^{129}I, ^{236}U, ^{239}Pu and ^{240}Pu profiles in a peatbog from the Southern Hemisphere

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Abstract

^{129}I, ^{236}U, ^{239}Pu and ^{240}Pu isotopes have been analyzed in the different layers of a peatbog core collected in Madagascar Island (South Hemisphere) and representing at least the last 100 years of atmospheric deposition. The determination of ultra-low levels of these isotopes has been possible by applying the Accelerator Mass Spectrometric Technique (AMS) by using the 1 MV Tandetrom AMS facility located at CNA (Seville, Spain). The elements of interest were extracted and isolated previous to the measurements by applying validated radiochemical procedures.

In this contribution, the behavior along the core of the different isotopes under analysis will be discussed, evaluating in particular the magnitude of their post-depositional retention/migration. In this sense, it will be highlighted the great mobility of the ^{129}I with a near uniform profile along the core, in opposition for example with the preservation for the Pu isotopes of the fallout bomb peak.

In the case of ^{236}U, the ^{236}U/^{239}Pu atomic ratios determined in a total of eight layers of the core are ranging in the interval 0.02 – 0.29 with an average value of 0.15. As far as we know, these are the first ^{236}U results seeing the light that were obtained in deposits as peatbogs collected in the southern hemisphere.

Experimental Set-up and Objectives

The main objective of the work was to obtain the profiles for several artificial radionuclides (^{239}Pu, ^{240}Pu, ^{129}I and ^{236}U) in a peatbog collected in the Southern Hemisphere (Madagascar) by Accelerator Mass Spectrometry (AMS). These determinations hardly can be done by radiometric techniques due to the low levels expected for the analyzed radionuclides.

The behavior of the mentioned radionuclides in the peatbog core has been analyzed, and information (scarce in the literature) about ^{240}Pu/^{239}Pu, ^{236}U/^{238}U and ^{236}U/^{239}Pu atom ratios in the environment of the southern hemisphere are given and briefly discussed.

The study have been done by using the SARA (Spanish Accelerator Radionuclide Analyses) system of 1 MV (LEAMS) located at the Spanish National Accelerator Centre in Seville, Spain. A schematic representation of the set-up is shown in Figure 1. Radiochemical and measurement procedures used at CNA with SARA for the radionuclides involved in this work, can be found in the following references: Santos et al. (2006), Chamizo et al. (2008), Gomez-Guzmán et al. (2012), Chamizo et al. (2015) and Scognamiglio et al. (20169. In addition, and in aliquots of the different layers of the profile, ^{238}U activity concentrations have been determined by alpha-particle spectrometry after applying a radiochemical method for isolation and conditioning for the measurement (Lehritani et al., 2012).

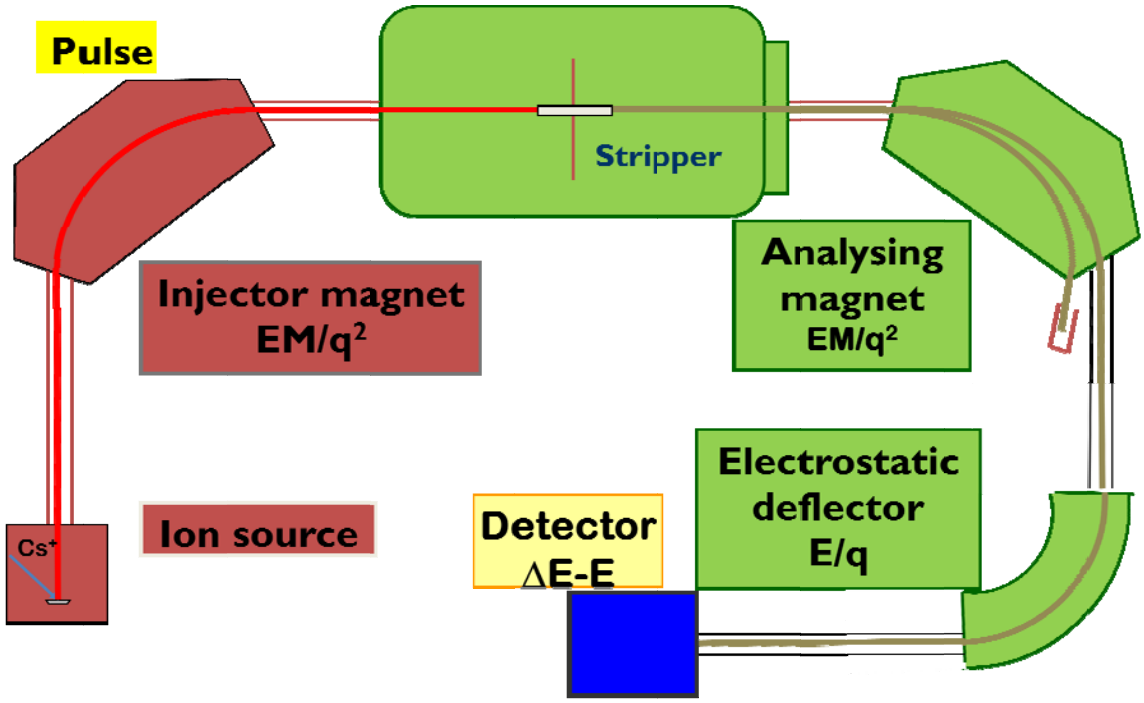

Figure 1.- Schematic representation of the SARA LEAMS system

Results

The ^{129}I, ^{239}Pu, ^{236}U and ^{238}U concentrations, expressed in atoms/g, in a total of eight layers from the analyzed peatbog are compiled in Table 1, while in Table 2 are compiled the $^{240}Pu/^{239}Pu$, $^{236}U/^{238}U$ and $^{236}U/^{239}Pu$ atom ratios.

The corresponding ^{129}I and ^{236}U profiles, in the Figure 1, and the $^{240}Pu/^{239}Pu$ and $^{236}U/^{239}Pu$ atom ratios, in the Figure 2, are also represented. All the set of results are briefly commented and discussed in the following section.

CODE	DEPTH (cm)	^{129}I (atoms/g)	^{239}Pu (atoms/g)	^{236}U (atoms/g)	^{238}U (atoms/g)
TOT – 2	2 -4 cm	$(11.47 \pm 0.81)10^6$	$(264.5 \pm 5.7)10^6$	$(59.9 \pm 3.4)10^6$	$(16.3 \pm 1.2)10^{14}$
TOT – 5	8 – 10 cm	$(10.36 \pm 2.42)10^6$	$(187.9 \pm 5.2)10^6$	$(35.0 \pm 1.9)10^6$	$(35.4 \pm 2.1)10^{14}$
TOT – 7	12 – 14 cm	$(7.25 \pm 2.11)10^6$	$(28.6 \pm 1.7)10^6$	$(4.9 \pm 1.1)10^6$	$(19.3 \pm 1.4)10^{14}$
TOT – 9	16 – 18 cm	$(3.49 \pm 0.25)10^6$	$(27.1 \pm 1.6)10^6$	$(2.5 \pm 0.6)10^6$	$(26.7 \pm 1.6)10^{14}$
TOT – 10	18 – 20 cm	$(4.63 \pm 1.29)10^6$	$(163.9 \pm 4.3)10^6$	$(3.7 \pm 0.6)10^6$	$(22.3 \pm 1.6)10^{14}$
TOT – 11	20 – 22 cm	$(2.09 \pm 0.12)10^6$	$(28.9 \pm 1.5)10^6$	$(3.4 \pm 0.8)10^6$	$(25.5 \pm 1.6)10^{14}$
TOT – 12	22 – 24 cm	$(2.38 \pm 0.86)10^6$	$(12.2 \pm 1.0)10^6$	$(1.3 \pm 0.4)10^6$	$(24.3 \pm 1.2)10^{14}$
TOT – 13	24 – 26 cm	$(3.44 \pm 0.34)10^6$	$(6.5 \pm 0.7)10^6$	$(1.9 \pm 0.4)10^6$	$(22.3 \pm 0.7)10^{14}$

Table 1.- ^{129}I, ^{239}Pu, ^{236}U and ^{238}U concentrations (atoms/g) in the different layers of the analyzed peatbog

CODE	DEPTH (cm)	$^{240}Pu/^{239}Pu$	$^{236}U/^{238}U$	$^{236}U/^{239}Pu$
TOT – 2	2 -4 cm	0.150 ± 0.007	$(3.67 \pm 0.34)10^{-8}$	0.23 ± 0.01
TOT – 5	8 – 10 cm	0.171 ± 0.010	$(0.99 \pm 0.09)10^{-8}$	0.19 ± 0.01
TOT – 7	12 – 14 cm	0.173 ± 0.024	$(0.25 \pm 0.06)10^{-8}$	0.17 ± 0.04
TOT – 9	16 – 18 cm	0.100 ± 0.018	$(0.09 \pm 0.02)10^{-8}$	0.09 ± 0.02
TOT – 10	18 – 20 cm	0.069 ± 0.005	$(0.17 \pm 0.04)10^{-8}$	0.02 ± 0.01
TOT – 11	20 – 22 cm	0.091 ± 0.014	$(0.13 \pm 0.03)10^{-8}$	0.12 ± 0.03
TOT - 12	22 – 24 cm	0.105 ± 0.025	$(0.05 \pm 0.02)10^{-8}$	0.11 ± 0.04
TOT - 13	24 – 26 cm	0.105 ± 0.037	$(0.09 \pm 0.02)10^{-8}$	0.29 ± 0.07

Table 2.- $^{240}Pu/^{239}Pu$, $^{236}U/^{238}U$ and $^{236}U/^{239}Pu$ atom ratios in the different layers of the analyzed peatbog.

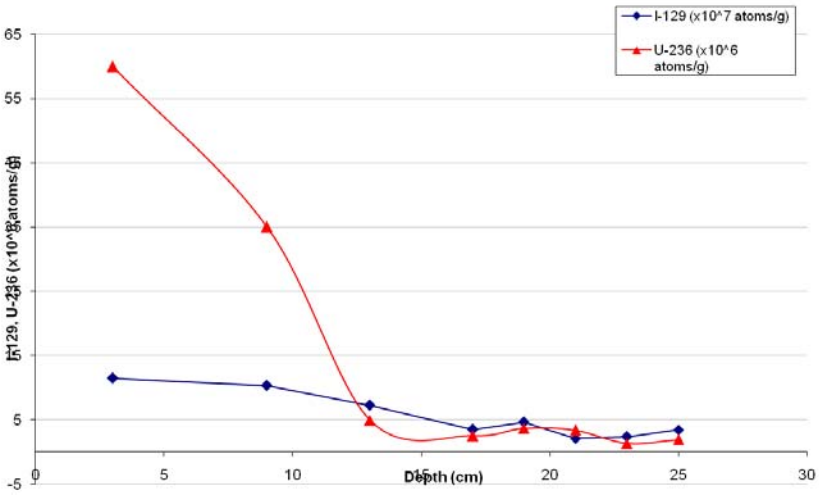

Figure 1.- ^{129}I and ^{236}U profiles in the analyzed core.

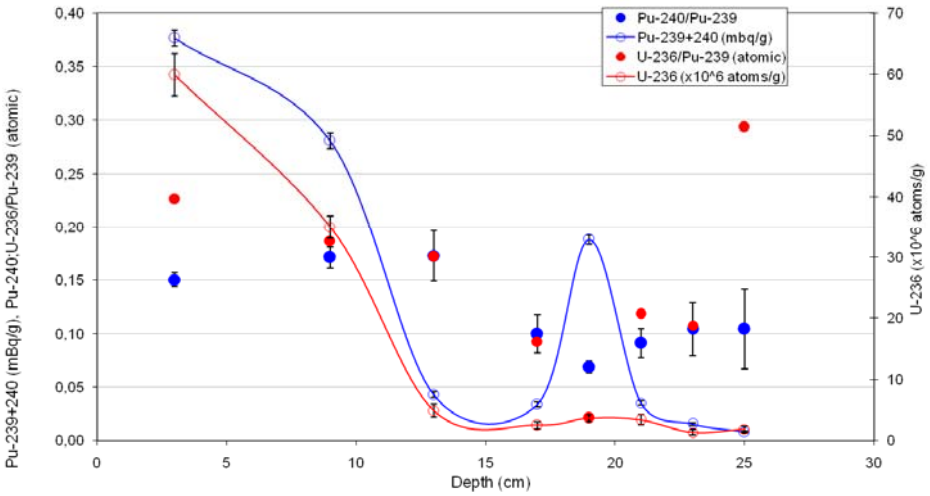

Figure 2.- $^{239+240}Pu$ (mBq/g), $^{240}Pu/^{239}Pu$ and $^{236}U/^{239}Pu$ atom ratios profiles

Discussion

The set of obtained results deserve the following comments:

-The ^{129}I deposited historically is not preserved without translocation in the different layers of the peatbog, moving along the entire core. The levels of ^{129}I are on the other hand low, indicating the possible no accumulation of all the ^{129}I atmospherically deposited over time.

-The ^{239}Pu profile shows enhanced concentrations of this radionuclide in the uppermost layers indicating an upward mobilization of a fraction of the historical record of deposited ^{239}Pu over time. However, a clear peak is observed around 18-20 cm, which can be associated to the maximum deposition of global fallout in 1962-63. The chronology established by the ^{210}Pb method ratifies the mentioned age-depth association.

-The ^{236}U profile indicates that this radionuclide in the peatbog suffers its remobilization, being not preserved any historical information about its deposition. Its mobility is clearly higher than the observed one for Pu.

-The mean ^{240}Pu/^{239}Pu atom ratio is lower than the value for global fallout, but in a good number of the uppermost layers, the results are consistent with the average ratio of 0.17 ± 0.03 for the southern hemisphere. In the deepest layers, associated to the years 1950-1970, even lower values have been found, fact that can be an evidence of the deposition by local fallout of some ^{239}Pu with origin in the disintegration over the collection area of the SNAP-9A satellite at the beginning of the 60´s. NEVERTHELESS, MORE EVIDENCES ARE NEEDED.

-The ^{236}U/^{238}U atom ratios are at least three orders of magnitude higher than natural background . ^{236}U anthropogenic origin of this radionuclide in the southern hemisphere is global fallout, as ^{236}U is produced by nuclear or thermonuclear bombs.

-The values of the ^{236}U/^{238}U atom ratio are on the other hand clearly lower than the found one in the vicinities of reprocessing plants and nuclear facilities, because another fraction of anthropogenic ^{236}U in the environment is originated in the mentioned facilities.

Generation of similar profiles in other peatbog cores collected in the area are now under development with special emphasis in the analysis of the actinide nuclides (^{236}U, ^{239}Pu and ^{240}Pu) by AMS and the determination of ^{238}Pu by alpha-particle spectrometry. The objective is to evaluate the magnitude of possible local fallout existing in the area with origin in the accident of the SNAP-9A satellite.

Acknowledgements

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References

E. Chamizo, S.M. Enamorado, M.García-León, M. Sutter and L. Wacker
Plutonium Measurements on the 1 MV AMS system at the Centro Nacional de Aceleradores (CNA)
Nuclear Instruments and Methods B 266 (2008) 4948-4954

E.Chamizo, M. Christi and L.K.Fifield
Measurement of ^{236}U on the 1 MV AMS system at the Centro Nacional de Aceleradores (CNA)
Nuclear Instruments and Methods B 358 (2015) 45-51

J.M.Gomez-Guzmán, J.M.Lopez-Gutierrez, A. R. Pinto, E.Holm
^{129}I measurements on the 1MV AMS facility at the Centro Nacional de Aceleradores (CNA, Spain)
Applied Radiation and Isotopes 70 (2012) 263-268

M. Lehritani, J. Mantero, N. Casacuberta, P. Masqué and R. García-Tenorio
Comparison of two sequential separation methods for U and Th determination in environmental samples by alpha-particle spectrometry
Radiochimica Acta 100 (2012) 431-438

F.J. Santos, J.M.Lopez-Gutierrez, E. Chamizo, M. García-León and H. Synal
Advances on the determination of ^{129}I by accelerator mass spectrometry (AMS)
Nuclear Instruments and Methods B 249 (2006) 772-775

G. Scognamiglio, E. Chamizo, J.M.Lopez-Gutierrez, A.M.Muller, S. Padilla, F.J. Santos, M. López-Lora, C.Vivo-Vilches and M. García-León
Recent developments of the 1MV AMS facility at the Centro Nacional de Aceleradores
Nuclear Instruments and Methods B 375 (2016) 17-25