A comparison between lead-210 determination in sediments by gamma and alpha spectrometry in sediments

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Abstract

Polonium-210 (Po-210) is analyzed for a variety of purposes, including for radiological impact assessment, as a tracer of environmental processes, and as an indirect measure of its progenitor lead-210 (Pb-210). A method for determining Po-210 by alpha spectrometry in sediments has been set. Chemical preparation was performed by acid digestion in open systems. Mixtures of HCl, HF, HNO_3 and $HClO_4$ in different proportions were employed at 80 °C in order to avoid losses due to volatilization. Results were compared with those for Pb-210 using high resolution gamma spectrometry. Sediment core samples were analyzed. A linear regression was performed showing a strong relationship among Pb-210 and Po-210 activities, proving that the method used for Po-210 determination is also suitable for Pb-210 in sediments. Mass accumulation rate (MAR) and sediment accumulation rate (SAR) were estimated using the constant flux: constant sedimentation (CF:CS) model, showing changes on them both along the sediment core studied.

Key words: *lead 210; polonium 210; sediments, alpha spectroscopy; gamma spectroscopy; ecological concentration.*

Comparación entre la determinación de plomo-210 en sedimentos mediante espectrometría gamma y alfa en sedimentos

Resumen

El polonio-210 (Po-210) se analiza con varios fines, incluyendo la evaluación de un impacto radiológico, como trazador de procesos ambientales, y como medición indirecta de su progenitor plomo-210 (Pb-210). Se estableció un método para la determinación de Po-210 por espectrometría alfa en sedimentos. La preparación química se realizó con una digestión ácida en plancha. Se empleó una mezcla de HCl, HF, HNO_3 y $HClO_4$ en diferentes proporciones a 80 °C para evitar pérdidas debido a volatilización. Los resultados se compararon con los obtenidos para Pb-210 por espectrometría gamma de alta resolución. Se analizaron muestras de un perfil de sedimentos. Se realizó una regresión lineal que mostró una relación fuerte entre las actividades de Pb-210 y Po-210, demostrando que el método utilizado para la determinación de Po-210 es también adecuado para la determinación de Pb-210 en sedimentos. Se estimaron las tasas de sedimentación y de acumulación másica usando el modelo de flujo constante: sedimentación constante, mostrando cambios en estas a lo largo del perfil estudiado.

Palabras clave: *plomo 210; polonio 210; sedimentos; espectroscopia alfa; espectroscopia gamma; concentración ecológica.*

Introduction

Po-210 is analyzed for a variety of purposes, including as a tracer of environmental processes, and as an indirect measure of its progenitor Pb-210. They have also been applied in studies of the chronology of sediments cores. Several analytical techniques have been reported for Po-210 chemical preparation before alpha-spectrometry measurements [1]. Sample preparation is thus time- and effort-consuming for Po-210 determination. Pb-210 can be measured directly from its 46,5 keV gamma-particle; this method enjoys the advanta-

ges of easy sample-preparation procedures, been the low energy of gamma-rays from Pb-210 and the low branching ratio (4%) their principal drawbacks [2]. The aim of the study is to determine the relative precision and comparability of both methods and apply them in a sediment core dating from an estuarine system.

Materials and methods

Sediments samples were collected by core in an estuarine system. The profile was sectioned into 1 cm slices, for a total of 71 samples. Samples were dried at 45 °C, grounded to fine powder (grain size less than 0,5 mm), and then homogenized thoroughly.

1.1. Alpha spectrometry

Po-209 was added (0,038 Bq) as a tracer to the dried sediment to determine the chemical recovery and plating efficiency. The dried sediment (~1 g) was then digested by wet ashing in open systems, with concentrated nitric and hydrofluoric acids (10 and 6 ml respectively), until almost dryness (samples were never dried in order to avoid volatilization) in a hot plate at 80 °C. Traces of hydrofluoric acid were removed by concentrated perchloric acid (4 ml) boiling. Residue was boiled in 6 ml of concentrated hydrochloric acid and finally the solution was turned into 1 M hydrochloric acid by several evaporations. Ascorbic acid was added to reduce Fe^{3}+ to Fe^{2}+, thus eliminating its interference. Before plating, any remaining undissolved sediment was removed by centrifuging the sample during 12 minutes at 3000 rpm and filtering. The polonium plated during the night, onto one side of the silver disk at room temperature. The plates were counted until each peak (Po-209 and Po-210) has 400 counts or more on a Canberra alpha-spectrometer with PIPS detectors. Detection limits were below 0,2 mBq/kg for measurements of 86400s.

1.2. Gamma spectrometry

Samples were analyzed in a well detector. An HPGe low background gamma-ray spectrometer was used. The coaxial detector with an iron shield 21 cm thick has an efficiency of 50,71% for 46,54 keV. The system is connected to a LTEL PC multichannel analyzer, with 8192 channels and the resolution of the 1,33 MeV gamma-ray peak from Co-60 is 1,86 keV. In this method, an average of 5,8692 g of sediment was placed in a vial of 4,5 ml, inside the well. Results were analyzed using the Winner 6.0 software.

1.3. Sediment dating

MAR and SAR were estimated using the CF:CS model. Supported Pb-210 was estimated using the constant values of total Pb-210 in the deeper samples of the core. Pb-210 values used were those obtained by alpha spectrometry.

Results

2.1. Alpha and gamma spectrometry comparison

Sample preparation for Po-210 determination was time consuming but low detection limits could be reached.

Specific activities above 6,5 Bq/kg of Pb-210 could be reported with an uncertainty less than 43 % for low activities for measurements of 172800 seconds.

As a result of fitting a linear model a Correlation Coefficient equals to 0,949391 was obtained.

The equation of the fitted model is:

Pb = -5,0106 + 1,13099*Po (1)

where:

Pb: Pb-210 Specific Activity expressed in (Bq/kg)

Po: Po-210 Specific Activity expressed in (Bq/kg)

Since the P-value 0,0000 is less than 0,01, there is a statistically significant relationship between Pb-210 specific activity and Po-210 specific activity at the 99% confidence level. The R-Squared statistic indicates that the model as fitted explains 90,1344 % of the variability in Pb-210 specific activity. The correlation coefficient equals 0,949391, indicating a relatively strong relationship between the variables.

The relationship between the data sets is shown graphically in figure 1.

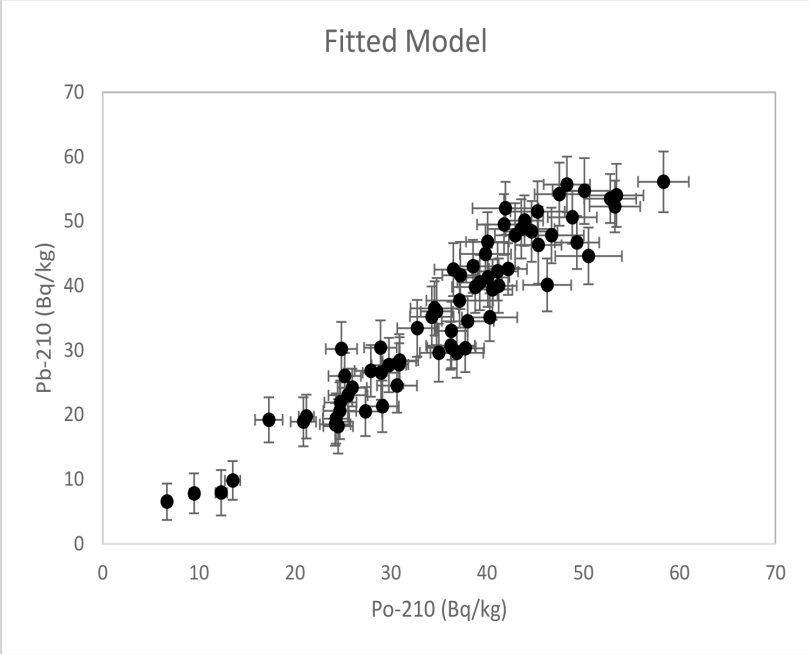

Figure 1. Plot of Po-210 and Pb-210 specific activities of sediment samples measured by gamma and alpha spectrometry.

2.2. SAR and MAR estimation

Two sets of SAR and MAR were found in the core figure 2 and 3, i.e. 0,298(21) cm yr- for the older years and 1,77(31) cm yr- for the recent ones, or 3,06(30) kg m^{-2} yr for the deeper layers and 13,3(2,5) kg m^{-2} yr for the new ones.

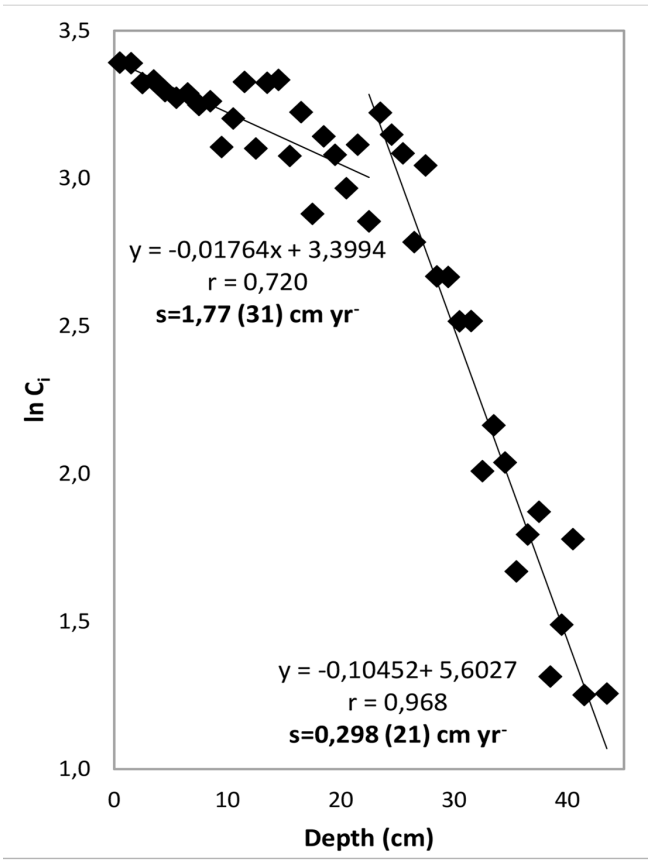

Figure 2. Sedimentation rates estimated by CF:CS model.

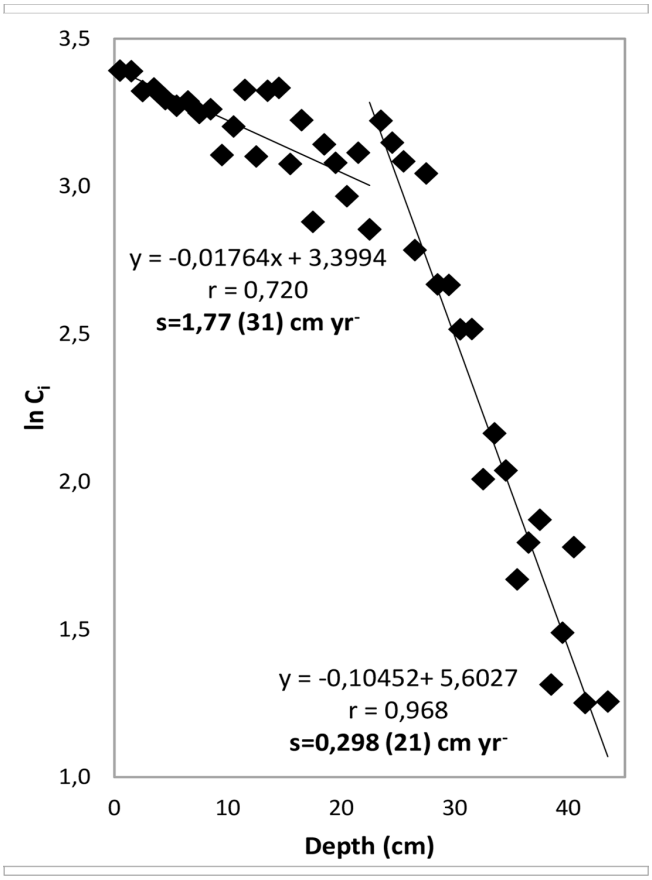

Figure 3. Mass accumulation rates estimated by CF:CS model.

Discussion

Alpha and gamma methods are both suitable for Pb-210 quantification, although alpha spectrometry yields lower relative uncertainties than gamma spectrometry, even with lower counting times. There is no statistically significant difference between the methods.

There has been a change in the amount of sediment entrance and the rate at which they are accumulated in that site. Stakeholders should take that into consideration, in order to avoid a negative impact in the environment.

Conclusions

Measurements based on Pb-210 alpha-counting and gamma ray spectrometry can yield comparable values for Pb-210 specific activities. Therefore, they both could be used for estimating sediment accumulating rates and mass accumulating rates in sediment cores. Although lower uncertainties can be achieved with alpha spectrometry.

References

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Recibido: 18 de septiembre de 2024
Aceptado: 31 de octubre de 2024

Los autores declaran que no existen conflictos de intereses en la realización, ni la comunicación del presente trabajo.

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