source discrimination. These methods are most robust when the assumption can be made that only a single release caused the detection and no nuclear reactor is in the surrounding area that could create a multi-source background.

Another application of activity ratio determinations is for dating a nuclear event, that is, to calculate the zero time of a nuclear event that generated the independent yield of the fission products (Yamba et al.,

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2016). The age determination of the origin of the radioxenon isotopes from a fission source was vital for the detection of the radionuclide proof for the nuclear nature of the explosion that caused a seismic event at the nuclear test of the Democratic People's republic on 12 February 2013. In April 2013, an episode with multiple measurements at abnormally high concentrations of both Xe-133 and Xe-131 m was observed at two IMS stations, first at JPX38 (Takasaki, Japan) and several days later at RUX58 (Ussuriysk, Russian Federation). By determining the origin time, Ringbom et al. (2014) as well as Carrigan et al. (2016) were able to show that the isotopic activity ratio of Xe-133 and Xe-131 m is consistent with the assumption that these isotopes were generated by a fission reaction at the time of the announced nuclear test of the DPRK on 12 February 2013, and observed with slightly more than 50 days delay.

In addition, the correlation between observed detections of radioxenon and the seismic signal involved Atmospheric Transport Modeling (ATM) in both backward and forward modes. Radioxenon isotope half-lives are key parameters in the ATM analysis and are also directly used in the calculation of radioxenon isotopic ratio over time. In turn, this affects the accuracy of zero time estimates. The half-life uncertainties constitute only a small contribution of the zero-time uncertainty in the case of radioxenon isotopes and the fission yields are to be crucial when calculating production scenarios and to determine the time-zero given a particular production and release scenario. An uncertainty quantification of radioxenon isotopic activity ratios for discrimination of nuclear events with regard to nuclear explosion monitoring has been studied by Sloan et al. (2016). That paper found by sensitive analysis that the uncertainties of half-lives and branching factors are less sensitive to xenon concentrations compared to that of independent fission yields. However, the evaluations of the fission yields are not in the scope of the present paper.

Radioxenon gamma emission probabilities are used in the efficiency calibration of IMS noble gas detectors as well as in the analysis of radioxenon isotopes in daily samples from IMS noble gas systems. Gamma and X-ray energies of radioxenon isotopes are fundamental in the data analysis of HPGe based noble gas systems.

For secondary emissions, such as electron conversions and XK-ray, both energies and probabilities are also data sources for detector calibration procedures (Khrustalev et al., 2016) and to determine the radioxenon activity concentrations and related minimum detectable activity (MDA) in spectral data of beta-gamma coincidence based noble gas systems (Axelsson et al., 2012).

Therefore, accurate nuclear and decay data for radioxenon isotopes are needed for the CTBT verification regime.

2. Radioxenon isotope decay schemes and evaluations

The primary decay mode of the radioxenon isotopes is by emitting a beta (or conversion electron) in prompt time coincidence with an X-ray or a γ-ray emission. This feature permits very high sensitive measurements either by gamma-ray measurements or by coincidence techniques (β and/or electron capture and γ- or X-ray coincidence counting). Four different radioxenon measuring systems used in the IMS (ARIX, ARSA, SAUNA, and SPALAX) have been developed for CTBT monitoring (Dubasov et al., 2005; Bowyer et al., 1996; Ringbom et al., 2003; Fontaine et al., 2004). A new generation of noble gas system (Haas et al., 2017; Le Petit et al., 2015) is currently under development for the IMS and the CTBT radionuclide laboratories are setting up noble gas systems as well. Further noble gas systems are being developed which are customized to the needs of On-site Inspections for CTBT verification (Prelovskii et al., 2007; Zhou et al., 2016).

In Table 1, principal nuclear data for each radioxenon used in the radioxenon detectors for the CTBT are presented.

The evaluations of the radioxenon nuclear and decay data have been performed within the DDEP methodology (Bé et al., 2011) and with help of the associated evaluation computational tools (SAISINUC and EMISSION codes). For the ICC and mixing ratios calculations, the updated versions of BrIcc (Kibédi et al., 2008) and BriccMixing (NNDC) have been used. Also other ENSDF analysis and utility codes, such as GABS and RADLIST were used and the results were compared with the DDEP tools. The good agreement showed between the calculated total decay energy with EMISSION and RADLIST code (using as inputs the data of the present evaluations) and the Q-value evaluated in the AME2016 (Wang et al., 2017), validates the consistency of the decay data evaluated presented in this paper. Extended tables and comments on each radioxenon evaluation may be found in the DDEP Recommended Data Web Page (http://www.nucleide.org/DDEP_WG/DDEPdata.htm).

2.1. Xe-131 m

Xe-131 m has the longest half-life of the four radioxenon isotopes under study. It is produced with low independent yield from nuclear explosions but can be present in airborne samples as a result of ingrowth from the decay chain. Normal operational releases occur from medical facilities or nuclear reactors. This isomeric state (11.962 d) of Xe-131 decays by a strongly converted (about 98%) gamma transition of 163.930 keV. The vacancy of the orbital electrons is instantly reoccupied, causing the emission of characteristic xenon X-rays. The principal X-ray abundance (%) in coincidence with the conversion electron emissions is shown in Table 1.

The last Xe-131 m nuclear and decay data evaluation was undertaken in DDEP in 2014 (Bé et al., 2016). This review included the available measured data related to its nuclear structure and radioactive decay to that date. No relevant measurements to be included in a new evaluation have been published since 2014. Therefore, this radioxenon has not been revised in this work.

2.2. Xe-133

Xe-133 decays by β disintegration (branching ratio = 99%) to three excited levels in Cs-133 (stable). The strongest β-emission goes to the 81 keV level and it is followed by a gamma transition to the Xe-133 ground state. This 81-keV γ-ray is internally converted and only occurs in about 37% of the decay processes. Therefore in 37% of the processes a β-γ coincidence will occur while in most of the decay cases (49%) a β-particle, a conversion electron and a 30–34 keV X-ray (from ^{133}Cs readjustment) will be emitted all in coincidence.

New corrected half-life value has been reported (Unterweger, 2014) since last DDEP evaluation done in 2008 (Galan, 2009). The new evaluated half-life resulted in 5.2441 (37) d. This represents a variation of 0.1% with the previous adopted value of 5.2474 (5) d.

For the relative gamma-ray intensities the experimental data used in the previous Xe-133 DDEP evaluation as well as the new recommended data from the Ba-133 evaluation (Chechev & Kuzmenko, 2016) for the ratio γ(79.6 keV)/γ(161 keV) have been considered. Also new theoretical ICCs for the 79.6- and 81-keV obtained with BrIcc code have been computed. These resulted in a new normalization factor of 0.370 (4) for absolute gamma probability determinations.

The total available energy consumed in the decay process given by SAISINUC code was found to be 427.0 (38) keV, in good agreement with 427.4 (24) keV from AME2016.

2.3. Xe-133 m

This radioxenon is produced only by 2.88 (2) % of I-133 decay and its cumulative fission yield relative to Xe-133 is quite small. The ratio isomer/isotope given in a sample is a key indicator of a nuclear explosion.

The decay of Xe-133 m is also via an isomeric transition through a high converted (90%) gamma-ray of 233.15 keV. Therefore, a similar spectrum of X-ray and conversion electron of higher energy than in the

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