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Nuclear event zero time determination using analytical solutions of radioxenon activities under in-growth condition

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HIGHLIGHTS

- Systems of differential equations describing the decay of the CTBT-relevant radioxenons are solved analytically;
- Analytical formulas of the number of nuclides for Xe-135, Xe-133m, Xe-133 and Xe-131m under in-growth condition are given;
- For validation, the analytical solution results are used in comparison with numerical solutions given by other authors;
- The calculated ages using radioxenon activity data from real observations are in accordance with the reported ages.

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ABSTRACT

Motivated by simplifying the calculation process of radioxenon isotopic activity used by scientist community in nuclear event characterization, the analytical formulas of the numbers of nuclides and isotopic activities of CTBT relevant radioxenon Xe-135, Xe-133m, Xe-133 and Xe-131m proposed in this work can be useful and incorporated in the calculation algorithms for nuclear event studies. The calculated ages using analytical formulas and radioxenon activity data from real observations compare well with the reported ages and show good results of event timing precision.

1. Introduction

The Comprehensive Nuclear-Test-Ban Treaty (CTBT) is an international juridical tool banning any nuclear tests anywhere on the earth (underground, on-ground, in water, and atmosphere). Some radioisotopes of xenon namely Xe-135, Xe-133m, Xe-133 and Xe-131m are considered as most relevant indicators in nuclear explosion monitoring (De Geer et al., 2001). Plots of activity ratios for one pair of isotopes vs. another pair of isotopes in logarithmic scale can be used to characterize the source of the emission and most importantly to discriminate between nuclear reactors and nuclear explosions (Kalinowski and Pistner, 2006).

These radioxenons, by also considering their nuclei ratio or isotopic activity ratio can allow determining the time of a nuclear release (Bin, 1998).

Using radioxenon activity ratios in nuclear event timing needs measurement data (observations from International Monitoring System) and appropriate algorithms that can be obtained from radioactive decay. After a nuclear test (underground in particular), the relevant radioxenon should stay mixed with its precursors for some time, and that fact necessitates taking into account the contribution of the precursors in the amount of xenon nuclei detected. Considering beta minus and gamma transitions in the decay chains of concerned radioxenon, 135 chain starts by Sn-135, 133 by In-133 and 131 by Cd-131, as shown in Fig. 1. This presentation takes into account one isotope more than (Sloan et al., 2016) and (Sun et al., 2015) about 131 chain, and two more than De GEER's 131 chain (De Geer, 2012).

The formulas giving the isotopic activities in that case, i.e. nuclear explosion without immediate fractionation, are very challenging to be written analytically. So instead of using analytical formula, a numerical approach based on Bateman equation (Bateman, 1910) was commonly used in many studies for characterizing nuclear events.

This work proposes comprehensive formulas based on an analytical approach of the isotopic activity of the four radioxenons of interest, Xe-135, Xe-133m, Xe-133 and Xe-131m.

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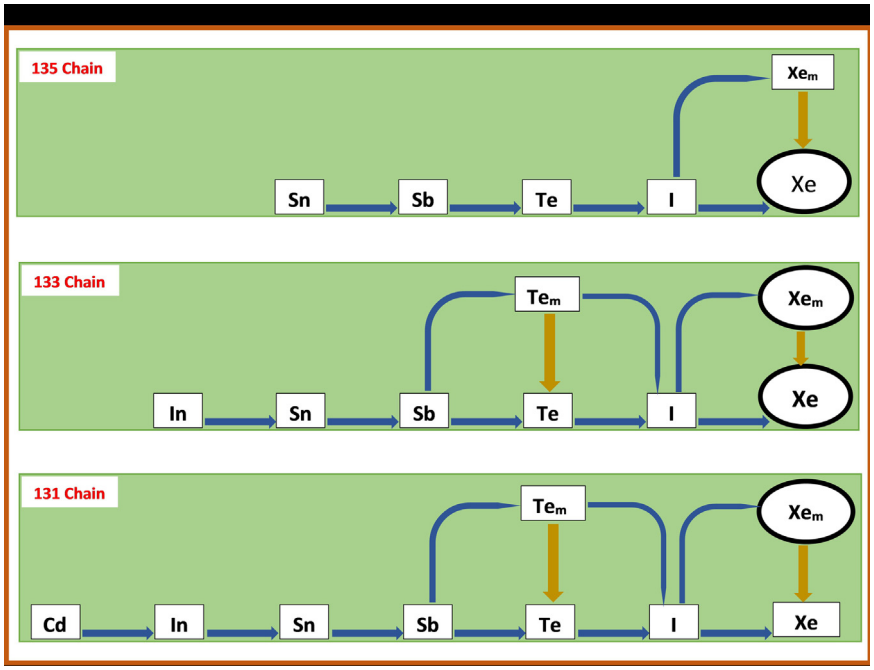

Fig. 1. Decay chains of the radioxenon of interest. Please note that the decay of Xe-131m, Xe-133 and Xe-135 take the isotope Cd-131, In-133 and Sn-135 respectively as starting points of the decay chains.

2. Isotopic activity determination for nuclear event timing

The results of measurements made in radio-xenon monitoring facilities are activities converted into and reported as activity concentrations. The activities are used for nuclear event timing (Axelsson and Ringbom, 2014; Yamba et al., 2015).

From the kind of nuclear release, three options with possible observations can be discussed in nuclear event timing:

- Effluents release from a Nuclear Power Plant (NPP)
- Nuclear explosion (nuclear test) along with a full fractionation and rapid venting
- Nuclear test in-growth condition and a delayed release

The rate of change in the nuclide number through a decay chain is described by a system of differential equations. For a straight chain with no branching that starts from a single parent nuclide with a number of $N_1(t = 0)$, while the daughters are initially zero, the general solutions for the number $N_n(t)$ of the n-th nuclide at time t have been given by Bateman (1910). Bateman's equation has to be extended for taking in consideration further parameters (Kalinowski, 2011).

To get the isotopic activity of each radioxenon of interest in- growth condition is to compute extended Bateman equation in order to take into account the initial numbers of nuclides (numbers of nuclides at zero time) for each concerned isotope. This extension allow also taking into account each decay branching (see Eq. (1)). (Kalinowski and Pistner, 2006)

$$N_{m,n}(t) = \sum_{k=1}^{n}\left[N^0_{m,k}\cdot\prod_{l=k}^{n-1}P_{m,l}\cdot\sum_{j=1}^{n}\left(\frac{T_{m,n}}{T_{m,j}}\cdot\prod_{\substack{i=1\\ i\neq j}}^{n}\frac{1}{1-\frac{T_{m,i}}{T_{m,j}}}\cdot e^{-\frac{ln2}{T_{m,j}}\cdot t}\right)\right] \quad (1)$$

$N_{m,n}(t)$ Number of nuclides;
$N^0_{m,k}$: Independent fission yield;
$P_{m,l}$: Decay branching ratio;
T: Half-life with $T = \ln(2)/\lambda$, λ is the decay constant;
m: Mass number (131, 133 or 135);
j, k, n, l: Index for isotopes and isomers of interest;
t: Elapsed time.

We can notice that this equation (extended Bateman's equation) has to be solved numerically for each isotope of interest.

In practice, the same differential equation (or systems of differential equations) is usable for calculating the formula giving isotopic activity of each radioxenon of interest for the first two cases mentioned above (NPP release and nuclear test with full fractionation), but by replacing into each formula the corresponding initial activity (activity at the time of release).

Differential equations describing the decay over time in the nuclear test with full fractionation and limited to the four CTBT relevant isotopes/isomers can be given as follows:

$$\frac{dN_1(t)}{dt} = -\lambda_1 \bullet N_1(t), \quad N_1(t=0) = N_1^0 \neq 0 \quad (2)$$

$$\frac{dN_2(t)}{dt} = -\lambda_2 \bullet N_2(t) + \lambda_1 \bullet N_1(t), \quad N_2(t=0) = N_2^0 \neq 0 \quad (3)$$

$N(t)$ Number of nuclides;
λ: Decay constant
t: Time

(2) refers to ^{131m}Xe; ^{133m}Xe and ^{135}Xe;
(3) refers to ^{133}Xe with index 1 for ^{133}Xe and index 2 for ^{133m}Xe.

Fig. 1 shows the decay chains giving the radioxenon Xe-135, Xe-133m, Xe-133 and Xe-131m in growth condition.

As we can see in Fig. 1, the determination of the isotopic activity of Xe-135, Xe-133m (and Xe-133) and Xe-131m takes necessarily into account the decay of their precursors. The three decay chains start with tin (Sn-135), indium (In-133) and cadmium (Cd-131) respectively.

The main difference between this new solution and Sun et al. (2015) is the contribution of some nuclides in our decay chains. First, our 131 chain starts with the nuclide Cd-131 instead In-131. The same difference can be noticed comparing our decay chain with Fig. 2 in De Geer (2012). Isotopes and isomers are not included in the differential equations, if their independent fission yield is smaller than 10^{-10}. For