

A comparison between ^{210}Pb and ^{210}Po activity concentration on Sediment Dating

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Abstract

Records stored in natural archives, such as lake sediments, are used in environmental programs 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 means to date recent sediments is through ^{210}Pb . This radionuclide occurs naturally as one of the ^{238}U decay series. It is widely distributed on the earth due to its formation from radium in the ground or from radon that emanates to the atmosphere or decays to ^{210}Pb . The ^{210}Pb is deposited as particulates and falls into lakes where it is scavenged from the water column and deposited on the basins. Excess of ^{210}Pb is measured through gamma spectrometry technique. However, this technique has a low sensibility and small values are difficult to detect or need to be analyzed for a long time. ^{210}Po is a decay product of ^{210}Pb and its determination through alpha spectrometry is more sensitive and faster. The aim of this paper is to compare both radionuclides activity concentration in lakes sediments cores to evaluate the use of ^{210}Po in dating of locals with low levels of ^{210}Pb .

Introduction

The increasing soil use and extraction have been generating perturbation and addition of contaminant materials to water sources. The sediments that are deposited in lakes usually reflect the erosion processes from upstream watersheds [1]. These records, that were considered just nutrient sites, are now supplies of contaminant information about water sources located next to mining-extracting industries and agricultural activities.

Records stored in natural archives, such as lake sediments are used in a wide range of environmental programs, for example, in the assessment of changing erosion rates in a catchment arising from disturbances such as afforestation, deforestation, changing agricultural practice or to monitoring pollution by heavy metals, organic pollutants and other contaminants [2]. Sediment quality are an important focus 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 concentration [3].

One of 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 . A fraction 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. ^{210}Pb falling directly into lakes is scavenged from the water column and deposited on the bed of the lake with the sediments [2].

Excess ^{210}Pb in the sediments over that in equilibrium with the *in situ* ^{226}Ra in sediments decays in accordance with the radioactive decay law,

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

(where λ is the ^{210}Pb radioactive decay constant). This equation can be used to date the sediments provided reliable estimates can be made of the *initial* ^{210}Pb activity $C_{Pb}(0)$ in each sediment layer *at the time of its formation*. Modelling and quantifying the process by which excess ^{210}Pb is produced and redeposited on the earth's surface is an important prerequisite to the development of reliable methods for calculating ^{210}Pb dates [2].

In most cases the choice of the method will be governed by what is available. Alpha spectrometry is more sensitive and most suitable for small samples of very low activity. The detectors are simpler and less expensive, though it is necessary to have access to radiochemical facilities. A significant disadvantage is the time required to establish $^{210}\text{Pb}/^{210}\text{Po}$ equilibrium, or to allow the ^{210}Po ingrowth. Further, the method only determines total ^{210}Pb and ^{226}Ra concentrations [4]. This method measures ^{210}Pb via alpha radiation emitted by ^{210}Po , the granddaughter product of ^{210}Pb decay. ^{210}Pb is extracted from the sample by chemical digestion and deposited onto silver planchets for assay in a low-background alpha spectrometer. [2]

Gamma spectrometry methods has a great advantage, it does not require the leaching and radiochemical separation of ^{210}Pb or ^{210}Po [5]. An advantage of gamma assay is 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 of 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, lower sensitivity for some radionuclides and the efficiency calibration is particularly demanding at low energies [2].

Methods

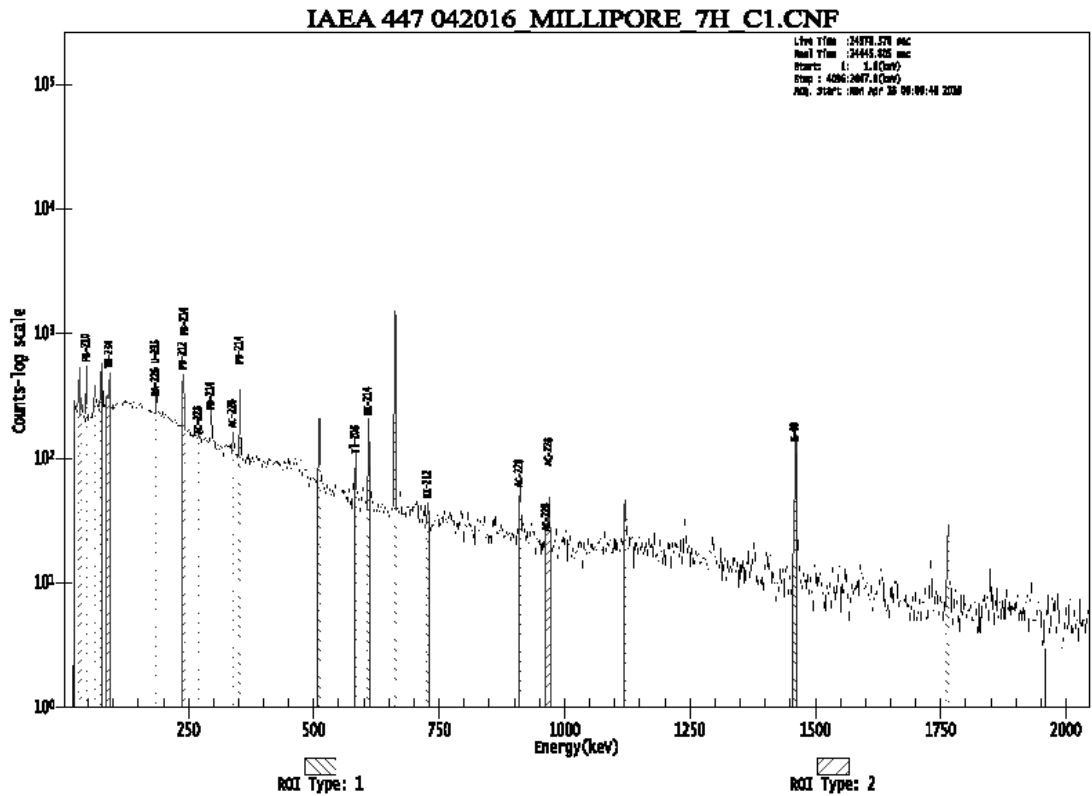
^{210}Pb : The certified reference material IAEA - 447 was prepared as the Reference Sheet of International Atomic Energy Agency describes. The material and the analyzed samples were dried at $110^\circ\text{C} \pm 5^\circ$ for 24h, then they were removed and cooled in a desiccator for 1h. A known mass of sediment was homogenized and compressed in a 49x13mm geometry propylene container until fullness, hermetically sealed with chloroform. Thirty days were expected to ^{222}Rn ingrowth inside the container and to the ^{210}Pb balance with ^{226}Ra , and then the sample was counted in a Gamma Spectrometry System, Canberra Detector, with a Germanium detector of high purity with 45% of relative efficiency, until it reaches at least 1000 counts in area. In order to create the efficiency curve, the efficiency was thoroughly set and then the samples and the reference material were evaluated.

^{210}Po : The reference material was prepared using about 1g of the dried sediment and 10 ml of ^{209}Po tracer. The samples were prepared using about 0,2g of dried sediment and 1ml of the tracer. They were transferred to a Teflon beaker and kept under agitation for 20 minutes for homogenization. Thus, the sample was totally destroyed 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 1g of ascorbic acid was added to eliminate interferences. The Po was deposited onto a silver planchet under agitation during 4h at 80°C . Then, the planchet was counted on a surface barrier detector in an Alpha Spectrometry System.

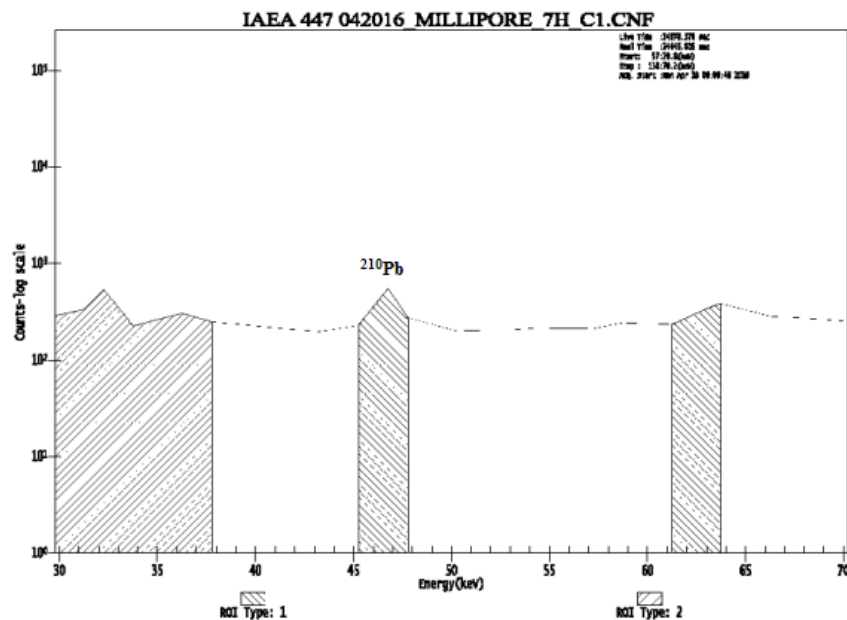
Results

First, in order to ensure the quality and accuracy of the method applied, samples of certified reference material of moss soil (IAEA – 447) were analyzed, two for Polonium and one for Lead.

For behavior comparison of the efficiency curve for gamma analysis, the values of activity found into the reference material for the ^{210}Pb is $(420 \pm 30) \text{ Bq}\cdot\text{kg}^{-1}$ in agreement with the value of the certified reference sheet $(420 \pm 20) \text{ Bq}\cdot\text{kg}^{-1}$.



(a)



(b)

Figure I: (a) Elements identified in IAEA – 447 Sample; (b) Detail of Lead Spectrum in IAEA – 447 Sample.

For ^{210}Po , considering the activity value available on the Reference Sheet and calculating the decay correction for the current date, it was expected to find an activity of $(344 \pm 8) \text{ Bq}\cdot\text{kg}^{-1}$ in the samples. The results show an activity of $(345 \pm 15) \text{ Bq}\cdot\text{kg}^{-1}$ and $(330 \pm 15) \text{ Bq}\cdot\text{kg}^{-1}$ for samples 1 and 2, respectively. Figure II shows the energy peaks for ^{210}Po and its tracer, ^{209}Po , with α spectrometry for Sample 1.

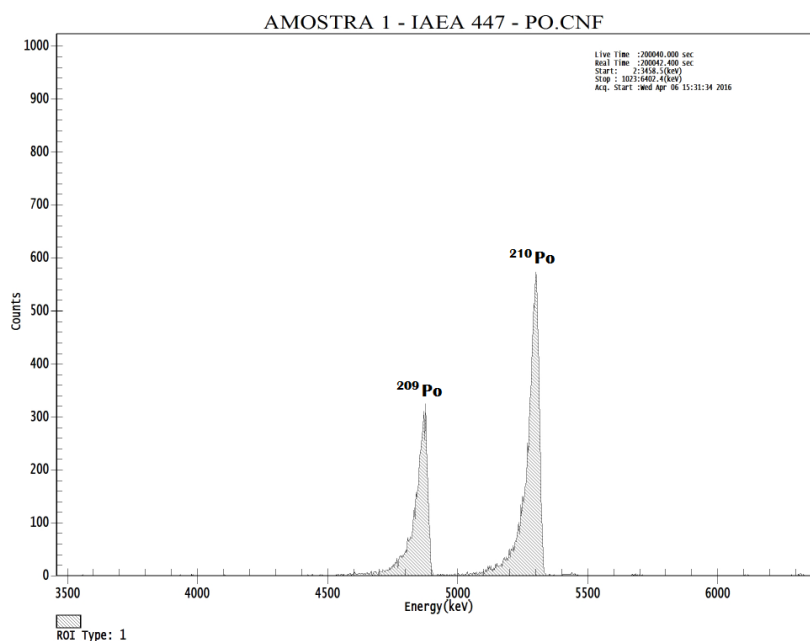


Figure II: Energy peaks for ^{210}Po and ^{209}Po

Both techniques were then applied for samples Bortolan 12, Bortolan 15, Bortolan 22 and Fazenda 03 with the following results for Lead respectively: $(294,1 \pm 20,9) \text{ Bq}\cdot\text{kg}^{-1}$, $(279,6 \pm 19,4) \text{ Bq}\cdot\text{kg}^{-1}$, $(289,8 \pm 11,8) \text{ Bq}\cdot\text{kg}^{-1}$, $(301,1 \pm 19,2) \text{ Bq}\cdot\text{kg}^{-1}$. The results for Polonium were respectively $(258,9 \pm 17,9) \text{ Bq}\cdot\text{kg}^{-1}$, $(246,0 \pm 17,2) \text{ Bq}\cdot\text{kg}^{-1}$, $(257,5 \pm 18,3) \text{ Bq}\cdot\text{kg}^{-1}$, $(299,2 \pm 16,2) \text{ Bq}\cdot\text{kg}^{-1}$.

Discussion

The results obtained through gamma spectrometry for Pb have shown a slightly variation in comparison with the results through alpha spectrometry for Po. This difference can occur either because of losses on the radiochemical analysis or the amount of tracer added to the sample, that could have an activity closer to the one found. Indeed, the results proved to be satisfactory for both methods. Once the Pb levels on the collected samples are high, and there is a suitable amount of material available, it is expected to choose the gamma spectrometry technique to perform further analysis. Even though the gamma spectrometry requires a longer time to the growing of ^{222}Rn inside the flask, it demands less sample manipulation.

Conclusion

The values obtained using the gamma method for analysis of Lead and the radiochemical analyses of Polonium were considered satisfactory, showing that both methods can be used. These results corroborate the intrinsic relationship of radioactive equilibrium between ^{210}Pb and its granddaughter ^{210}Po , proving the existence of chemical balance as proposed by Cutshall [5]. In conclusion, since there is a substantial amount of lead in the analyzed samples, gamma spectrometry technique will be used in further determination of ^{210}Pb activity concentration and subsequent sediment dating.

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