AIP Conference Proceedings

RESEARCH ARTICLE | AUGUST 07 2008

Measurement of ^{210}Pb and its Application to Evaluate Contamination in an Area Affected by NORM Releases

F. Mosqueda; M. Villa; S. Hurtado; A. Absi; G. Manjón; F. Vaca; R. García-Tenorio

AIP Conf. Proc. 1034, 315–318 (2008)
https://doi.org/10.1063/1.2991236

Measurement of ^{210}Pb and its Application to Evaluate Contamination in an Area Affected by NORM Releases

F. Mosqueda(*)[1], M. Villa[2], S. Hurtado[2], A. Absi[3], G. Manjón[3], F. Vaca[1], R. García-Tenorio[3]

(*) Corresponding author. Tel: +34-95-9219798; fax: +34-95-9219777
E-mail address: fernando.mosqueda@dfa.uhu.es
[1]Dpto. de Física Aplicada. Facultad de Ciencias Experimentales. Universidad de Huelva. Campus de El Carmen. 21007-Huelva, Spain
[2] Centro de Investigación, Tecnología e Innovación. CITIUS. Universidad de Sevilla. Av. Reina Mercedes 4B. 41012-Sevilla, Spain
[3] E. T. S. Arquitectura. Dpto. de Física Aplicada II. Universidad de Sevilla. Av. Reina Mercedes, 2. 41012-Sevilla, Spain

Abstract. Liquid scintillation counting (LSC) is an easy and straightforward technique, and combined with its low limit of detection, makes it a powerful tool for both routine and low level measurements that can be applied to ^{210}Pb low level counting in environmental samples. ^{210}Pb can be easily measured following a sulphate co-precipitation method; the addition of a carrier and the weighing of the recovered amount is a widespread technique to evaluate radiochemical yield, however, this evaluation of the recovery is sometimes questioned. The samples employed in this work were recollected in 1999 and 2005 from the estuary of the Odiel and Tinto rivers (SW of Spain), which were affected by phosphogypsum (pg.) discharges until 1998. Phosphogypsum contains most of the ^{210}Pb from the treated raw material, for that reason analysed riverbed sediments have enhanced ^{210}Pb activity concentrations and hence, enhanced activity concentration of its daughter ^{210}Po, both in secular equilibrium after two years.

Keywords: ^{210}Pb, ^{210}Po, Liquid Scintillation Counting, Cerenkov radiation
PACS: 06.06.Ei, 06.06.Mr.

INTRODUCTION

To get the most accurate results in LSC, it is not only necessary to calibrate the counting efficiency, taking into account alpha interferences, but also to have a robust tool to evaluate the yield of the radiochemical procedure. Thus the objectives of this work can be divided in: 1.- Studying the accuracy of the methods to obtain the lead recuperation in the co-precipitation. Two methods are compared, ICP-MS for direct evaluation of stable lead, and weighting the final sulphate precipitate. 2.- Comparing the LSC with three techniques employed for the evaluation of ^{210}Pb: LSC, Cerenkov counting, gamma spectrometry and alpha spectrometry (^{210}Po), in order to evaluate the accuracy of the LSC technique. And 3.- Evaluating the evolution of ^{210}Pb concentration in sediments from Huelva Estuary (in years 1999 and 2005), affected by former discharges of pg.

CP1034, *The Natural Radiation Environment—8th International Symposium,* edited by A. S. Paschoa

SAMPLING AND METHODS

^{210}Pb was evaluated through four different radiometric techniques in sediment samples, collected in the years 1999 and 2005 in the Huelva estuary affected by former pg. discharges.

Through LSC and Cerenkov radiation, ^{210}Pb was determined following a selective sulphate precipitation method [1]. After co-precipitating Ra and Pb as Ra-Pb-$BaSO_4$ [2] by the (addition of dilute H_2SO_4). The Ra-Pb-$BaSO_4$ precipitate was re-dissolved using EDTA. Re-precipitation of $PbSO_4$ was obtained by adjusting the solution pH = 3. Precipitates were dissolved using EDTA in ammonia medium. For LSC, the sample volume was mixed with scintillator (Optiphase Hisafe 3) and for Cerenkov counting of ^{210}Pb MilliQ water was added to the vial to complete 20 ml. The detectors employed were a Quantulus Wallac 1220 and a Packard Tri-Carb 3170.

On the other hand ^{210}Pb was determined through gamma spectrometry, employing a Canberra n-type Reverse Electrode Germanium detector (REGe); and through alpha spectrometry, measuring ^{210}Po in secular equilibrium with ^{210}Po, through auto-deposition onto metal surfaces using 1.5 M HCl, employing a Camberra PIPS alpha detector [1], [3].

RESULTS AND DISCUSSION

Evaluation of the Recovery Yield

To calculate the recovery yield in the co-precipitation method, a widespread technique consists of the weighing of the final mass of the $PbSO_4$ precipitate, after adding a known amount of carrier. To obtain the radiochemical yield from the precipitate of $PbSO_4$, it has been made a calibration curve, stable lead mass versus radiochemical yield[1]. To check if it is possible to estimate the radiochemical yield by weighting the final precipitate, the ICP-MS technique is applied to measure the final stable lead recovered (Figure 1).

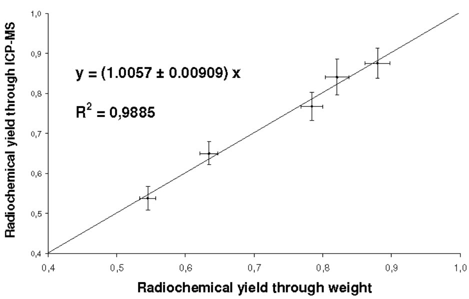

FIGURE 1. Radiochemical yield calculated through mass measurement vs. radiochemical yield evaluated through stable lead measurement through ICP-MS.

According to these results, the gravimetrical method provides accurate results for recovery calculations and therefore, could be used as an alternative to the ICP-MS technique when necessary.

Evaluation of ^{210}Pb

^{210}Pb was determined through LSC and by Cerenkov (CK) counting through beta emissions of its descendant ^{210}Bi. For CK counting, the radiochemical procedure is identical, except that there is no final mixing with scintillator cocktail [4][3]. Furthermore, in CK counting it is necessary waiting 28 days to reach the secular equilibrium ^{210}Pb–^{210}Bi.

The results, in Figure 2, show good agreement between both techniques. The main advantage of LSC is that the determination is immediate and is not necessary to wait for secular equilibrium. On the other hand, in CK counting, no scintillator is employed, so heterogeneity between sample and cocktail is avoided; this is of great advantage in the case of high amounts of sulphates when higher concentrations of EDTA and ammonia are needed for the dissolution of the sulphates [3].

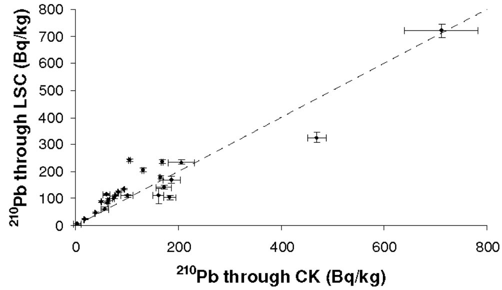

FIGURE 2. ^{210}Pb (Bq/kg) by LSC vs. ^{210}Pb (Bq/kg) by Cerenkov counting.

The evaluation of the goodness of the LSC procedure for ^{210}Pb determination is also done through intercomparing samples from the Estuary of Huelva, through two more alternative techniques (Figure 3): gamma spectrometry and alpha spectrometry (measuring its daughter ^{210}Po, after secular equilibrium is reached). The results show coincident results with both gamma and alpha techniques.

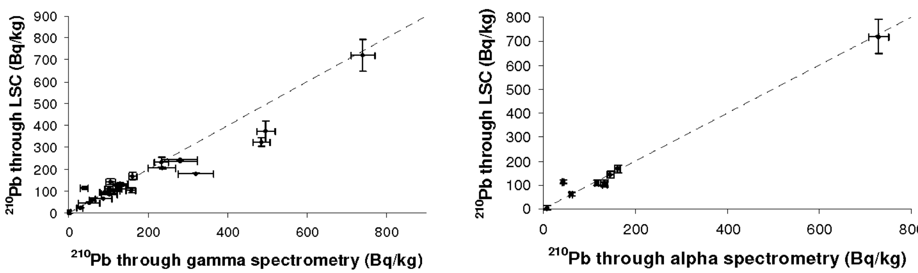

FIGURE 3. ^{210}Pb (Bq/kg) by LSC vs. ^{210}Pb (Bq/kg) by gamma spectrometry (a) and by alpha spec. (b).

Evolution of ^{210}Pb Concentration in Sediments of Huelva Estuary

In 1998, the new policy of waste management changed completely and releases to Odiel and Tinto river ceased [5]; activities in 1999 are higher than in 2005, since the changes caused a clear decrease in ^{210}Pb concentrations in the riverbed sediments.

It can also be seen a remobilization of the bottom sediments that homogenizes ^{210}Pb concentrations in the sampling points. The diminishing of the activity concentration in the Tinto River is not as noticeable as in the Odiel part due to the lixiviation of old phosphogypsum open-air piles next to the Tinto area.

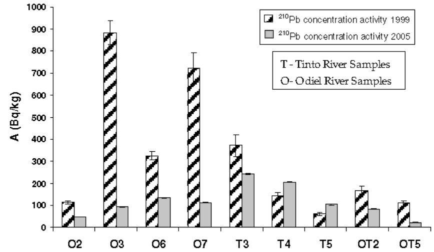

FIGURE 4. ^{210}Pb (Bq/kg) by LSC in sediment samples colleted in Tinto and Odiel rivers (years 1999 and 2005)

CONCLUSIONS

The determination of ^{210}Pb through LSC, applying the sulphate precipitation method, is suitable for samples affected by phosphogypsum discharges, being an accurate and precise method. Furthermore, it is proved that it is possible to evaluate the recovery yield of the procedure by weighting the final mass of the sulphate precipitate, comparing the results obtained through the measurement of stable lead by ICP-MS. This is supported for the excellent agreement within the uncertainties of the four different intercompared techniques, LSC, Cerenkov, gamma and alpha spectrometry.

REFERENCES

1. M. Villa, S. Hurtado, G. Manjón, R. García-Tenorio. *Calibration and measurement of ^{210}Pb using two independent techniques*. Article in press in: Radiation Measurements.
2. M. Villa, H.P. Moreno, G. Manjón. *Determination of ^{226}Ra and ^{224}Ra in sediments samples by liquid scintillation counting*. Radiation Measurements **39** (2005) 543-550
3. S. Hurtado, M. García-León, R. García-Tenorio. *^{210}Pb determination in lead shields for low-level small gamma, γ-spectrometry applying two independent radiometric techniques*. Nucl. Instrum. Meth. A **497** (2004) 381-388
4. F. Mosqueda, M. Villa, F. Vaca, J.P. Bolívar. *Colour quenching corrections on the measurement of ^{90}Sr through Cerenkov counting*. Sent for publication to Anal. Chim. Acta.
5. A. Absi, M. Villa, H. P. Moreno, G. Manjón and R. Periañez. *Self-cleaning in an estuarine area formerly affected by ^{226}Ra anthropogenic enhancements*. Science of The Total Environment, **329** (2004) 183-195.