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To cite this article: K. De Los Ríos *et al* 2018 *J. Phys.: Conf. Ser.* **1078** 012009

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Characterization of the LEMA isotope separator to measure concentrations of ^{10}Be from atmospheric filters

K. De Los Ríos, C. Méndez-García, S. Padilla, C. Solís, E. Chávez, A. Huerta and L. Acosta

Instituto de Física. Universidad Nacional Autónoma de México. Apartado Postal 20-364 Cd.Mx 01000, Mexico.

E-mail: acosta@fisica.unam.mx

Abstract. The understanding and measurement of ^{10}Be is useful for earth science. Its way of production, deposition and mainly half-life allow phenomenological reconstructions of great interest on this area. In order to characterize the 1 MV isotope separator of the National AMS Laboratory LEMA (Laboratorio de Espectrometría de Masas con Aceleradores) at Mexico City for the measurement of ^{10}Be concentrations, atmospheric filters with aerosols content were used. The filters were processed with a radio-chemical extraction procedure detailed here. Then, the prepared samples were pressed in cathodes and measured by using the AMS technique, tuning the machine till focus the optics on the beryllium peak. The preliminary statistical analysis of the $^{10}\text{Be}/^9\text{Be}$ ratios obtained from this characterization is presented in this work. Two statistical methods were used in order to improve the data information. With them, statistical stability behavior is observed on different campaigns of measurements. The results obtained comparing both methods are here discussed.

1. Introduction

The Accelerator Mass Spectrometry technique (AMS) was created in the early 80's in order to measure rare and long-lived radioisotopes, which could not be measured using conventional mass spectrometry or decay counting techniques [1]. Among the different radioisotopes that have been measured using the AMS technique, it has been found that ^{10}Be is of a relevant importance for the development of different sciences [2]. ^{10}Be is a radioisotope that is produced mainly in the atmosphere, by spallation reactions with N_2 and O_2 . The ^{10}Be has a half-life of 1.5×10^6 years [3], it remains approximately 12 months in the stratosphere, from where it passes to the troposphere staying there for about 2 weeks and is finally deposited on the Earth surface by wet or dry deposition [4]. For this reason, it is considered as a potentially useful tracer to be studied.

In this work, the characterization of the isotope separator of the LEMA IF-UNAM [5] was carried out for the study of ^{10}Be concentrations in natural samples. The samples chosen were atmospheric filters which were processed making small modifications to the radiochemical procedures established in the literature for the extraction of the radioisotope from this kind of samples [6, 7]. Four different campaigns of measurements were performed at LEMA, calibrating the separator by using reference standards (Labeled as STD**) and blanks (Labeled as BLK**).



For data analysis the method exploration data analysis (EDA) was used [8]. As a measure of central tendency the median was chosen and as a spread, the median absolute deviation (MAD) [9] was determined. To validate the results, a comparison with the traditional methods was made as well. This kind of analysis allows to determine the ^{10}Be concentration existing on each processed sample. However in this preliminary results just the $^{10}\text{Be}/^9\text{Be}$ ratios for the statistical analysis obtained are presented. In order to demonstrate the success of the characterization, a preliminary comparison of some ^{10}Be concentrations obtained from two different campaigns of measurements, is also presented.

2. Methods

2.1. Sample extraction and preparation technique.

The radiochemical extraction was carried out with atmospheric filters collected in 2012 [10]. Mainly the filters of the "Ciudad Universitaria" (Labeled as CU**) and the "Intituto Mexicano del Petroleo" (Labeled as T0**) site were chosen (details regarding samples are well described in [10]). The radiochemical extraction is divided in three main steps: sample preparation, separation and purification of the Be and obtention of the BeO molecule, necessary to produce the negative ions of BeO^- in the ion source to be processed with the AMS technique (see Fig. 1). The radiochemical procedure established here is the result of small modifications (as it is shown in Fig. 1 diagram) of the described procedures found in the literature [6, 7].

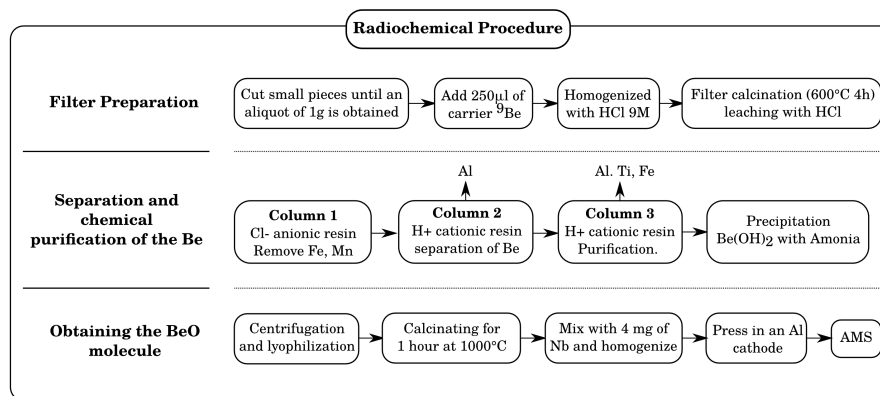


Figure 1. Radiochemical procedure adapted for the extraction of ^{10}Be from atmospheric filters in LEMA.

2.2. Measurement in the LEMA isotope separator.

For the isotope separation it is necessary to take into account the charge, mass and energy properties of the Be isotope, and thus to filter and carry the beam through the beam line, by electric and magnetic fields. The Isotope Separator of LEMA is a Tandetron of 1 MV with a complex optical system coupled. A detailed description of this kind of system can be consulted in [5]. Cathode filled with processed sample are introduced in a carousel of up to 50 positions and then are transported (one by one) to the ion source, where the beam of negative ions is produced. To perform the tuning, the stable isotope beam (^9Be) is initially focused and the current I is measured in a Faraday cup close to the detection system. Then it is possible to tune the radionuclide of interest (^{10}Be) modifying the appropriate parameters in the accelerator. The ^{10}Be beam is undetectable by the Faraday cups of the accelerator. For this reason it has to be identified and counted event by event, in a gas detector mounted at the end of the line. The

final filtration of the beam is carried out there. Finally the ΔE -E technique in the gas detector is used in order to observe well discriminated, events of ^{10}Be . To improve the discrimination between ^{10}Be and its isobar (^{10}B), a 75 nm thick absorber foil is introduced after the electrostatic analyzer. The whole procedure is carried out both for the cathode filled with STD51 (nominal value $^{10}\text{Be}/^9\text{Be} = 2.709 \times 10^{-11}$ [11]) and for the cathode with the sample. It is necessary to introduce a blank cathode (BLK**), with carrier material chemically processed in the same way as the sample, to determine the background level of the technique and to obtain the best possible calibration. The STD ratio in the isotopic separator should be between 8-10% of the nominal STD51 value.

2.3. Exploratory Data Analysis (EDA).

In the measurement process a large set of data is provided by the system. The ratios values at the beginning or at the end of the run may change. It is important to establish a statistical method for the data analysis that not be affected by those variations. The method of exploratory data analysis EDA is a favourable way to obtain information from a large data set. The EDA method is robust and resistant for making assumptions regarding the nature of a data set [8]. Within EDA, a reasonably simple complete alternative for the numerical summary is the median, as a measure of central tendency and the median absolute deviation (MAD), as a spread measure. In this case, the MAD is just the transformed value of $y_i(x) = x_i - q_{0.5}$ [9]:

$$MAD = \text{median}|y_i(x)| \quad (1)$$

where x_i is referred to each value of the data set and $q_{0.5}$ is the median of the set.

For the statistical analysis of this work, the graphical analysis was made by means of schematic diagrams [8]. This is an extension of the boxes and whiskers diagrams, since this method offers information on the "outliers" behavior, which must be observed for the analysis of this characterization. On this kind of diagram, the box represents the values where the 75% of the data from the statistical distribution is moving, while the whiskers indicate the maximum and minimum value of the data series. Those values that are out of the range established by the whiskers are the so called outlier values. A number of the series is considered an "outlier", when its value is more than 1.5 times the absolute value enclosed by the box, called the inter-quartile range (IQR). Then, the outliers are values so far from the expected behavior of the mean value.

3. Results

During the measurement with the AMS technique it is necessary to observe the ΔE -E spectra to be able to determine the validity of the data. This kind of plot shows the separation of the ^{10}Be and its isobar ^{10}B , due to the energy loss of the ions in the two anodes of the gas detector. The ΔE -E spectra observed during the tuning process are related to the measurement of: a standard, a blank and a natural sample.

In the first measurement made (April-2017) it was not possible to obtain the desired sequence of spectra since they were far to fulfill the requirements mentioned above. With these results, part of the radio-chemical extraction process pre-established in the literature was changed (as it is described on the Fig. 1). For the second measurements made (September-2017), the spectra were better adjusted to the aforementioned requirements. However, the spectrum of the sample showed a considerable background, evidencing some contaminant in the sample or a calibration problem in the system. For the measurements of November 2017 and February 2018 the radio-chemical technique was established (see Fig. 2) and it was possible to obtain a 10% of the nominal STD51 value, with a beam transmission of 57%. The behavior of the ΔE -E spectra allows to ensure that the radio-chemical extraction was correct and the detected counts effectively were related to ^{10}Be events and therefore the results of the ratios could be analyzed.

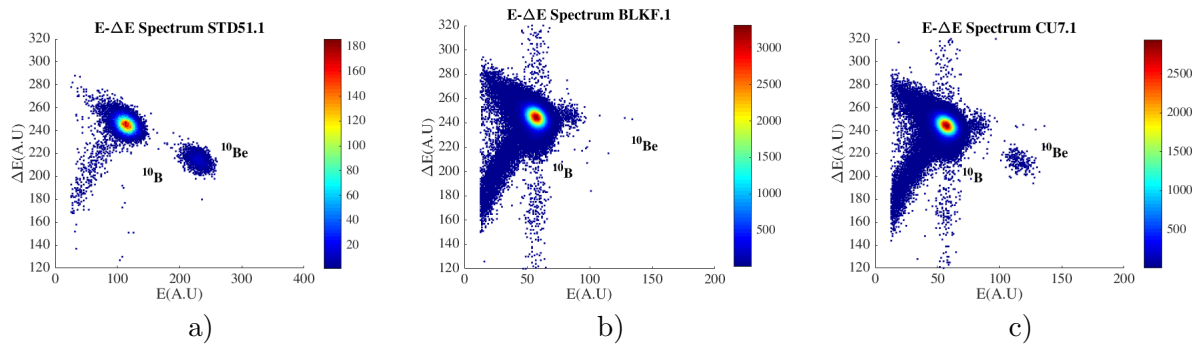


Figure 2. E- ΔE spectra observed with the AMS technique for the best campaign of measurements (February 2018) for an acquisition time of 5 minutes: a) STD51 with $^{10}\text{Be}/^9\text{Be}=1.43\times 10^{-12}$ b) Blank processed BLKF with $^{10}\text{Be}/^9\text{Be}=4.42\times 10^{-15}$ and c) Atmospheric sample processed CU7.1 with $^{10}\text{Be}/^9\text{Be}=2.19\times 10^{-13}$.

Since the AMS technique is destructive, the cathodes wear out and for this reason the values at the beginning or at the end of the run may change. To observe this influence on the $^{10}\text{Be}/^9\text{Be}$ ratios thrown by the system, a dispersion diagram was made showing the outliers using the EDA schematic plot graphic method (see Fig. 3). The mean and the standard error of each cathode involved in the run are also graphed in the Fig. 3. In the data series it is easy to distinguish the standard samples from natural samples and blanks. This behavior is the expected for a good characterization of the isotopic separator. For this reason, the data of Fig. 3 are those considered as the correct and better results, thus completing the characterization of LEMA separator for ^{10}Be natural samples.

The anomalous samples showed cathodes partially melted during the ionization process, so they do not represent a reliable measurement. In order to make a comparison between the different results, all experimental data coming from anomalous samples were removed from this analysis (purple-shaded in Fig. 3).

4. Discussion

4.1. Comparison of statistical methods for the different campaigns of measurements.

Performing the schematic plot for the radio-chemically processed samples (See Fig. 4 a), b) and c)), stability was observed in the dispersion data for the three plots: September-2017, November-2017, and February-2018. A better stability is observed in November-2017 and February-2018 plots, considering that more outliers appear in September-2017. This behavior can be explained, for the case of September-2017, as a problem of contamination in the radiochemical procedure.

For the three campaigns of measurements some of the cathodes were filled with material coming from a same sample. In this context it was possible to develop some systematic comparisons between the data resulting from the different campaigns after the AMS analysis (shaded in pink and green in Fig. 4). These data were identified for each campaign as TO3** and TO4** and then analyzed taking into account both, the traditional statistical method and the EDA method.

Figs. 5 a) and b) show in two different plots, the comparison between results coming from same samples but different campaigns: In Fig. 5 a) the plot shows the results of September-2017 measurements for the natural sample T03, on the right side of the dashed line; In Fig. 5 b) the plot shows the results of November-2017 measurements for the natural sample T04, as well on the right side of the dashed line. In both plots, the February-2018 measurements for some of the same samples (also used on September-2017 (Fig. 5 a)) and November-2017 (Fig. 5 b))

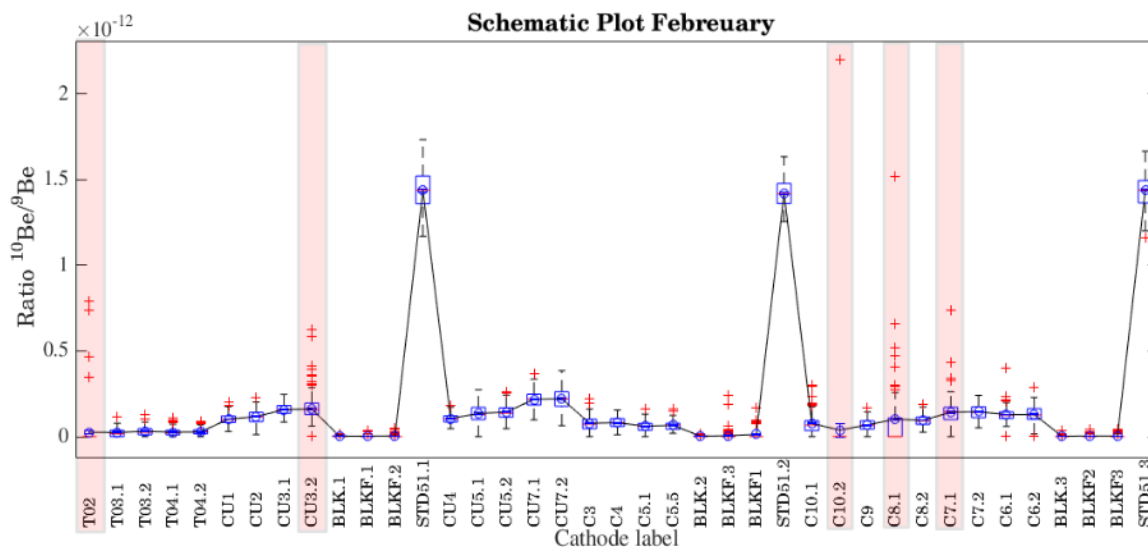


Figure 3. Schematic plot for the best campaign of measurements (February-2018), showing $^{10}\text{Be}/^9\text{Be}$ ratios as a function of the natural samples, standards and blanks. The blue boxes enclose the median (red horizontal line) and the the mean (blue circles) of each statistical distribution. Dashed bars are whiskers representing maximum an minimum values of each series, whereas solid bars are the statistical errors. Red crosses are the outliers values, i.e. values larger than 1.5 times the IQR value (see the text for details). The samples shaded in purple, represent the results with more outliers during the measurement.

campaigns) are shown on the left side of the dashed line.

For the calculation of these results, the normalization was made with respect to the nominal value of STD51. Although the data are from the same sample in both plots of Fig. 5 (T03 and T04), the ratios showed some differences (September, November or February). This difference can be related to the amount of ^9Be carrier added in the radiochemical procedure, then the samples are more or less diluted. Either way, the behavior of the different data series compared in Fig. 5 showed similar conditions observing in all of them a trend comparable taking in to account the uncertainties calculated in every measure. Considering the noise found on the September campaign spectra, such data can be discarded. Then, the best data were those from November and February campaigns, so the parameters related to the sample protocol preparation and the isotope separator tuning were fixed with these last two campaigns. With minor changes in the tuning of the machine from November to February measurements the data were improved, showing a better agreement between both statistical methods used for the analysis (see Fig. 5). This results are saying that February measurements were those when for most of the samples, the spectrometer was more stable on the time of measure.

Anyhow, in order to make a real comparison of the measurements, it is necessary to calculate the final concentrations of ^{10}Be radioisotope. This has to be made by using the values of standard and blanks (typically around the numbers reported in Fig. 2) and many other parameters related to the chemical preparation for the normalization. On the Table 1, the preliminary concentrations obtained for the sample T04 in November-2017 and February-2018 campaigns are shown. As it can be seen on the Table, this values show a good agreement for both campaigns. Nevertheless, many other tests will be make with the different data campaigns, in order to support the whole technique here presented.

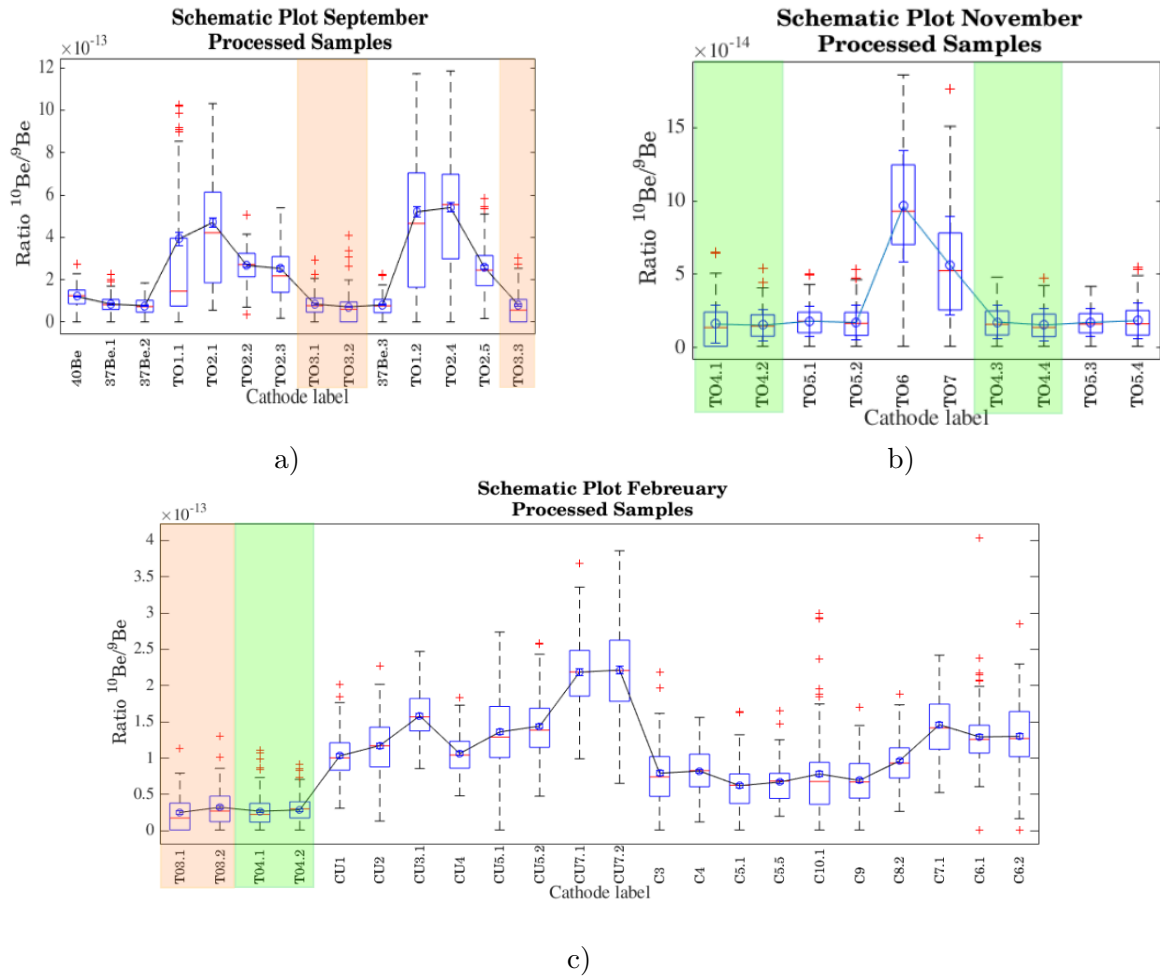


Figure 4. The same kind of plot than in Fig. 3, but for different campaigns of measurements: a) September-2017 b) November-2017 and c) February-2018. In this plots, the standards and the blanks ratios measured, as well as those samples with many outliers, were removed. The samples shaded in pink and green on the three plots, denote those samples that can be used to make comparisons: pink, September-2017 with February-2018 and green, November-2017 with February-2018.

Sample	C_{mean}	Error C_{mean}	C_{median}	Error C_{median}
T04NO17	2.4452e+08	2.5749e+07	2.6516e+08	6.3303e+07
T04FE18	2.7126e+08	1.1995e+07	2.9467e+08	1.8501e+07

Table 1. Preliminary concentrations and their corresponding errors for the sample T04 on November-2017 and February-2018 campaigns. The values obtained with both statistical methods (EDA and conventional) are reported (C_{median} and C_{mean} , respectively). The units for the concentrations measured are atoms/g.

5. Conclusions

In this work a protocol for sample preparation and its further analysis by using AMS technique were presented. Four measurements were carried out in order to establish the effectiveness of the protocol and the best parameters for the tuning of the LEMA isotope separator.

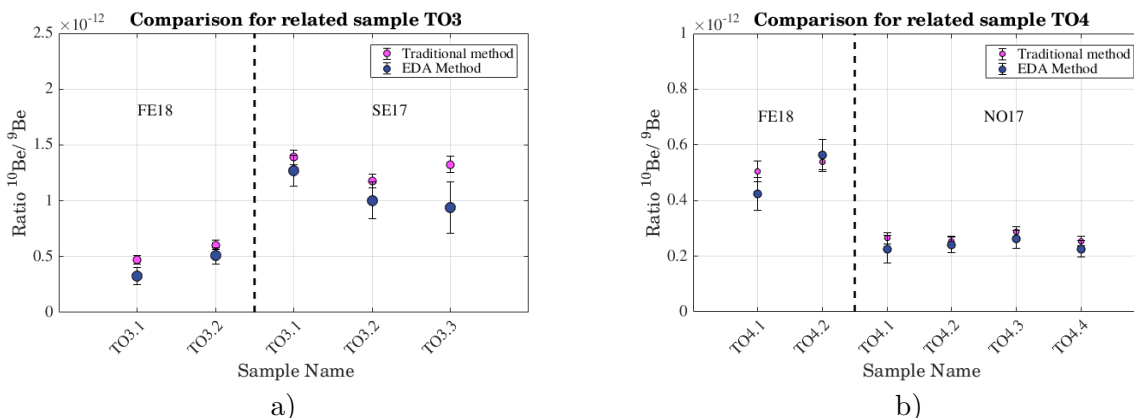


Figure 5. Comparison of some of the ratios measured in three different campaigns: In the left side of the dashed line in a) and b) are the $^{10}\text{Be}/^{9}\text{Be}$ ratios for samples from February-2017 campaign. In the right side of the dashed line of both plots are shown some ratios coming from same samples as in February-2017 campaign but for September-2017 campaign in a) and, November-2017 campaign in b). Pink circles are related with a conventional statistical analysis and blue circles with the EDA analysis method. Both data series present their statistical error bars.

In the characterization process carried out, the radiochemical extraction technique is critical to obtain reliable data. The ΔE -E spectra is one of the ways to verify that the extraction procedure is successful. In the measuring process the ratios values at the beginning or at the end of the run may change, considering this, the EDA method allows to determine measures of central tendency and the spread, being unaffected by these variations (if they occur). The method as well is useful to observe the stability of the data as a function of the time in this kind of measurements. Finally, stable and good data from the campaign of February-2018 show the expected behavior, demonstrating that the LEMA isotope separator is ready for the application of AMS technique to the study of ^{10}Be concentrations in atmospheric filters with PM10 particles. The agreement found between preliminary concentrations coming from two different campaigns of measurements, demonstrates the good stability of the whole process.

6. Acknowledgments.

This work was partially supported by CONACYT 51600, 82692, 123655 Projects and DGAPA-PAPIIT IA101616, IA103218 Projects.

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