

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

ABSTRACT

This work focuses on the determination of time zero nuclear events considering the Xe-135/I-135 isotopic activity ratio. Considered as relevant isotopes for CTBT, these two radionuclides are usable for radioactive release timing. According to the considered databases, some parameters namely A, U and F are evaluated taking into account the numerical difference between the radioactive values from nuclear database. We found that these parameters give different values by considering the databases ENDF 2011 and TENDL 2011. These proposed parameters can be used for a quick and accurate evaluation of the nuclear event time and its upper and lower age limits.

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

1. INTRODUCTION

The Comprehensive Nuclear-Test-Ban Treaty (CTBT) is an international legal tool banning any nuclear tests anywhere on the earth (underground, on-ground, in water, and atmosphere). Opened for signature in September 1996, the CTBT was signed on July 2022 by more than 186 States. Each state party to the treaty works with the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO) under the treaty's verification regime, which is a set of scientific elements ensuring compliance with the terms of the treaty by the States parties. The verification regime includes the International Monitoring System (ISS), the International Data Center (IDC) and the On-Site Inspection (OSP). The CTBT has not yet entered into force, but its verification regime is already in place and operational. When radionuclides are detected abnormally, one wonders about their potential origin: debris from a nuclear explosion? Rejection of a nuclear power plant in operation? "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". [4]

"Graphical representations of the activity ratios for one pair of isotopes relative to another pair is usable to characterize the emission source and, importantly, to distinguish between nuclear reactors and nuclear explosions" [1]. "By considering the nuclear ratio or the isotopic activity ratio of the measured radionuclides, it is possible to determine their release time (release age)" [2, 3]. "It has also been shown that dating using isotope activity ratios is very sensitive to certain decay parameters such as decay constants" [4]. "The decay data used in this work come from nuclear databases such as the French NUCLEIDE of the Laboratoire National Henry Becquerel (LNHB – CEA) [5] and the US Evaluated Nuclear Data Structure File (ENSDF) of the National Nuclear Data Center" [6]. "The NUCLEIDE nuclear database has updated some radioxenon decay data relevant to CTBT as part of the Decay Data Evaluation Project (DDEP)". [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. How to use the Xe-135/I-135 ratio in the characterization of the radioactive release? How to improve the accuracy of calculated zero times? This work answers to these questions and proposes calculation constants allowing a rapid and precise evaluation of the radioactive release dates using Xe-135/I-135.

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: Some specifications for the aerosol and particulate monitoring stations (CTBTO/PCII//Add.2 P.48) [4]

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 $\phi = 0.2 \mu\text{m}$ Global ^d : ≥60% at $\phi = 10 \mu\text{m}$	9
Measurement mode	HPGe: High-resolution gamma spectrometry	10
HPGe relative efficiency	≥40%	11
HPGe resolution	<2.5 keV at 1332 keV	12
Baseline sensitivity ^{e,f}	10–30 $\mu\text{Bq m}^{-3}$ 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

The experimental measurement process takes place in three phases: a sampling phase which lasts 24 hours, a rest phase which lasts 24 hours and a measurement phase which also lasts 24 hours. The measurement of isotopic activities is made by the technique of gamma spectrometry. The ratio measured is then calculated using the activities of the isotopes Xe-135 and I-135. The expected activity is obtained by using the differential equations of radioactive decay. In this case, we will use an analytical method of resolution.

2.1 Radioactive release characterization

“Nuclear explosions can be distinguished from civil explosions in terms of release of radionuclides or actual physical impact. Thus, in the event of suspicious events, a preliminary examination of the physical characteristics of the samples is necessary. The high temperature of an underground nuclear explosion causes vitrified rock to form at the bottom of the cavity, which could be found by drilling into the ground. A repository for nuclear reactor waste would not have such a feature. This fact serves as the sole criterion for determining the occurrence of a nuclear reaction” [10].

Secondly, the emission of gamma and beta radiation from the samples is measured and qualified. A highly radioactive sample could clearly indicate the presence of a nuclear fission source.

"The third step will consist in estimating the abundance of the isotopes of interest in the samples by measuring the intensity of the radiation. On the basis of the principles cited above, one could also determine the moment of fission if the analyzed isotopes are uniformly mixed. As described, certain noble gases, volatile elements and their daughter elements would have been depleted in the vitrified rock. We should avoid using such elements in fission time estimation" [10].

"It can also be noted that a fission reaction lasts from months to years. Isotopes with half-lives of less than a few weeks would be present in the waste in reduced quantities. Therefore, if the amount of short half-life isotopes in the samples is less than would be expected from nuclear explosions, the drill sample could come from a reactor waste dump. In contrast, in a nuclear explosion, the fission reaction occurs almost simultaneously and the timing of fission could be calculated by measuring the abundance ratios of different isotope pairs. Any ratio could be used for estimating a cleavage time. If the average fission time determined from the isotopes and the timing of a suspicious event detected by the IMS match, the event could be confirmed as a nuclear explosion" [10].

Radioactive decay can be used in several ways to date nuclear events. Isotope ratios of parent-daughter radionuclides or of the same element are the most reliable. 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:

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

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

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

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

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

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

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

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:

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

Eq. (6) can be simple rewritten as

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

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

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

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

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

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

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

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

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, cc_{f_I} and $cc_{f_{Xe}}$ 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:

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

$$\text{where } \begin{cases} C = \frac{(1 - e^{-\lambda_I t_c})}{(1 - e^{-\lambda_{Xe} t_c})} \\ L = \frac{\lambda_{Xe}}{\lambda_I} \\ B = \frac{N_{Xe} cc_{f_{Xe}} \varepsilon_I P_I}{N_I cc_{f_I} \varepsilon_{Xe} P_{Xe}} \end{cases} \quad [13]$$

A is the parameter defined previously

The following are the steps involved in uncertainty calculations:

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

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

$$\Delta B = B \cdot \sqrt{\left(\left(\frac{\Delta N_I}{N_I}\right)^2 + \left(\frac{\Delta\varepsilon_I}{\varepsilon_I}\right)^2 + \left(\frac{\Delta P_I}{P_I}\right)^2 + \left(\frac{\Delta cc_{f_I}}{cc_{f_I}}\right)^2 + \left(\frac{\Delta N_{Xe}}{N_{Xe}}\right)^2 + \left(\frac{\Delta\varepsilon_{Xe}}{\varepsilon_{Xe}}\right)^2 + \left(\frac{\Delta P_{Xe}}{P_{Xe}}\right)^2 + \left(\frac{\Delta cc_{f_{Xe}}}{cc_{f_{Xe}}}\right)^2\right)} \quad [16]$$

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

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

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:

$$\frac{dN_I(t)}{dt} + \lambda_I N_I(t) = K_{Sr} \quad [\text{Error! Bookmark not de ined.}]$$

$$\frac{dN_{Xe}(t)}{dt} + \lambda_{Xe}N_{Xe}(t) = \lambda_I N_I(t) + K_Y \quad [\text{Error! Bookmark not de ined.}]$$

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

$$A_I = K_I(1 - e^{-\lambda_I t}) \quad [\text{Error! Bookmark not de ined.}]$$

$$A_{Xe} = K_I \frac{\lambda_{Xe}}{\lambda_{Xe} - \lambda_I} (1 - e^{-\lambda_I t}) + \left(K_{Xe} - K_I \frac{\lambda_I}{\lambda_{Xe} - \lambda_I} \right) (1 - e^{-\lambda_{Xe} t}) \quad [\text{Error! Bookmark not de ined.}]$$

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:

$$r(t) = \frac{\lambda_{Xe}}{\lambda_{Xe} - \lambda_I} + \left(\frac{K_{Xe}}{K_I} - \frac{\lambda_I}{\lambda_{Xe} - \lambda_I} \right) \frac{(1 - e^{-\lambda_{Xe} t})}{(1 - e^{-\lambda_I t})} \quad [\text{Error! Bookmark not de ined.}]$$

The equilibrium level of this activity ratio is reached at:

$$r(t \rightarrow \infty) = 1 + \frac{K_{Xe}}{K_I} \quad [\text{Error! Bookmark not de ined.}]$$

3. RESULTS AND DISCUSSION

Equation (22) is only applicable 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 $r(t \rightarrow \infty)$ 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-nuclear explosion source can be obtained by using the databases ENDF B.VII.I and LNHB/CEA. The weighted least-squares method [12] can be used to obtain the average of different values of age calculated by using various nuclear databases, which is expressed as:

$$J_{average} = \frac{\sum_{i=1}^2 \left(\frac{1}{\Delta J_i^2} \right) J_i}{\sum_{i=1}^2 \frac{1}{\Delta J_i^2}}, \quad [\text{Error! Bookmark not de ined.}]$$

where J is a parameter that is defined earlier (Eq. 9).

The measured radioactivity rate cannot be used to determine the release time assuming an equilibrium non-explosive scenario because its value is approximately equal to the equilibrium level. This is indeed consistent with the assumption that there is no delay between emission at the source and start of spectral acquisition when there is a 24-hour sampling and 24-hour decay (Table 1) between emission and radiation measurement. In general, non-explosive equilibrium source ignition scenarios can be excluded if the measured radioactivity ratio is less than 1.

Dating a radioactive release using Xe-135/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 shorter 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.

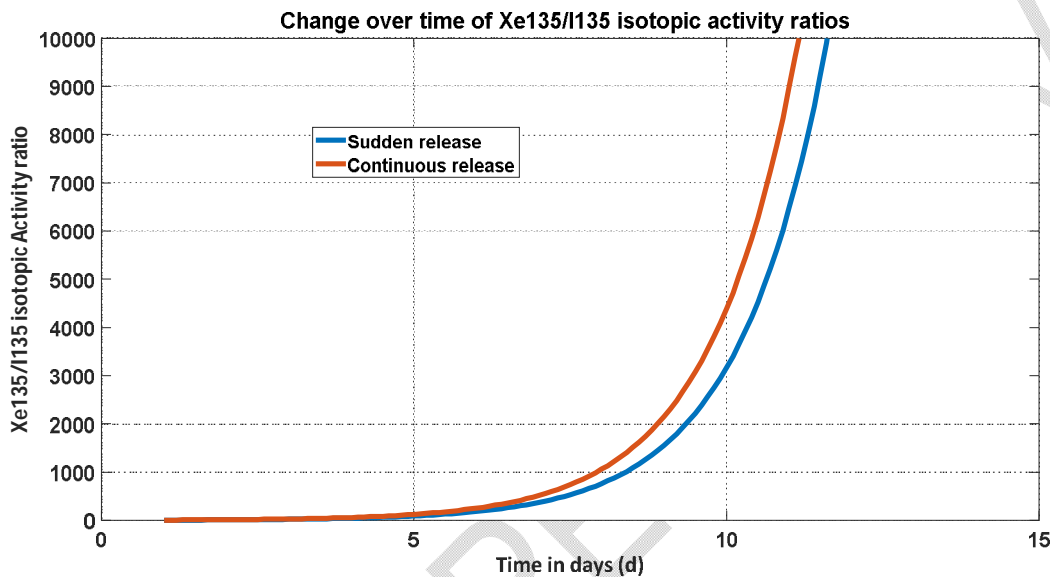


Figure 1: Change of I-135 and XE-135 activity with time.

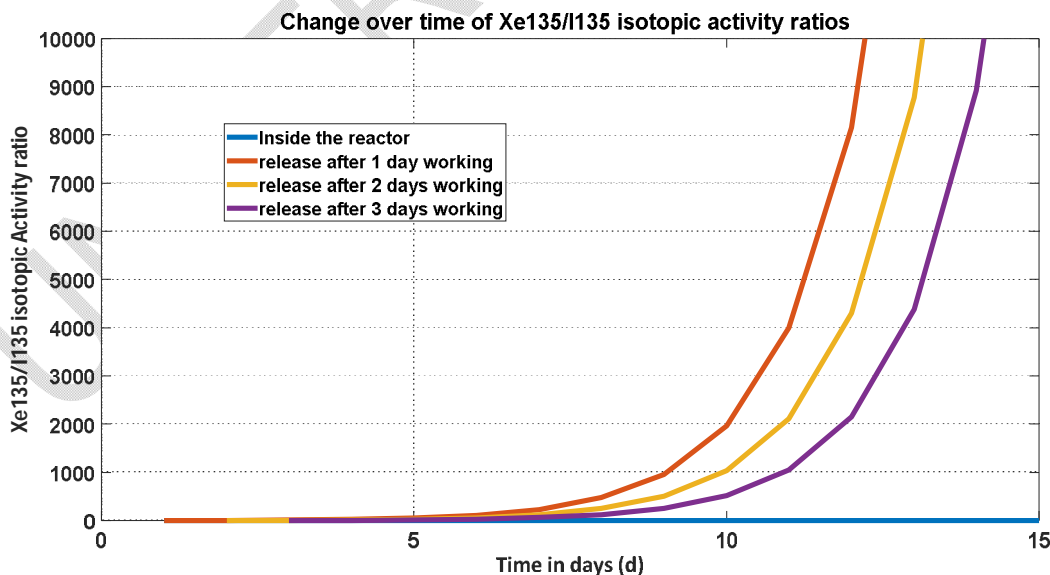


Figure 2: Change over time of the activity ratio Xe-135/I-135

4. CONCLUSION

In this report, we have mentioned the result of our investigation on the different possible approaches to evaluate the zero time of a nuclear event and its age limits. The radioisotopes pair used are I-135 and Xe-135 and by taking into account numerical differences between radioactive data from databases. The two databases considered are ENDF 2011 and TENDL 2011 that can be accessible online. We found that all the parameters U, A and F considered in this study are showing different values according to the database. So in order to calculate accurately the age of the radioactive release, the weighted least-squares method could be used.

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