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La-140/Ba-140 activity concentration ratio use for investigation on nuclear event occurrence

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ABSTRACT

During the month of May 2010, many radionuclides monitoring stations of the International Monitoring System (IMS) of CTBO detected some radionuclides. Some of them are La-140 and Ba-140. In the present paper we have done an investigation on the possible origin of these radionuclides as well as their release date, by using the isotopic activity ratio calculated from activity concentration recorded at the japan monitoring station JPP37 between 15 May and 23 May 2010 by using eight (08) samples. We concluded that these radionuclides are from the nuclear explosion that occurred in North Korea in May 2010 between 11 to 13th.

RESUME

En Mai 2010, plusieurs stations de surveillance des radionucléides du système de surveillance international (SSI) de l'Organisation du traité d'interdiction complète des essais nucléaires (OTICE) ont détecté des particules radioactives. Parmi ces détections se trouve une quantité non négligeable des radionucléides La-140 et Ba-140. Cet article propose une méthode d'investigation sur les origines possibles de ces détections enregistrées au niveau du système SSI, par l'utilisation des rapports d'activité isotopique calculés à partir des concentrations d'activité. Huit (8) échantillons prélevés du 15 au 23 Mai 2010 ont été analysés. Il en résulte que l'évènement nucléaire ayant conduit à ces libérations ou rejets des radionucléides est lié probablement à une explosion nucléaire survenue entre le 11 et le 13 Mai 2010 en Corée du Nord.

I. INTRODUCTION

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 for atmospheric tests, seismic for underground tests, hydro acoustic for underwater tests and radionuclides for all environments. The Comprehensive Nuclear Test Ban Treaty (CTBT) is not yet interred into force, but the Verification Regime is already almost established. Among the monitoring station established, 80 are radionuclides and of these, 40 are equipped with noble gases detection system. This radionuclides network includes 16 certified radionuclide laboratories. They may evaluate filter samples further if requested (Schulze, Auer, and Werzi

2000). Regarding the records in the radionuclides stations, the data are sent by ARR and RRR format, and the results are given in activity concentration (in Bq/m3). To assess efficiently the timing information, we have to use the activity ratios (Axelsson and Ringbom 2014). There is a fundamental difference between these two quantities (activity concentration ratio and activity ratio), because activity concentration is not calculated with the formula ‘’Measured activity/sampled air volume’’. Our previous study (Yamba et al. 2016) investigated only on one (01) available sample (sample1) collected on 15 May 2010 by using directly the activity ratio. In this paper, eight (08) samples are used to convert La-140 and Ba-140 activities concentration ratios in activities ratios in order to evaluate the age of nuclear release. The La-140 and Ba-140 data used in this paper were recorded at JPP37 between 15 May and 23 May 2010.

II. METHODOLOGICAL APPROACH

Ba-140 decays by beta minus emissions to the excited level of La-140, and hence La-140 decays by beta minus emissions to the Ce-140 level. The radioactive activity of these disintegrations can be evaluated using spectroscopic measurements such as gamma spectrometry

II.1. Activity calculation

ARR (Automated Radionuclide Report) and RRR (Reviewed Radionuclide Report) give always, after sampling, processing and acquisition, activity concentration value in Bq/m3. We can extract from them the activities values in order to make an assessment of the age. Globally, the activity concentration value is given as (Axelsson and Ringbom 2014):

$$C = \frac{n \cdot \lambda}{\varepsilon \cdot (1 - e^{-\lambda t_A})} \cdot \frac{\lambda}{e^{-\lambda t_p}(1 - e^{-\lambda t_c})} \cdot \frac{1}{S} \qquad [1]$$

Where C is the activity concentration in Bq/m3, ε is the efficiency of detector, λ is the decay constant, n is the net peak area, S is the sampling rate, t_A is the measurement acquisition time, t_p is the sample processing time, t_C is the sample collection time.

In view of that the measured activity is given by:

$$A = \frac{n \cdot \lambda}{\varepsilon \cdot (1 - e^{-\lambda t_A})} \qquad [2]$$

So, we can get from the activity concentration, the measured activity as:

$$A = \frac{C \cdot S}{\lambda} \cdot e^{-\lambda t_p}\left(1 - e^{-\lambda t_c}\right) \qquad [3]$$

Regarding the calculation of La-140 and Ba-140 activities, we have to use these two following equations:

$$A_{Ba} = \frac{C_{Ba} \cdot S}{\lambda_{Ba}} \cdot e^{-\lambda_{Ba} t_p}\left(1 - e^{-\lambda_{Ba} t_c}\right) \qquad [4]$$

And

$$A_{La} = \frac{C_{La} \cdot S}{\lambda_{La}} \cdot e^{-\lambda_{La} t_p}\left(1 - e^{-\lambda_{La} t_c}\right) - A_{Ba} \frac{\lambda_{La}}{\lambda_{La} - \lambda_{Ba}} \left(\frac{\lambda_{La}}{\lambda_{Ba}} \frac{1 - e^{-\lambda_{Ba} t_A}}{1 - e^{-\lambda_{La} t_A}} - 1 \right) \qquad [5]$$

II.2. Activity ratio calculation

After the evaluation of isotopic activities of La-140 and Ba-140, we can now calculate the isotopic activity ratio. The measured activities ratio $r = \frac{A_{La}}{A_{Ba}}$ is given as:

$$r = \frac{C_{La}\lambda_{Ba}}{C_{Ba}\lambda_{La}} \frac{1 - e^{-\lambda_{La} t_c}}{1 - e^{-\lambda_{Ba} t_c}} e^{(\lambda_{Ba} - \lambda_{La}) t_p} - \frac{\lambda_{La}}{\lambda_{La} - \lambda_{Ba}} \left(\frac{\lambda_{La}}{\lambda_{Ba}} \frac{1 - e^{-\lambda_{Ba} t_A}}{1 - e^{-\lambda_{La} t_A}} - 1 \right) \qquad [6]$$

By making a simplification, we obtain:

$$r = K_1 K_2 - K_3 K_4 \qquad [7]$$

With

$$\Delta r = \sqrt{((K_2 \cdot \Delta K_1)^2 + (K_1 \cdot \Delta K_2)^2 + (K_4 \cdot \Delta K_3)^2 + (K_3 \cdot \Delta K_4)^2)} \qquad [8]$$

Where:

$$K_1 = \frac{C_{La}\lambda_{Ba}}{C_{Ba}\lambda_{La}} \qquad [9]$$

$$\Delta K_1 = K_1 \sqrt{\left(\left(\frac{\Delta C_{La}}{C_{La}}\right)^2 + \left(\frac{\Delta C_{Ba}}{C_{Ba}}\right)^2 + \left(\frac{\Delta \lambda_{La}}{\lambda_{La}}\right)^2 + \left(\frac{\Delta \lambda_{Ba}}{\lambda_{Ba}}\right)^2 \right)} \qquad [10]$$

$$K_2 = \frac{1 - e^{-\lambda_{La} t_c}}{1 - e^{-\lambda_{Ba} t_c}} e^{(\lambda_{Ba} - \lambda_{La}) t_p} = \frac{e^{-\lambda_{La} t_p} - e^{-\lambda_{La}(t_c + t_p)}}{e^{-\lambda_{Ba} t_p} - e^{-\lambda_{Ba}(t_c + t_p)}} \qquad [11]$$

$$\Delta K_2 = \sqrt{\left(\left(\frac{\left(e^{-\lambda_{La} t_p} - e^{-\lambda_{La}(t_c + t_p)}\right) \cdot \left(t_p \cdot e^{-\lambda_{Ba} t_p} - (t_c + t_p) \cdot e^{-\lambda_{Ba}(t_c + t_p)}\right)}{\left(e^{-\lambda_{Ba} t_p} - e^{-\lambda_{Ba}(t_c + t_p)}\right)^2} \cdot \Delta\lambda_{Ba} \right)^2 + \left(\frac{-t_p \cdot e^{-\lambda_{La} t_p} + (t_c + t_p) \cdot e^{-\lambda_{La}(t_c + t_p)}}{e^{-\lambda_{Ba} t_p} - e^{-\lambda_{Ba}(t_c + t_p)}} \cdot \Delta\lambda_{La} \right)^2 \right)} \qquad [12]$$

$$K_3 = \frac{\lambda_{La}}{\lambda_{La} - \lambda_{Ba}} \qquad [13]$$

$$\Delta K_3 = K_3 \cdot \sqrt{\left(\left(\frac{\Delta\lambda_{La}}{\lambda_{La}}\right)^2 + \left(\frac{\sqrt{(\Delta\lambda_{La})^2 + (\Delta\lambda_{Ba})^2}}{\lambda_{La} - \lambda_{Ba}}\right)^2 \right)} \qquad [14]$$

$$K_4 = \frac{\lambda_{La}}{\lambda_{Ba}} \frac{1 - e^{-\lambda_{Ba} t_A}}{1 - e^{-\lambda_{La} t_A}} - 1 \qquad [15]$$

$$\Delta K_4 = \sqrt{\left(\left(\frac{\lambda_{La}}{1 - e^{-\lambda_{La} t_A}} \cdot \frac{\lambda_{Ba} \cdot t_A \cdot e^{-\lambda_{Ba} t_A} - (1 - e^{-\lambda_{Ba} t_A})}{(\lambda_{Ba})^2} \cdot \Delta\lambda_{Ba} \right)^2 + \left(\frac{1 - e^{-\lambda_{Ba} t_A}}{\lambda_{Ba}} \cdot \frac{(1 - e^{-\lambda_{La} t_A}) - \lambda_{La} \cdot t_A \cdot e^{-\lambda_{La} t_A}}{(1 - e^{-\lambda_{La} t_A})^2} \cdot \Delta\lambda_{La} \right)^2 \right)} \qquad [16]$$

Thus, we can calculate with its uncertainty, the isotopic activity ratio by using the equations above. The activity concentrations are measured with the same detector at the radionuclides station JPP37, and so, our calculation parameters can be directly applied for all the samples recorded. For the assessment of the age, the reference time is considered as the beginning time of acquisition. Next table (table 1) shows a summary of measurement parameters and the measured activity concentrations.

Table 1 :La140 and Ba140 Activities concentrations measured at the radionuclides station JPP37, at OKINAWA on May 2010.

COLLECTION START	SAMPLING TIME (h)	NAME	ACTIVITY µBq/m3
15-MAY-2010	24.00	BA-140	81,8582253
15-MAY-2010	24.00	LA-140	163,186919
16-MAY-2010	24.00	BA-140	22,6648369
16-MAY-2010	24.00	LA-140	42,6135387
17-MAY-2010	24.00	BA-140	27,5041321
17-MAY-2010	24.00	LA-140	59,0066529
18-MAY-2010	24.00	LA-140	69,4410997
18-MAY-2010	24.00	BA-140	28,1064215
19-MAY-2010	24.00	BA-140	50,8134929
19-MAY-2010	24.00	LA-140	110,623558
20-MAY-2010	24.00	BA-140	43,7932773
20-MAY-2010	24.00	LA-140	99,4365867
21-MAY-2010	24.00	BA-140	5,24481568
21-MAY-2010	24.00	LA-140	19,0249998
22-MAY-2010	24.00	BA-140	5,00346554
22-MAY-2010	24.00	LA-140	6,5478

By using Eq.6, we have evaluated from the measured activity concentrations, the activity ratio for each sample. The following table (table 2) shows the calculated activity ratios for the samples 1 to 8.

Table 2: Isotopic activity ratio is calculated from activity concentration measured at the radionuclides station JPP37, at OKINAWA on May 2010. C1 and A1 represent respectively the isotopic activity and the concentration of Ba-140. C2 and A2 represent respectively the isotopic activity and the concentration of La-140.

SAMPLE	COLLECTION START	CONCENTRATION RATIO (C2/C1)	ACTIVITY RATIO (A2/A1)
SAMP1	15-MAY-2010	1.9935 ± 9.86748e-2	0.9592692 ± 5.80296e-2
SAMP2	16-MAY-2010	1.8802 ± 9.30632e-2	0.8925988 ± 5.47295e-2
SAMP3	17-MAY-2010	2.1454 ± 10.61906e-2	1.0486 ±6.24495e-2
SAMP4	18-MAY-2010	2.4706 ± 12.22909e-2	1.2399 ± 7.19178e-2
SAMP5	19-MAY-2010	2.1771 ± 10.77585e-2	1.0672 ± 6.33716e-2
SAMP6	20-MAY-2010	2.2706 ± 11.23885e-2	1.1222 ± 6.60943e-2
SAMP7	21-MAY-2010	3.6274 ± 17.95467e-2	1.9201 ± 10.55889e-2
SAMP8	22-MAY-2010	1.3087 ± 6.47750e-2	0.5565085 ± 3.80939e-2

III. RESULTS AND DISCUSSIONS

IV.1. Using activity ratio for the dating

From the activity concentration given by ARR or RRR, we use the measured activities ratio for the assessment of the nuclear event age. In our case, we are interested by the measured activity concentrations of La-140 and Ba-140, recorded at the radionuclide station JPP37 on May 2010.
In our previous study on the fast and accurate assessment of the nuclear event zero-time (Yamba et al. 2016), we have proposed some nuclear constants that one can use directly in the calculation algorithms. This study has been done by taking into account some of nuclear databases, and this, in order to improve the calculation accuracy. The sample age is calculated using the following schemes:

$$t = U \cdot ln\left(\frac{r-A}{F-A}\right) \qquad [17]$$

where

$$\begin{cases} A = \frac{\lambda_{La}}{\lambda_{La}-\lambda_{Ba}} \; with \; \Delta A = \frac{\lambda_{La}}{\lambda_{La}-\lambda_{Ba}} \cdot \sqrt{\left(\frac{\Delta\lambda_{La}}{\lambda_{La}}\right)^2 + \left(\frac{\sqrt{(\Delta\lambda_{La})^2+(\Delta\lambda_{Ba})^2}}{\lambda_{La}-\lambda_{Ba}}\right)^2} \\ r \text{ is the measured activity ratio} \\ U = \frac{1}{\lambda_{Ba}-\lambda_{La}} \; with \; \Delta U = \frac{\sqrt{(\Delta\lambda_{Ba})^2+(\Delta\lambda_{La})^2}}{(\lambda_{Ba}-\lambda_{La})^2} \\ F = \frac{\lambda_{La}\gamma_{La}}{\lambda_{Ba}\gamma_{Ba}} \; with \; \Delta F = F\sqrt{(\frac{\Delta\lambda_{La}}{\lambda_{La}})^2 + (\frac{\Delta\lambda_{Ba}}{\lambda_{Ba}})^2 + (\frac{\Delta Y_{Ba}}{Y_{Ba}})^2 + (\frac{\Delta Y_{La}}{Y_{La}})^2} \end{cases} \qquad [18]$$

where γ_{Ba} and γ_{La} are respectively the cumulative fission yield of Ba-140 and the independent fission yield of La-140

$$J = \frac{r-A}{F-A} \; with \; \Delta J = J \cdot \sqrt{\left(\frac{\sqrt{(\Delta r)^2+(\Delta A)^2}}{r-A}\right)^2 + \left(\frac{\sqrt{(\Delta F)^2+(\Delta A)^2}}{F-A}\right)^2} \qquad [19]$$

So, the upper (t_{up}) and lower (t_{low}) of age limit are assessed using the following formulas:

$$\begin{cases} t = U ln(J) \\ t_{up} = (U+\Delta U) ln\,(J+\Delta J) \\ t_{low} = (U-\Delta U) ln\,(J-\Delta J) \end{cases} \qquad [20]$$

The following tables show the values of parameters used for dating of a nuclear explosion (see Table 3) or a continuous release (non-nuclear explosion) (see Table 4).

Table 3 : Parameters used for a fast assessment of nuclear explosion zero time

	U	A	F
$ENDF.B.VII.I$	-2.7888	1.1516	$6.3759e^{-3}$
	$\pm 2.419154e^{-4}$	$\pm 1.294469e^{-3}$	$\pm 4.0811e^{-3}$
$LARA - LNHB/CEA$	-2.7886	1.1516	$6.3766e^{-3}$
	$\pm 3.512159e^{-4}$	$\pm 1.861144e^{-2}$	$\pm 4.0815e^{-3}$

Table 4 : parameters used for a fast assessment of continuous nuclear release zero time

	U	A	F
$ENDF.B.VII.I$	-2.7888	1.1516	1
	$\pm 2.419154e^{-4}$	$\pm 1.294469e^{-3}$	± 0
$LARA - LNHB/CEA$	-2.7886	1.1516	1
	$\pm 3.512159e^{-4}$	$\pm 1.861144e^{-2}$	± 0

This plot (figure 1) shows the difference of behavior between a nuclear explosion scenario and a continuous nuclear release scenario. The dotted line represents a release from a nuclear reactor for example for radionuclides Ba140 and La140. During a sufficiently long time into the nuclear reactor, the activity ratio La-140/Ba-140 reaches its equilibrium level which approach 1. So, in this case, the activity ratio allowing to date the nuclear event starts to 1, and reaches the equilibrium level 1.1516. The solid line represents a case of nuclear explosion, where the activity ratio starts to 0 and reaches its equilibrium level at 1.1516.

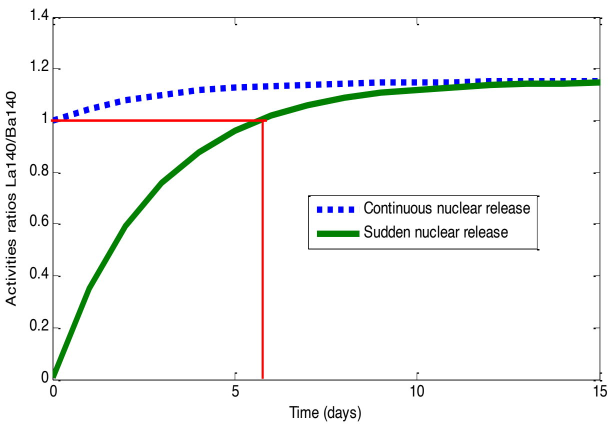

Figure 1 : Change in over time, the activity ratio in the case of nuclear explosion and in the case of a continuous release.

Before estimating the sample age, we need to analyze all the data in order to evaluate the possible kind of nuclear event. Two kind of nuclear events are considered: a sudden release (nuclear explosion scenario) and a continuous release. Next table (table 5) gives an estimation of various scenarios of release by analyzing of activity ratios. YES means a possibility according to the activity ratio value. For a nuclear explosion, the activity ratio must be between 0 and 1.1516 (equilibrium level), and for a continuous release, it must be between 1 and 1.1516.

Table 5 : Discrimination of nuclear explosion against a continuous release scenario.

SAMPLE	ACTIVITY RATIO (A2/A1)	POSSIBLE SUDDEN RELEASE	POSSIBLE CONTINUOUS RELEASE
SAMP1	0.9592692 ± 5.80296e-2	YES	NO
SAMP2	0.8925988 ± 5.47295e-2	YES	NO
SAMP3	1.0486 ±6.24495e-2	YES	YES
SAMP4	1.2399 ± 7.19178e-2	NO	NO
SAMP5	1.0672 ± 6.33716e-2	YES	YES
SAMP6	1.1222 ± 6.60943e-2	YES	YES
SAMP7	1.9201 ± 10.55889e-2	NO	NO
SAMP8	0.5565085 ± 3.80939e-2	YES	NO

After the discrimination of the kind of nuclear event, we calculate the sample age under the conditions of release give in Table 3 and Table 4. These conditions of release (related to the initial activity ratio) can be also see in Figure 1. Table 6 shows the results of estimations according to the kind of scenario.

Table 6 : This table shows the results of our calculations according to the case of release.

SAMPLE	COLLECTION START	ACQUISITION START	Zero-time in the case of Sudden release	Zero-time in the case of Continuous release
SAMP1	15-MAY-2010	17-MAY-2010	12-MAY 01:00 +17h/-24h	
SAMP2	16-MAY-2010	18-MAY-2010	13-MAY 21:00 +12h/-15h	
SAMP3	17-MAY-2010	19-MAY-2010	12-MAY 07:00 +31h/-62h	no consistent
SAMP4	18-MAY-2010	20-MAY-2010		
SAMP5	19-MAY-2010	21-MAY-2010	13-MAY 18:00 +37h/-93h	no consistent
SAMP6	20-MAY-2010	22-MAY-2010	11-MAY 19:00 +78h/-...	17-MAY 11:00 +78h/-...
SAMP7	21-MAY-2010	23-MAY-2010		
SAMP8	22-MAY-2010	24-MAY-2010	no consistent	

We notice that, according to the zero-time values, Sample1 and Sample3 would come from the same event. Also, Sample2 and Sample5 would come from the same event. This event is considered as a sudden nuclear release, like a nuclear explosion. These two hypotheses are very consistent because it is not possible to consider that these four samples come from a continuous nuclear release. The expression ‘’no consistent’’ means that, the calculated age is less than 2hours. A calculated age less than 2hours means that the nuclear event has been performed inside the measurement facilities, and that is no consistent.

III.2. Discussion

This Study has been done using 8 samples recorded in May 2010 at the radionuclide station JPP37. Data used in this study are activities concentrations, which are thus converted in isotopic activities.
Among these eight samples, two (Sample4 and Sample7) cannot give a reasonable age because the measured activities ratio are greater than the level equilibrium which is 1.1516. The calculated ages from Sample2 and Sample5, and from Sample1 and Sample3 seem to characterize a same nuclear event, like a nuclear explosion.
In all cases, it is very clear that four of these eight samples seem to indicate the same nuclear event, and this nuclear event is a nuclear explosion occurred between 11 May and 13 May. We can also notice that, by using data based on nuclei ratio, Lars Erik DE GEER in his interesting study (De Geer 2012, 2013) gave an estimated date of 11 May 2010 17:00. The same sample has been investigated by others authors (Ihantola, Toivonen, and Moring 2013) based on activity ratios. In our previous study (Yamba et al. 2016) on the same sample (sample1), by using directly the activity ratio and taking into account the numerical difference between some radioactive data from four databases, the estimated date is found to be 11 May 2010, 10:00. This estimation was done using the weighted least-squares method (Björck 1990).

IV. CONCLUSION

In this study, we have analyzed eight samples recorded at the radionuclides station at OKINAWA-Japan. We have firstly converted the measured data from the activity concentration data into the isotopic activity. We have then analyzed these data according to the two possible release scenarios: a sudden release for a nuclear explosion and a continuous release. Our best conclusion regarding the kind of nuclear event and the age, is that the nuclear event would be a nuclear explosion performed between 11 may and 13 may. This work could be improve by taking into account others monitoring technologies used by the IMS, like seismic.

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