



Preliminary Measurements of Be-10/Be-7 Ratio in Rainwater for Atmospheric Transport Analysis

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ARTICLE INFORMATION

Received: October 10, 2019

Accepted: January 22, 2020

Published online: February 28, 2020

Keywords:

Rainwater, Beryllium-7, Beryllium-10, AMS, HP-Ge



DOI: [10.15415/jnp.2020.72018](https://doi.org/10.15415/jnp.2020.72018)

ABSTRACT

The meteoric cosmogenic beryllium has been used as an essential geophysical tracer in the analysis of atmospheric flows and erosion soils since 1960. The first measurements Be-7 and Be-10 concentrations in rainwater from Mexico, have been carried out by using gamma decay spectroscopy and AMS techniques, respectively for each isotope. With this it was possible to report a preliminar value for the Be-10/Be-7 isotopic ratio in such environmental samples. The present work described preliminary results related to rainwater collected at mountain and metropolitan areas. Results are compared with predictions and previous measurements for both radioisotopes, observing a very sensible behavior particularly for the case of Be-7 activities.

1. Introduction

The nuclear and radiation physics development has provided useful experimental methodologies and techniques to solve several problems at different science disciplines. One of the most important applications of nuclear applied physics is the use of radionuclei and its radioactivity as a powerful tool for the Environmental Science and Earth Science [1].

The Be-7 and Be-10, are natural radionuclei produced by spallation reactions among the neutron component and the secondary cosmic rays with the nitrogen and oxygen nuclei, present in the atmosphere. Such a production is given in a ratio of 2/3 and 1/3 in the stratosphere and troposphere, respectively [2, 3]. Due to their production depends of the amount of secondary cosmic rays, to attain a good interpretation and analysis, it is important to take in consideration the modulation factors of cosmic rays that make variable the production. These factors are: astrophysical events (which increase the cosmic ray flux), the solar activity, the geomagnetic field and, the atmosphere height. Be-10 has a half-life of 1.387 ± 0.012 My [4, 5] and Be-7 of 53.3 days. Once this radionuclei are produced, it is assumed the beryllium is oxidized and absorbed in the aerosols surface. The residence time in the stratosphere is one or two year approximately, whereas in the troposphere is in the order of weeks (21 days, approximately), so almost all Be-7 deposited

in the earth surface has its origin in the troposphere [6-9]. The removal of the atmospheric beryllium occurs by dry deposition as well as by scavenging. The scavenging is carry out in two ways, in-cloud and below cloud, and this dry deposition contains the dissolved Be as well as the non-dissolved one (dust). Once are removal from the atmosphere, the Be isotopes are deposited in the lithosphere and they constitute environmental records which can be used to explain climatologic and geophysical phenomena.

The atmospheric transport and the interchange of air masses in the atmosphere are affected by the season variability and the meteorological regional changes [10, 11]. The dry and wet depositions determine the beryllium concentration in the environmental records, due to both isotopes are transported till the earth surface in similar ways. The proportions of dry and wet deposition may change depending of the region where the environmental record will be localized. According to the studies developed, there are different enrichment sources for the Be-10, due to its long half-life. Among this sources is the resuspended Be-10, the transported by air masses to other places and the coming from the solar system. For these reasons, it is established that one of the ways to identified or to exclude information for the analysis of environmental records is by using the Be-10/Be-7 ratio [10, 12].

To measure such radionuclei, commonly is used the combination of two different techniques: the Accelerator Mass Spectrometry (AMS), for the case of Be-10 and gamma ray spectroscopy, to measure the Be-7 taking advantage of its gamma decay [13].

In this work is presented a preliminary discussion regarding Be-7 data achieved in rain samples from the Alzomoni Mountain, and Ciudad Universitaria-Centro de Ciencias de la Atmósfera, Mexico City. In parallel it was developed an estimation of the Be-10 concentration which should be achieved for rain samples, taking into account prediction models. Finally, it was make a comparison with the Be-10 concentration in rain for a previous sampling campaign (1 year before) in one of the region here reported.

2. Methodology

The first step was to make a correct characterization of a ultra-low background gamma array, which consist of an HPGe detector fully covered by a lead shielding, both performed by CAMBERRA. Such array is part of the spectroscopy systems at the Radiation Laboratory II of the Science Faculty from Universidad Nacional Autónoma de México. To carry out the characterization, pilot rain samples as well as different gamma calibration source were used. Once the whole array was characterized and calibrated, the main samples of interest started to be measured. In the following sections, the main characteristics of this study are described in detail.

Additionally, for comparison purposes, a previously measured rain sample analyzed with AMS technique is as well reported here. Description of the detailed process for this kind of measurement can be found in [14, 15].

3. Rainfall Information

As it was mention in the previous section, to perform this preliminary study, rain samples from the Observatorio Meteorológico Alzomoni, State of Mexico (19.1187°N, 98.6552°W, Altitude: 3,985 m.a.m.s.l) and the Observatorio Meteorológico of Centro de Ciencias de la Atmósfera, UNAM, Mexico City (19.3262 °N, 99.1761 °W, Altitude: 2,280 m.a.m.s.l) were taken, particularly this latter, during the contingency period caused by Popocatepetl volcano ashes (April 22nd to May 2nd 2019). Samples were transferred to Nalgene containers previously acidified in order to maintain rain in acid pH to avoid the aging of the sample. Later, the samples were deposited in a single marinelli to take them to the interior of the detection system. Such samples were not previously processed or filtered, considering the requirement of a first measure taking into account all the natural compounds

of the sample, either suspended material, transported material for other air sources or by extraterrestrial particulated material.

4. Measurements of the Rain Samples

The sample measurements is carry out considering the isotopes to measure; for the specific case of this test, the measure procedure was divided in two parts: the first consisted in the simple preparation and its Be-7 measurement with the HPGe detector; the second consisted in the sample preparation to be later measured by using the AMS technique.

The first measurement does not imply a destructive technique, the samples were just acidified and carefully transported to the marinelli to be measured. Later on, to carry out the second part of the procedure, the samples were taken to a Radiochemical Laboratory where they were processed by using cationic interchange resins, with the aim to separate and purify the Be to be measured with AMS technique. This second part of the study is still in progress.

For this reason, this work is particularly centered on the samples measurements developed with the Germanium detector, to achieve the Be-7 concentrations. One of the most important thing to be considered for the Be-7 analysis is the natural background in the gamma spectrum, which can be manage the detection limits. During the measurement procedure may appears signals interfering a clear data tacking. The Compton Effect, occurs for instance when the high energy gammas eject an electron with just a fraction of their total energy, producing a “count” in the spectrum along with a low energy gamma which could be also registered by the detector. Hence, the high energy photons (> 500 keV) from the disintegration pattern of U, Th, K and cosmic rays, make random noise in the region of the gamma spectrum where the Be-7 signal is expected. In most of the measurements developed with HPGe detectors, a shielding array of more than 4” thickness is used, in order to reduce the background produced in the region of interest, allowing the identification of the real gamma decay related to the process to be measured [16].

Commonly, the Be-7 is measured in environmental samples by means of low background techniques. The Be-7 decays in Li-7 via electron capture (86%) emitting extremely low energy X-rays to lays directly to the ground state, and to the first excited state of Li-7 (10.4%) emitting a gamma of 477.6 keV. Usually the more used technique is the detection with a germanium crystal, which is recommendable due to its good energy resolution, allowing for instance, the separation of gammas coming from the Th and Ra decay, which appears in very high and very low rates, respectively, in the 480-487 keV energy range [8].

between data achieved is observed. By using these values is possible to calculate the detection efficiency for Be-7 (see Fig. 4), considering the counts resulting for the isotope (following section), getting a value of 0.0014, which is in agreement with other ones found in the literature [17].

5. Results and Discussion

After to establish the calibration and the good characterization of the detection array, the rain water samples were measured for different time periods. In the Figure 5 (a) and (b) are shown the typical spectra obtained during this measurements for the two different latitudes, Alzomoni Observatory (State of Mexico) and CCA-UNAM, Ciudad Universitaria (Mexico City). In the region around 400 keV can be easily observed the peak related to Be-7 events in both plots, demonstrating the validity of our setup to discriminate the radioisotope. The background and fit of the spectra can be seen in different colors than the experimental data. The comparison between background and experimental data shows a well identified Be-7 peak which did not appear in the first one. With this results it was possible to estimate the concentration of the radionucleus in the rain samples.

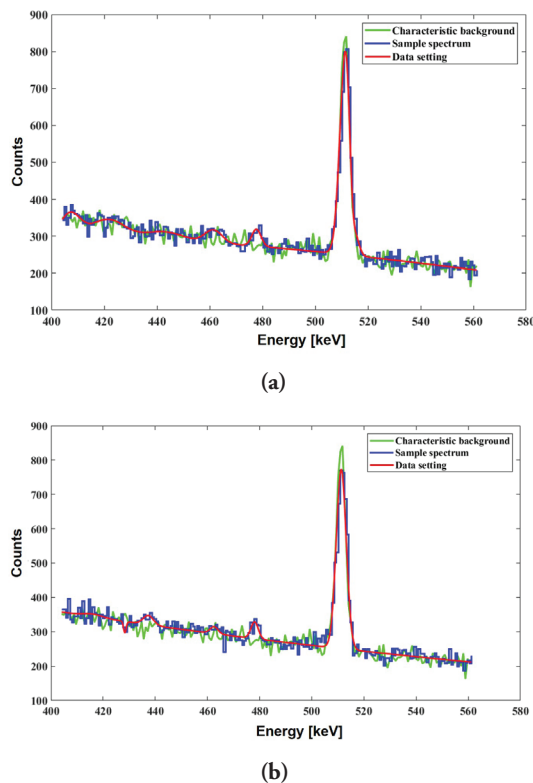


Figure 5: Rain samples gamma spectra where Be-7 events are well identified. (a) Alzomoni mountain, State of Mexico and (b) Centro de Ciencias de la Atmósfera, UNAM, Ciudad Universitaria, Mexico

City. In blue the experimental data, compared with background in green. A fit of data is included in red, showing a very acceptable identification of the gamma coming from Be-7 decay at 477.6 keV.

In Table 1 are presented the experimental values, obtained after the extraction of the Be-7 concentrations coming from rain samples described before. “ALTZ” is referred to Alzomoni rain samples and “CCA-UNAM” to the samples collected at Ciudad Universitaria. The Table shows the value of nominal gamma energy ($E_{\gamma T}$) the measured value ($E_{\gamma m}$), the efficiency for Be-7 (as it was estimated with the fit shown in Fig. 4) gamma decay (ϵ), the estimation for the activities (A) and the concentrations achieved (C).

Table 1: Activities and concentrations found after the analysis of Be-7 samples from two different regions in the Mexican territory: Mountain (ALTZ ~ 4000 m.a.m.s.l) and high city (CCA-UNAM ~ 2300 m.a.m.s.l).

Sample site	$E_{\gamma T}(\text{keV})$	$E_{\gamma m}(\text{keV})$	$\epsilon(E_{\gamma})$	(Bq/L)	C (atoms/g)
ALTZ	477.621	478.1 (0.4)	1.4×10^{-3}	9.40 (4)	6.23 (4)
CCA-UNAM		477.7 (0.5)		33.72 (4)	22.37 (4)

According to the concentrations estimated for Be-10 and Be-7 reported by Nagai et. al., [8] for production rates of such radioisotopes in the atmosphere, the expected concentrations for Be-10/Be-7 ratio should be ~ 2.0 for the North hemisphere. However, other estimations developed using global atmospheric models [3] establish the Be-10/Be-7 should oscillate between 1.1 in autumn, when the Stratosphere-Troposphere Exchange (STE) is minimum and 1.5 in spring, when the STE is maximum. Particularly in the tropics, the STE expected is low and practically null. With this last consideration, the expected value for the Be-10/Be-7 ratio could be lower than 1, for the latitudes considered in this work.

The experimental value achieved using AMS technique at the Laboratorio Nacional de Espectrometría de Masas con Aceleradores (LEMA) IF-UNAM, for Be-10 concentration from Alzomoni mountain rain samples, taken in the same season than the one here analyzed for Be-7 (Table 1) but, in the previous year (2018), is of $2.15 \pm 0.26 \times 10^{-4}$ atoms/cm². With this value, the Be-10/Be-7 ratio achieved is of 0.34, a number much lower than the estimated in the predictions reported in [8]. Taking in to account the considerations mentioned in the previous paragraph, the ratio is possibly been affected by tropical latitudes climate conditions. AMS studies of the present samples and a more complete series of samples has to be studied to establish

whether present results are constant and consistence for the latitudes here analyzed.

In order to evaluate the results obtained for Be-7 activity of the samples measured, a number of studies for different regions around the work are shown in Table 2. The results for the highest site (ALTZ) are well comparable with many of the activities found at different places and latitudes, for instance Chilton and Milford Haven [18], Southeastern Australia [22], Cantabria [24] and Geneva [25]; besides, they falling inside the range reported for the northern Loess Plateau in China [10] and Galveston, Texas [21]. This last two regions reported the highest activities for rainfall Be-7, which as well are in agreement for the values measured for the lowest site (CCA-UNAM), here reported. Nevertheless, there are many other places where a lower activity is reported, oscillating between 0.2 and almost 3 Bq/L [19, 20, 23, 26-29].

For the first (larger activities) and second (lower activities) groups, it is not apparently simple to justify some kind of relationship regarding latitudes or seasons. Then the message is there not exist a well-known behavior regarding Be-7 concentrations found for rain water around the world, appearing notorious differences when a group of measurements reported in different periods and latitudes are compared. Knowing this, it seems the exchange between different atmospheric layers is very sensible to any variable, at least the season, the latitude, the altitude and the year, as far as this preliminary study is showing. More and long campaigns of sampling at different latitudes has to be performed as well as to develop more complex models, in order to find more mechanisms to give better criteria to describe the behavior of the rainfall Be-7.

Table 2: Compilation of Be-7 activities measured in rainfall at different geographical sites.

Location	Reference	(Bq/L)
Chilton and Milford Haven (UK)	Peirson (1963) [18]	0.02–5.9
Bermuda	Turekian et al. (1983) [19]	1.57
Lake Zurich (Switzerland)	Schuler et al. (1991) [20]	2.43
Galveston, TX (USA)	Baskaran et al. (1993) [21]	0.11–24.84
Southeastern Australia	Wallbrink, and Murray (1994) [22]	0.02–5.9
Onagawa(Japan)	Ishikawa et al. (1995) [23]	1.54–1.60
Cantabria (Spain)	Ródenas et. al. (1997) [24]	0.73–5.05

Geneva (Switzerland)	Caillet et al. (2001) [25]	0.93–10.45
Detroit, MI (USA)	McNeary and Baskaran (2003) [26]	2.87
The northern Loess Plateau (China)	Zhang et al. (2013) [10]	1.2 – 20.8
Hsinchu, Taiwan (China)	Chao et al. (2014) [27]	0.2 – 1.3
Malaysia	Sharib et al. (2015) [28]	0.2 – 2.26
Southwest UK	Taylor et al. (2016) [29]	1.56 - 2.67

Conclusions

The present work shows a preliminary study related with the extraction of Be isotopes from rain samples, taken at two different regions at Mexico. Be-7 was measured in rain samples at mountain and metropolitan areas, 4000 and 2300 m.a.m.s.l., respectively. The concentration found for the mountain region was used to estimate the Be-10/Be-7 ratio, taking a previously measured concentration of Be-10 for the same region. The resultant ratio is much lower than the estimated by using predictions reported in literature. The lower value may be attributed to the expected low stratosphere-troposphere exchange at tropical latitudes. Further measurements of both Be isotopes must be carry out, in order to define whether this behavior is consistent for similar samples taken in similar seasons.

Particularly, in the case of Be-7 activity, data from different works taken at different latitudes how large oscillations (since 0.02 to 24.84 Bq/L), not finding in a simple way, some kind of relationship for this variations. The preliminary results here reported shows a good agreement with between 2 and 6 of the values previously reported, validating the methodology used. However, the notorious discrepancies between the different activities revised is pointing the scientific need of more and detailed studies, not sampling campaign only but also, the improvement of existent models to predict the behavior of Be-7 isotope in a global way, since it appears in the atmosphere till arrives to the earth by rainfall. Evidently, the most experimental results there are, most precise boundaries for calculations. A most extended number of measurements for Be isotopes at the Mexican territory will be presented in future works.

Acknowledgements

This work was partially supported by the CONACyT LN-280769, LN-294537, and the DGAPA-PAPIIT IG100313, IA101616 and IA103218 Projects. The authors want to

thank the valuable help of Arcadio Huerta Hernández and Santiago Padilla Domínguez for his technical support.

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Journal of Nuclear Physics, Material Sciences, Radiation and Applications

Chitkara University, Saraswati Kendra, SCO 160-161, Sector 9-C,
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Volume 7, Issue 2

February 2020

ISSN 2321-8649

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