Poster Presentation

COMPARISON OF ^{210}Pb AND ^{210}Po ACTIVITY CONCENTRATIONS FOR SEDIMENT DATING

H.C. ALMEIDA*, T.B.S. NASCIMENTO*, M.H.T. TADDEI*, B.P. MAZILLI**

* Poços de Caldas Laboratory,
National Commission on Nuclear Energy,
Poços de Caldas
Email: heleinec@yahoo.com.br

** Institute of Energy and Nuclear Research,
São Paulo

Brazil

Abstract

Records stored in natural archives, such as those for lake sediments, are used in environmental programmes for the assessment of changing erosion rates in a catchment arising from disturbances, and to monitor pollution by heavy metals and other contaminants. Accurate sediment chronologies are important to interpret those practices. One of the most important methods for dating recent sediments is through ^{210}Pb. This radionuclide occurs naturally as one of the ^{238}U decay series. It is widely distributed on Earth owing to its decay from radium in the ground or from radon that emanates to the atmosphere. The ^{210}Pb is deposited as particulates and falls into lakes where it is scavenged from the water column and deposited in the basins. The elevated ^{210}Pb concentrations are measured using the gamma spectrometry technique. However, this technique has a low sensitivity and small values are difficult to detect or require a long measurement time. The determination of ^{210}Po (a decay product of ^{210}Pb) using alpha spectrometry is more sensitive and rapid. The aim of this paper is to compare the activity concentrations of the two radionuclides in lake sediment samples to evaluate the use of ^{210}Po in the dating of sediments with low levels of ^{210}Pb.

1. INTRODUCTION

The increasing use and extraction of soil has been disturbing the natural environment and adding contaminants to water sources. The sediments deposited in lakes usually reflect the erosion processes in upstream catchments [1]. These sediments, previously considered only as nutrient sites, are now sources of contaminant information about water sources located near mining and ore processing industries and agricultural activities. Lake sediments stored in natural archives are used as records in a wide range of environmental programmes for purposes such as the assessment of changing erosion rates in a catchment (arising from disturbances such as afforestation, deforestation or changing agricultural practice) or the monitoring of pollution by heavy metals, organic pollutants and other contaminants [2]. Sediment quality is an important parameter in the assessment, protection and management of aquatic ecosystems. Because sediments influence the fate of many chemicals, concern exists about the potential impact on organisms that are exposed to sediments with elevated chemical concentrations [3].

One of the most important means for dating recent sediments (0–150 years) is through ^{210}Pb, which occurs naturally as one of the radionuclides in the ^{238}U series. Disequilibrium between ^{210}Pb and its parent isotope in the series, ^{226}Ra, arises through diffusion of the intermediate gaseous isotope ^{222}Rn. Some of the ^{222}Rn atoms produced by the decay of ^{226}Ra in soils escape into the atmosphere where they decay through a series of short-lived radionuclides to ^{210}Pb. This is removed from the atmosphere by precipitation or dry deposition, falling onto

the land surface or into lakes and oceans. The ^{210}Pb falling directly into lakes is scavenged from the water column and deposited on the bed of the lake with the sediments [2]. The excess ^{210}Pb in the sediments, beyond the amount that is in equilibrium with the in situ ^{226}Ra, decays in accordance with Eq. (1).

$$C_{Pb} = C_{Pb}(0)\, e^{-\lambda t} + C_{Ra}\left(1 - e^{-\lambda t}\right) \quad (1)$$

where C_{Pb} is the ^{210}Pb activity concentration in the sediment layer, $C_{Pb}(0)$ is the initial ^{210}Pb activity concentration in the sediment layer at its time of formation, C_{Ra} is the ^{226}Ra activity concentration, λ is the ^{210}Pb decay constant and t is the time since sediment formation. This equation can be used to date a sediment provided a reliable estimate can be made of the initial ^{210}Pb activity $C_{Pb}(0)$. Modelling and quantifying the process by which excess ^{210}Pb is produced and redeposited on the Earth's surface is an important prerequisite for the development of reliable methods for calculating ^{210}Pb dates [2]. In most cases the choice of method for determining ^{210}Pb activity concentration will be governed by what is available:

(1) Alpha spectrometry is more sensitive and most suitable for small samples of very low activity. The method is based on the measurement of ^{210}Pb via alpha radiation emitted by ^{210}Po, a decay product of ^{210}Pb. The ^{210}Pb is extracted from the sample by chemical digestion and deposited onto silver planchets for assay in a low background alpha spectrometer [2]. The detectors are simple and inexpensive, but it is necessary to have access to radiochemical facilities. A significant disadvantage is the time required to establish equilibrium between ^{210}Pb and ^{210}Po, or to allow the ingrowth of ^{210}Po. Furthermore, the method only determines total ^{210}Pb and ^{226}Ra concentrations [4].

(2) The gamma spectrometry method has a great advantage in that it does not require the leaching and radiochemical separation of ^{210}Pb or ^{210}Po [5]. A gamma assay requires minimal sample preparation, since gamma photons can travel significant distances without absorption. Furthermore, the measurements can be carried out on dried sediment samples without the need for radiochemical separation. The measurements are non-destructive so that, after gamma assay, samples can be used for further analyses. In addition, the method allows simultaneous determination of a range of radionuclides, including ^{210}Pb, ^{226}Ra, ^{137}Cs and ^{241}Am. On the other hand, the method presents higher overheads and lower sensitivity for some radionuclides, and the efficiency calibration is particularly demanding at low energies [2].

2. METHODS

Four sediment samples (Bortolan 12, Bortolan 15, Bortolan 22 and Fazenda 03) were selected for measurement of ^{210}Pb and ^{210}Po activity concentrations. To ensure the quality and accuracy of the methods used, samples of certified reference material IAEA–447 (moss soil) were prepared as described in the IAEA reference sheet and analysed, one sample for lead and two for polonium. The analysis methods used for the sediment samples and the reference material were as follows:

(1) For ^{210}Pb determination, the sediment samples and reference material were dried at 110 ± 5°C for 24 h, and then removed and cooled in a desiccator for 1 h. A known mass of sample was homogenized and compressed into a 49×13 mm propylene container until full, and hermetically sealed with chloroform. Thirty days were allowed for ^{222}Rn ingrowth inside the container and for achieving equilibrium between ^{210}Pb and ^{226}Ra. The sample was then counted in a Canberra gamma spectrometry system with a high purity germanium detector of 45% relative efficiency until at least 1000 counts were reached. In order to create the efficiency curve, the efficiency was thoroughly set and then the samples and the reference material were evaluated.

(2) For ^{210}Po determination, the sediment samples were prepared using about 0.2 g of dried sediment and 1 mL of ^{209}Po tracer. The reference material was prepared using about 1 g of dried sediment and 10 mL of ^{209}Po tracer. The samples were transferred to a Teflon beaker and kept under agitation for 20 min for homogenization. Each sample was totally digested in an open system with mineral acids, and then the residue was dissolved using 1.5M HCl and filtered. Finally the solution was transferred to a deposition cell using 1.5M HCl and 1 g of ascorbic acid was added to eliminate interference. The polonium was deposited onto a silver planchet under agitation for 4 h at 80°C. Then, the planchet was counted on a surface barrier detector in an alpha spectrometry system.

3. RESULTS

For ^{210}Pb, the activity concentration in the reference sample was found to be 420 ± 30 Bq/kg, the same as the value specified in the reference sheet (420 ± 20) Bq/kg. The details are shown in Fig. 1. For ^{210}Po, the activity concentrations in the two reference samples were found to be 345 ± 15 and 330 ± 15 Bq/kg. The activity concentration specified in the reference sheet, after applying a decay correction for the current date, was 344 ± 8 Bq/kg. The energy peaks for the first of the reference samples are shown in Fig. 2.

Both measurement techniques were then applied to the four sediment samples. The results are shown in Table 1.

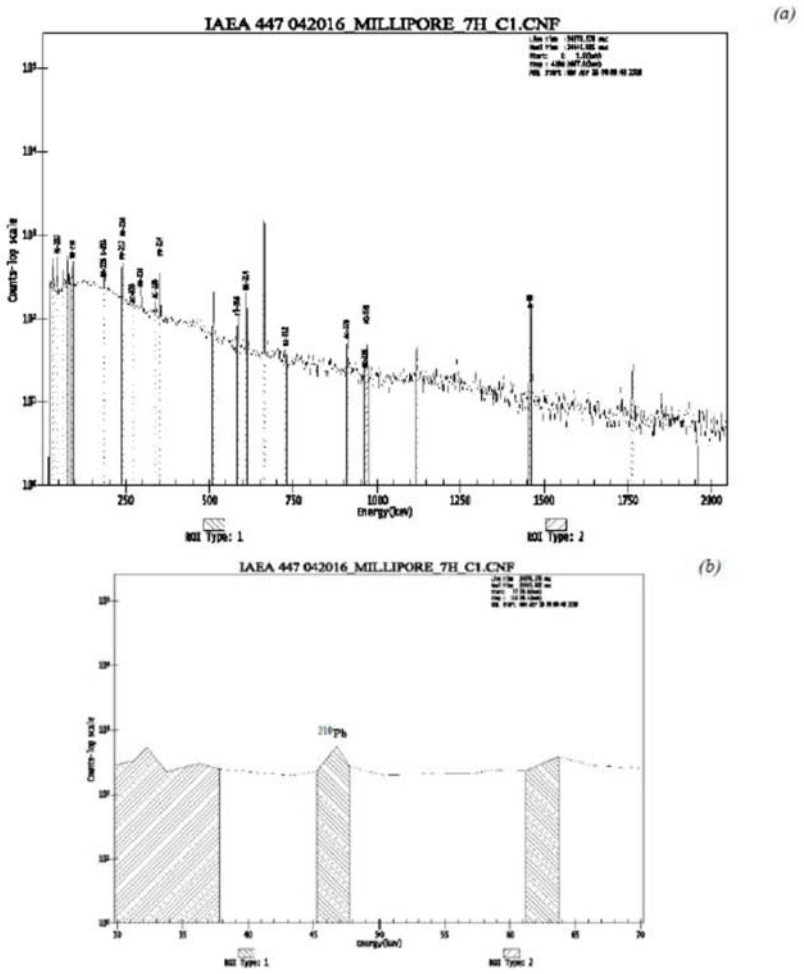

FIG. 1. IAEA–447 reference sample: (a) Elements identified by gamma spectrometry; (b) Detail of lead spectrum.

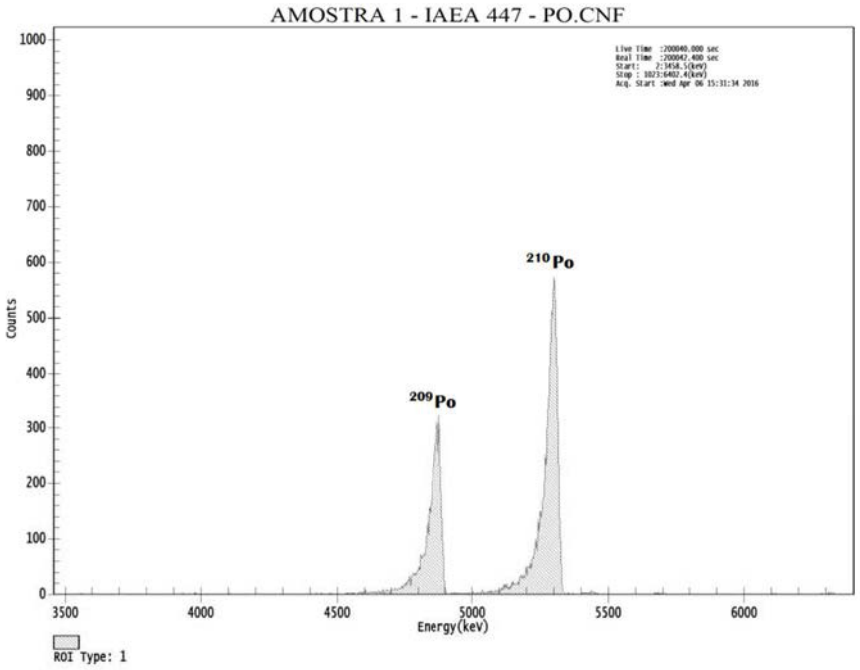

FIG. 2. IAEA–447 reference sample, alpha energy peaks for ^{210}Po and ^{209}Po.

TABLE 1. ACTIVITY CONCENTRATIONS MEASURED IN SEDIMENT SAMPLES.

	Activity concentration (Bq/kg)	
	Pb-210	Po-210
Sample 1	294.1 ± 20.9	258.9 ± 17.9
Sample 2	279.6 ± 19.4	246.0 ±17.2
Sample 3	289.8 ± 11.8	257.5 ± 18.3
Sample 4	301.1 ± 19.2	299.2 ± 16.2

4. DISCUSSION

The results obtained through gamma spectrometry for ^{210}Pb are slightly higher than those obtained through alpha spectrometry for ^{210}Po. This difference could be explained by losses in the radiochemical analysis or the amount of tracer added to the sample, that could have an activity concentration closer to the one found. The results were considered to be satisfactory for both methods. When the ^{210}Pb levels in the collected samples are high and there is a suitable amount of material available, it would be appropriate to choose the gamma spectrometry technique for performing further analyses. While the gamma spectrometry technique requires a longer time, to allow for the ingrowth of ^{222}Rn inside the flask, it involves less sample manipulation.

5. CONCLUSION

The values obtained using the gamma spectrometry method for ^{210}Pb analysis and using the radiochemical method for ^{210}Po analysis were considered satisfactory, showing that either method can be used. These results corroborate the intrinsic relationship of radioactive equilibrium between ^{210}Pb and its progeny ^{210}Po, proving the existence of a chemical balance as proposed in Ref. [5]. Since there is a substantial amount of ^{210}Pb in the analysed sediment samples, the gamma spectrometry technique will be used for further determinations of ^{210}Pb activity concentration and subsequent sediment dating.

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