Original Research Article

Using I135-Xe135 parent-daughter isotopic activity ratio for dating a recent nuclear event

ABSTRACT

This study reports on an accurate assessment of zero time of certain nuclear events using Xe-135/I-135 isotopic activity ratio. For a non-steady nuclear fission reaction, the dating is not possible. For the hypothesis of a nuclear explosion and for a release from a steady state nuclear fission reaction the zero-times will differ. This assessment is fast, because we propose some constants that can be used directly for the calculation of zero time and its upper and lower age limits. The assessment is accurate because of the calculation of zero time using a mathematical method, namely the weighted least-squares method, to evaluate an average value of the age of a nuclear event. This was done using two databases that exhibit differences between the values of some nuclear parameters, ENDF 2011 and TENDL 2011.

Keywords: Zero time, Isotopic Activity ratio, Xe-135, I-135

1. INTRODUCTION

The Comprehensive Nuclear-Test-Ban Treaty (CTBT) is an international legal instrument banning any nuclear tests anywhere on the earth (underground, on-ground, in water, and atmosphere). The International Monitoring System (IMS), built as part of verification regime of the Comprehensive Nuclear Test Ban Treaty Organization (CTBTO), comprises four monitoring technologies, namely infrasound mainly for atmospheric tests, seismic mainly for underground tests, hydroacoustic mainly for underwater tests, and radionuclides for all environments. The Comprehensive Nuclear Test Ban Treaty (CTBT) is not yet in force, but the verification regime has been almost established.

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 [1]. By considering the nuclei ratio or isotopic activity ratio of measured radionuclides, it is possible to evaluate their time of release (age of release) [2, 3]. It has also been shown that the age determination using isotopic activity ratios is very sensitive to some decay parameters like decay constants [4]. In that work, decay data used come from nuclear databases such as the French NUCLEIDE from Laboratoire National Henry Becquerel (LNHB - CEA) [5], the US Evaluated Nuclear Data structure File (ENSDF) from National Nuclear Data Center [6]. The nuclear database NUCLEIDE has updated some CTBT-relevant radioxenon decay data Within the framework of the DDEP (Decay Data Evaluation Project) project [7, 8].

Iodine I-135 and Xenon Xe-135 are two parent-daughter radionuclides that can be used for dating a fresh radioactive release. Some of these were detected in some samples taken by atmospheric radionuclide monitoring stations.

2. METHODOLOGY

Among the 321 CTBT monitoring stations, 80 have the ability to analyze radionuclides, of which 40 are equipped with noble gas detection system. The IMS includes 16 certified radionuclide laboratories. They evaluate filter samples further, if required [9]. The specifications for the radionuclide stations are listed in Table 1.

Table 1 Specifications for the aerosol and particulate monitoring stations (CTBTO/PCII/I/Add.2 P.48)

Specifications for the aerosol and particulate monitoring stations

Characteristics	Minimum requirements	
System	Manual or automated	1
Airflow	500 m^3 h^{-1}	2
Collection time[a]	24 h	3
Decay time[b]	≤24 h	4
Measurement time[c]	≥20 h	5
Time before reporting	≤72 h	6
Reporting frequency	Daily	7
Filter	Adequate composition for compaction, dissolution, and analysis	8
Particulate collection efficiency	For Filter: ≥80% at ϕ = 0.2 µm Global[d]: ≥60% at ϕ = 10 µm	9
Measurement mode	HPGe: High-resolution gamma spectrometry	10
HPGe relative effciency	≥40%	11
HPGe resolution	<2.5 keV at 1332 keV	12
Baseline sensitivity[e,f]	10–30 µBq m^{-3} for ^{140}Ba	13
Calibration range	88–1836 keV	14
Data format for gamma spectra and auxiliary data	RMS (radionuclide monitoring system) format[g]	15
State of health	Status of data transmitted to IDC	16
Communication	Two-way	17
Auxiliary data	Meteorological data Flow rate measured every 10 min	18
Data availability	≥95%	19
Downtime[h]	≤7 consecutive days ≤15 days annually	20

IDC: International Data Center

[a] Time specifications allow for an uncertainty of 10%, except for the reporting time parameter.
[b] This value can be reduced to a minimum of 6 h, if a suspicious event is detected by other stations or techniques.
[c] This value allows for measurement of authentication for manual systems.
[d] This global value includes 80% filter efficiency and collection efficiency of the incoming air circuitry.
[e] The upper limit is intended for high background areas.
[f] Certification procedures to be defined for baseline sensitivities (a posteriori Minimum Detectable Concentration MDC) as well as efficiency. Sample preparation losses should not affect baseline sensitivities.
[g] This format should make provision for auxiliary data, authentication data, and state-of-health data.
[h] Provision should be made for spare parts in particular areas where periodicity of transportation facilities is >7 days.

This study, based on the nuclear data analysis, takes into account the numerical differences between various constants used for dating nuclear events. This allows us to propose some parameters for a fast and accurate evaluation of zero time.

2.1 Nuclear event characterization

Nuclear explosions can be distinguished from civilian sources, for example, in terms of release of radionuclides or physical impact. Therefore, suspicious events first require examination of physical features of the samples. The high

temperature of an underground nuclear explosion forms vitrified rock at the bottom of the cavity, which could be found by drilling. A repository for waste from nuclear reactors would not exhibit such a feature. This serves as a unique criterion to determine the occurrence of a nuclear reaction [10].

The second step is the measurement of the emission of gamma and beta radiations from the samples. A highly radioactive sample would significantly suggest the presence of a nuclear fission source.

The third step is the estimation of abundances of the isotopes in the samples by measuring the intensities of the radiation. On the basis of the aforementioned principles, we could also determine the time of fission, if the analyzed isotopes are uniformly mixed. As described, some noble gases, volatile elements, and their daughter elements would have been depleted in the vitrified rock. We should avoid using such elements during the estimation of fission time [10].
It can also be noted that a fission reaction lasts from months to years. Isotopes with half-lives less than a few weeks would be present in reduced quantities in the waste. Therefore, if the amount of isotopes with short half-lives in the samples is less than that expected in nuclear explosions, the drilling sample could be originated from a deposit of reactor waste. By contrast, the fission reaction occurs almost at the same time in a nuclear explosion, and the time of the fission could be calculated by measuring the ratios of abundances of various pairs of isotopes. Each ratio could be used to estimate a fission time. If the average value of the fission time estimated from isotopes and the time of a suspicious event detected by the IMS coincide, the event could be confirmed as a nuclear explosion [10].
Regarding dating of nuclear events, the radioactive decay can be used in different ways. Most reliable are the isotopic ratios of parent–daughter radionuclides or those of the same element. In principle, any two radionuclides can be used.

2.2 Using Xe-135/I-135 isotopic activity ratio as a clock

Many radionuclides (with or without resulting from the same decay chain) can help date the age of a nuclear event. I-135 and Xe-135 are two among those with filiation (i.e., parent–daughter relationship). Iodine 135 (I-135) disintegrates by beta minus emissions to the Xenon 135 (Xe-135) excited level, and then, Xe-135 decays by beta minus emissions to the Cs-135 excited level. This decay chain can be used to evaluate the age of the nuclear event.

2.3 Sudden radioactive release

A nuclear explosion results in the sudden creation and possible subsequent release of radionuclides. The differential equations of decay are given as follows:

$$\frac{dN_I(t)}{dt} + \lambda_I N_I(t) = 0 \qquad [1]$$

$$\frac{dN_{Xe}(t)}{dt} + \lambda_{Xe} N_{Xe}(t) = \lambda_I N_I(t) \qquad [2]$$

where λ_I is the decay constant of I-135, λ_{Xe} is the decay constant of Xe-135, N_I represents the particle number of I-135, and N_{Xe} represents the particle number of Xe-135. In order to solve these equations, we assume that, during the release of radionuclides, the particle numbers $N_I(t = 0)$ and $N_{Xe}(t = 0)$ are nonzero. In case of a nuclear explosion this is the initial yield. Thus, we obtain

$$A_I(t) = A_I(0)e^{-\lambda_I t} \qquad [3]$$

$$A_{Xe}(t) = A_{Xe}(0)\frac{\lambda_{Xe}}{\lambda_{Xe} - \lambda_I}\left(e^{-\lambda_I t} - e^{-\lambda_{Xe} t}\right) + A_{Xe}(0)e^{-\lambda_{Xe} t} \qquad [4]$$

where A_I and A_{Xe} are the isotopic activities of I-135 and Xe-135, respectively. Then, the isotopic activity ratio $r(t) = \frac{A_{Xe}(t)}{A_I(t)}$ of these radionuclides is given by

$$r(t) = \frac{\lambda_{Xe}}{\lambda_{Xe} - \lambda_{Xe}} + \left(\frac{A_{Xe}(0)}{A_I(0)} - \frac{\lambda_{Xe}}{\lambda_{Xe} - \lambda_I}\right)e^{(\lambda_I - \lambda_{Xe})t} \qquad [5]$$

where $\frac{A_{Xe}(0)}{A_I(0)}$ is the activity ratio at the time of release, and the following approximation can be made: $\frac{A_{Xe}(0)}{A_I(0)} = \frac{\lambda_{Xe}\gamma_{Xe}}{\lambda_I\gamma_I}$, where γ_I and γ_{Xe} are respectively the cumulative fission yield of I-135 and the independent fission yield of Xe-135. The use of the cumulative fission yield for I-135 is justified by the fact that the longest half-life of any precursor of I-135 is 63.7 seconds. The error in zero-time estimation that is introduced by this approximation can be expected to be less than a few minutes. Zero time (i.e. the age) of a nuclear explosion can be evaluated by solving Eq. (5) with respect to t as follows:

$$t = \frac{1}{\lambda_I - \lambda_{Xe}} Ln\left(\frac{r - \frac{\lambda_{Xe}}{\lambda_{Xe} - \lambda_I}}{\frac{\lambda_{Xe}\gamma_{Xe}}{\lambda_I\gamma_I} - \frac{\lambda_{Xe}}{\lambda_{Xe} - \lambda_I}}\right) \quad [6]$$

Eq. (6) can be simple rewritten as

$$t = U \cdot ln\left(\frac{r - A}{F - A}\right) \quad [7]$$

where $\begin{cases} A = \frac{\lambda_{Xe}}{\lambda_{Xe}-\lambda_I} \; with\, \Delta A = \frac{\lambda_{Xe}}{\lambda_{Xe}-\lambda_I} \cdot \sqrt{\left(\frac{\Delta\lambda_{Xe}}{\lambda_{Xe}}\right)^2 + \left(\frac{\sqrt{(\Delta\lambda_{Xe})^2+(\Delta\lambda_I)^2}}{\lambda_{Xe}-\lambda_I}\right)^2} \\ r\ is\ the\ measured\ activity\ ratio \\ U = \frac{1}{\lambda_I - \lambda_{Xe}} \quad with\, \Delta U = \frac{\sqrt{(\Delta\lambda_I)^2+(\Delta\lambda_{Xe})^2}}{(\lambda_I-\lambda_Y)^2} \\ F = \frac{\lambda_{Xe}\gamma_{Xe}}{\lambda_I\gamma_I} \; with\, \Delta F = F\sqrt{(\frac{\Delta\lambda_{Xe}}{\lambda_{Xe}})^2 + (\frac{\Delta\lambda_I}{\lambda_I})^2 + (\frac{\Delta\gamma_I}{\gamma_I})^2 + (\frac{\Delta\gamma_{Xe}}{\gamma_{Xe}})^2} \end{cases}$ [8]

This equation can be simplified further to facilitate the calculation of uncertainty:

$$J = \frac{r - A}{F - A} \; with\, \Delta J = J \cdot \sqrt{\left(\frac{\sqrt{(\Delta r)^2 + (\Delta A)^2}}{r - A}\right)^2 + \left(\frac{\sqrt{(\Delta F)^2 + (\Delta A)^2}}{F - A}\right)^2} \quad [9]$$

The upper (t_{up}) and lower (t_{low}) age limits are assessed as follows:

$$\begin{cases} t = Uln(J) \\ t_{up} = (U + \Delta U)ln\,(J + \Delta J) \\ t_{low} = (U - \Delta U)ln\,(J - \Delta J) \end{cases} \quad [10]$$

The measured activity ratio (at the reference time) using gamma-ray spectrometry [2, 11] is written as

$$r = \frac{A_{Xe}}{A_I} = \frac{N_{Xe} ccf_{Xe} \lambda_{Xe} \varepsilon_I P_I (1 - e^{-\lambda_I t_c})}{N_I ccf_I \lambda_I \varepsilon_{Xe} P_{Xe} (1 - e^{-\lambda_{Xe} t_c})} - \frac{\lambda_{Xe}}{\lambda_{Xe} - \lambda_I}\left(\frac{\lambda_{Xe}}{\lambda_I}\frac{(1 - e^{-\lambda_I t_c})}{(1 - e^{-\lambda_{Xe} t_c})} - 1\right) \quad [11]$$

where N_I and N_{Xe} are the net peak areas of I-135 and Xe-135, respectively, ε_I and ε_{Xe} are respectively the efficiencies of the detector at a particular energy level, ccf_I and ccf_{Xe} are the coincidence correction factors depending on measurement conditions, coincidence correction factors, P_I and P_{Xe} are the emission probabilities at the specified energy level, and t_c is the measurement time.
In order to facilitate the uncertainty calculations, the following simplification is possible:

$$r = C\,L(B - A) + A, \quad [12]$$

where $\begin{cases} C = \frac{(1-e^{-\lambda_I t_c})}{(1-e^{-\lambda_{Xe} t_c})} \\ L = \frac{\lambda_{Xe}}{\lambda_I} \\ B = \frac{N_{Xe}}{N_I}\frac{ccf_{Xe}}{ccf_I}\frac{\varepsilon_I}{\varepsilon_{Xe}}\frac{P_I}{P_{Xe}} \\ A\ is\ the\ parameter\ defined\ previuosly \end{cases}$. [13]

The following are the steps involved in uncertainty calculations:

$$\Delta C = \sqrt{(\frac{t_c\, e^{-\lambda_I t_c}}{1 - e^{-\lambda_{Xe} t_c}})^2 (\Delta\lambda_I)^2 + (C\frac{t_c\, e^{-\lambda_{Xe} t_c}}{1 - e^{-\lambda_{Xe} t_c}})^2 (\Delta\lambda_{Xe})^2} \quad [14]$$

$$\Delta L = \frac{\lambda_{Xe}}{\lambda_I} \cdot \sqrt{\left(\frac{\Delta\lambda_{Xe}}{\lambda_{Xe}}\right)^2 + \left(\frac{\Delta\lambda_I}{\lambda_I}\right)^2} \quad [15]$$

$$\Delta B = B\sqrt{\left(\begin{array}{c}\left(\frac{\Delta N_I}{N_I}\right)^2+\left(\frac{\Delta \varepsilon_I}{\varepsilon_I}\right)^2+\left(\frac{\Delta P_I}{P_I}\right)^2+\left(\frac{\Delta ccf_I}{ccf_I}\right)^2+\\ \left(\frac{\Delta N_{Xe}}{N_{Xe}}\right)^2+\left(\frac{\Delta \varepsilon_{Xe}}{\varepsilon_{Xe}}\right)^2+\left(\frac{\Delta P_{Xe}}{P_{Xe}}\right)^2+\left(\frac{\Delta ccf_{Xe}}{ccf_{Xe}}\right)^2\end{array}\right)} \qquad [16]$$

Finally, we obtain the uncertainty of the measured activity ratio at the reference time (beginning of measurements) by

$$\Delta r = \sqrt{\left(C\cdot L\cdot (B-A)\cdot\sqrt{\left(\frac{\Delta C}{C}\right)^2+\left(\frac{\Delta L}{L}\right)^2+\left(\frac{\sqrt{\Delta B^2+\Delta A^2}}{B-A}\right)^2}\right)^2+(\Delta A)^2} \qquad [17]$$

Radioactive decay data used in this study come from many databases available online. Table 2 shows the cumulative and independent yields induced by thermal neutrons according to the databases ENDF.B.VII.I (USA), JENDL.VI.0 (Japan), JEFF.III.I (Europe), and TENDL (Europe). As we can see in, there are many numerical differences between these values that can affect the event zero time determination. By contrast, the analysis of the half-life data from databases ENDF and LARA give similar values and its uncertainties, as we can see in table 3 where half-life values of the radionuclides I-135 and Xe-135 are displayed.

Table 2: I-135 and Xe-135 fission-product yield (cumulative and independent) induced by thermal neutrons, for databases ENDF.B.VII.I (USA), JENDL.VI.0 (Japan), JEFF.III.I (Europe), and TENDL (Europe).

	$Cumulative\ Yield$ (%)		$Independent\ Yield$ (%)	
	I_{135}	Xe_{135}	I_{135}	Xe_{135}
$ENDF.B.VII.I$ 2011	$6.2819E-02$	$6.5385E-02$	$2.9274E-02$	$7.8513E-04$
	$\pm 8.7946E-04$	$\pm 4.5769E-04$	$\pm 8.1966E-04$	$\pm 4.7107E-05$
$JENDL.VI.0$ 2010	$6.2668E-02$	$6.5230E-02$	$2.9229E-02$	$7.5080E-04$
	$\pm 8.9225E-04$	$\pm 4.7337E-04$	$\pm 8.1842E-04$	$\pm 4.5048E-05$
$JEFF.III.I$ 2005	$6.3853E-02$	$6.6140E-02$	$2.5486E-02$	$6.9118E-04$
	$\pm 2.1926E-03$	$\pm 2.2490E-03$	$\pm 5.4041E-03$	$\pm 2.3815E-04$
$TENDL$ 2011	$7.3773E-02$	$7.6530E-02$	$2.9582E-02$	$7.9338E-04$
	$\pm 1.3279E-02$	$\pm 1.1479E-02$	$\pm 8.2829E-04$	$\pm 4.7604E-05$

Table 3: Half-life of the radionuclides I-135 and Xe-135. Values and their uncertainties for Xe-135 and I-135 are different.

		$half-life\ (hours)$
$ENDF.B.VII.I$	$I-135$	6.57 (2)
	$Xe-135$	9.14 (2)
$LARA-LNHB/CEA$	$I-135$	6.57 (2)
	$Xe-135$	9.14 (2)

2.4 Continuous fission reaction

In the case the source is not a nuclear explosion, we assume a release of radionuclides from a continuously running nuclear fission reaction in a nuclear reactor. The differential equations of decay are given as follows:

$$\frac{dN_I(t)}{dt} + \lambda_I N_I(t) = K_{Sr} \qquad [18]$$

$$\frac{dN_{Xe}(t)}{dt} + \lambda_{Xe} N_{Xe}(t) = \lambda_I N_I(t) + K_Y \qquad [19]$$

where K_I and K_{Xe} are the production rates of I-135 and Xe-135, respectively. By solving these equations, we obtain:

$$A_I = K_I\left(1 - e^{-\lambda_I t}\right) \qquad [20]$$

$$A_{Xe=} K_I \frac{\lambda_{Xe}}{\lambda_{Xe} - \lambda_I}\left(1 - e^{-\lambda_I t}\right) + \left(K_{Xe} - K_I \frac{\lambda_I}{\lambda_{Xe} - \lambda_I}\right)\left(1 - e^{-\lambda_{Xe} t}\right) \qquad [21]$$

where A_I and A_{Xe} are the isotopic activities of I-135 and Xe-135, respectively. The activity ratio $r = \frac{A_{Xe}}{A_I}$ is expressed as:

$$r(t) = \frac{\lambda_{Xe}}{\lambda_{Xe} - \lambda_I} + \left(\frac{K_{Xe}}{K_I} - \frac{\lambda_I}{\lambda_{Xe} - \lambda_I}\right)\frac{\left(1 - e^{-\lambda_{Xe} t}\right)}{\left(1 - e^{-\lambda_I t}\right)} \qquad [22]$$

The equilibrium level of this activity ratio is reached at:

$$r(t \to \infty) = 1 + \frac{K_{Xe}}{K_I} \qquad [23]$$

3. RESULTS AND DISCUSSION

Equation (22) is applicable only in a time interval $[t_c\,;\, t_0]$, where t_0 is the time of release and t_c is the time of onset of fission. For $t \in [t_c\,;\, t_0]$, $r(t)$ is denoted r_0.

After t_0, the equation of $r(t)$ in the case of a nuclear explosion (Eq. 5) must be used, by replacing $\frac{A_{Xe}(0)}{A_I(0)}$ by r_0. By assuming $K_{Xe} \ll K_I$ (or no production of radionuclide daughters), the isotopic activity ratio before t_0 reaching its equilibrium at $r(t \to \infty)$ becomes 1 (i.e., $r_0 = 1$). Therefore, in these conditions, in order to evaluate zero time for a non-explosion source, we must assume $F = 1$ and $\Delta F = 0$ by using the equation of isotopic activity ratio for nuclear explosion, with others values remaining unchanged. In addition, the numerical differences between decay data values and their uncertainties according to the mentioned databases, affect the parameters A, U and F as shown in table 4.

Table 4: Calculated parameters for a fast and accurate evaluation of zero time by using the activity ratio Xe-135/I-135.

	U	A	F
$ENDF.B.VII.I$ 2011	0.63361	−2.9775	0.0090118
	±0.058048	±0.2733	±0.0040627
$TENDL$ 2011	1.4046	−2.5564	0.0077304
	±0.017116	±0.031651	±0.001467

Zero time values for nuclear explosion and non-explosion source can be obtained by using the databases ENDF B.VII.I and LNHB/CEA. The weighted least-squares method [12] is used to obtain the average of different values of age calculated by using various nuclear databases, which is expressed as:

$$J_{average} = \frac{\sum_{i=1}^{2}(\frac{1}{\Delta J_i{}^2})J_i}{\sum_{i=1}^{2}\frac{1}{\Delta J_i{}^2}}, \qquad [24]$$

where J is a parameter that is defined earlier (Eq. 9).
It is not possible to use the measured activity ratio to determine the release time under the assumption of a non-explosion scenario at equilibrium, as its value is almost identical to the equilibrium level. This would be consistent with the assumption of no delay between the release from a source and begin of spectrum acquisition, whereas in reality 24 hours of sampling and 24 hours of decay (table 1) are between release and the radiation measurement. In general, for measured activity ratios of less than 1, the release scenario for a non-explosion source at equilibrium can be excluded. Dating a radioactive release using Xe135/I-135 is possible only for a recent release due to their short half-life value. In addition, Iodine I-135 having an half-life sorter than Xenon Xe-135, the change over time of the activity isotopic activity ratio Xe-135/I-135 diverge, as we can see in figure 1 and figure 2.

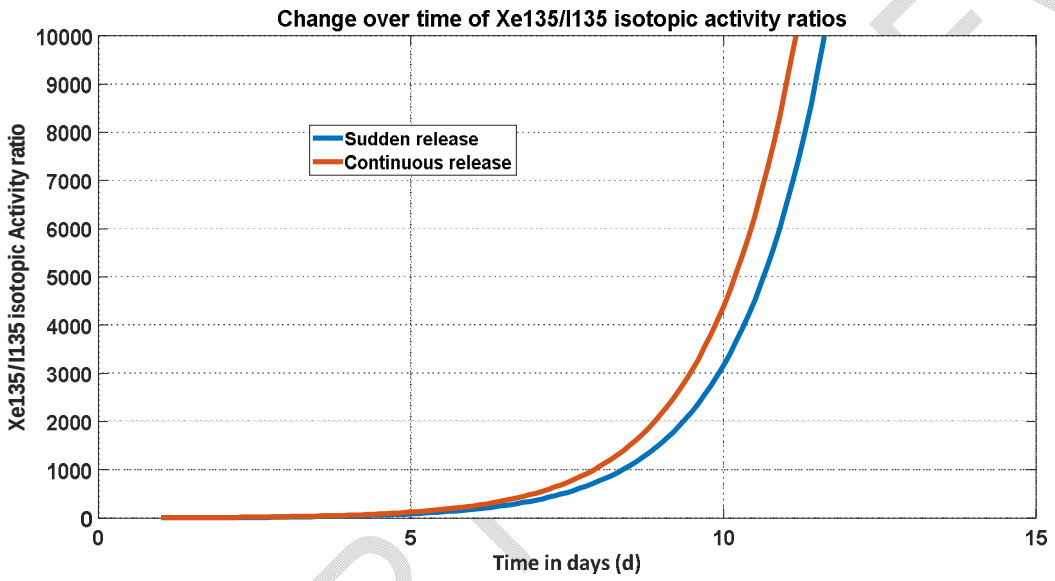

Figure 1: Change of I-135 and XE-135 activity with time.

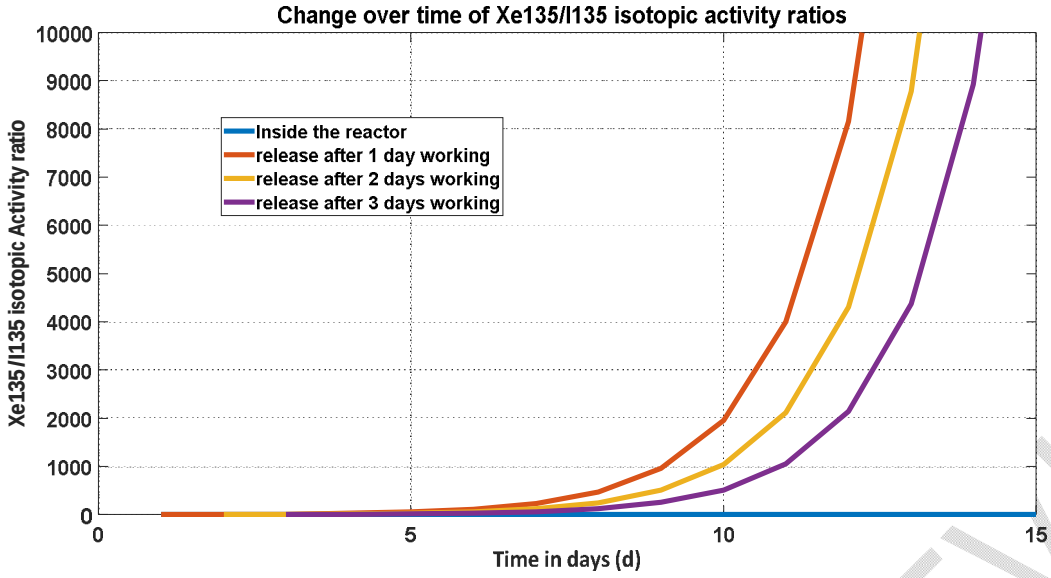

Figure 2: Evolution of the activity ratio Xe-135/I-135 (dashed blue line) for a non-explosion source (continuous production, i.e. nuclear reactor before reaching the steady state) and a release of I-135 and Xe-135 at certain times (colored solid lines).

4. CONCLUSION

In this study, we investigated the different possible approaches to evaluate the zero time of a nuclear event and the corresponding age limits, by using the isotopic activity ratio of the radionuclide pair I-135 and Xe-135. We also proposed a mathematical tool, applying the weighted least-squares method, to calculate accurately the zero time of a nuclear event. As the nuclear databases do not always show the same values of some constants used for the dating process, it is very important to take into account the values provided by nuclear databases such as ENDF V.II.I or LARA-LNHB/CEA.

It will be interesting to apply this calculation method to other isotopic ratios, in order to propose some nuclear constants that allow a simple and efficient calculation of the age of a nuclear event in case these pairs of isotopes are observed.

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