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Plutonium and americium concentrations and vertical profiles in some Italian mosses used as bioindicators

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We have examined the uptake of actinide elements Am and Pu by different species of lichen and moss collected in two locations (Urbino, Central Italy; Alps region, North-east Italy). Plutonium and americium were separated and determined by extraction chromatography, electrodeposition and alpha-spectrometry. This paper summarizes our results with a special emphasis on the vertical profiles of these actinides in two different species of mosses. Several 1–2 cm depth sections were obtained and dated by ^{210}Pb method. A typical peak for ^{239,240}Pu and ^{241}Am was found in the very old moss species ("*Sphagnum Compactum*") at a depth corresponding to the period 1960–1970 which was the period characterized by the maximum nuclear weapon tests. In a younger moss species ("*Neckeria Crispa*") no peak was observed and the regression curves showed that Am is more mobile than ^{239,240}Pu and ^{238}Pu.

Introduction[+]

Bioindicators are often used to assess the occurence of airborne pollutants. In fact, they can provide basic information on pollution levels as well as on the bioavailability of airborne chemical species of present and past times. Mosses and lichens have been shown to be useful as bioindicators.[1–4] Furthermore, some peculiar mires appear to be particularly suitable to investigate atmospheric contamination and they may provide a record of the history of atmospheric fall-out, especially when collected in appropriate sampling sites (open areas, absence of trees and vascular plants). this is the case of "Sphagnum" mosses. Under such conditions these moss species are very effective collectors of airborne materials, and they represent the most productive plant of these ecosystems to reconstruct past pollution events.

This paper examines the concentration values of ^{238}Pu, ^{239,240}Pu and ^{241}Am (^{241}Pu daughter) in some Italian mosses and lichens. Peculiar vertical distributions were also obtained by determining these radionuclides in two different species of moss as a function of the plant height.

Experimental

Sampling

Figure 1 shows the two sites where the samples were collected. The first site was at 400–500 m above sea level (a.s.l.) in Central Italy: lichens from tree trunks, mosses from tree trunks and terrestrial mosses were collected taking care to remove tree bark, soil and other foreign matter. One very high and young moss species called "*Neckeria Crispa*" was also collected in order to assess trends in the vertical distribution and migration.

The second sampling site was located in the Dolomitic Alps (North-east Italy at 1500 m a.s.l.). In this case the collected moss species was "*Sphagnum Compactum*", a very old, compact and high kind of moss.

To obtain the vertical distribution samples were cut into sections and analyzed separately. All the samples were dried to constant weight at 105 °C for 24 h, ground and homogenized.

Radioanalytical method

Plutonium and americium radioanalytical determination was carried out by extraction chromatography, electrodeposition and alpha-spectrometry.

0.5–2 g of dried sample were placed in a beaker with known activities of ^{242}Pu and ^{243}Am (0.01–0.03 Bq) as the chemical yield tracers. After a strong chemical attack with concentrated HNO_3 and H_2O_2, plutonium was separated by using a Microthene-Tri-n-octylphosphine oxide (TOPO) column, electroplated from a $(NH_4)_2SO_4$ solution and counted by alpha spectrometry. The final yield ranged from 70% to 85%.

^{241}Am was determined in the effluent collected from the chromatographic column. A second column of Microthene-HDEHP [di (2-ethyl hexyl) phosphoric acid] and a liquid-liquid extraction with PMBP (1-phenyl-3-methyl-4-benzoyl-5-pyrazolone) + TOPO were used to separate and purify ^{241}Am. After an electrodeposition from a $H_2C_2O_4$+HNO_3 solution the measurement by alpha spectrometry was carried out. The final yield ranged from 50% to 70%.

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Table 1. Plutonium and americium concentration in mosses and lichens of Urbino area

Kind and number of samples	Concentration range, Bq/kg			Mean concentration, Bq/kg		
	^{239,240}Pu	^{238}Pu	^{241}Am	^{239,240}Pu	^{238}Pu	^{241}Am
Tree trunk mosses (12)	0.32±0.07 – 4.96±0.23	0.03±0.02 – 0.17±0.04	0.20±0.05 – 1.93±0.16	1.39	0.09	0.65
Tree trunk lichens (8)	0.83±0.11 – 1.87±0.14	0.05±0.02 – 0.15±0.05	0.18±0.05 – 0.77±0.09	1.13	0.10	0.44
Terrestrial mosses (5)	0.034±0.004 – 0.56±0.04	0.002±0.001 – 0.036±0.007	0.015±0.004 – 0.128±0.014	0.19	0.01	0.07

Fig. 1. Sampling sites: *Urbino Area, Central Italy, 400–500 m a.s.l.; ♦ Dolomitic Alps, North-east Italy, 1500 m a.s.l.

The counting was performed for 2,000–10,000 minutes, depending on the activity levels, by using a 450 mm^2 detector: the counting efficiency was 31% and the background $5 \cdot 10^{-4}$ cpm. By measuring the sample for 10,000 minutes the detection limits for 2 g sample were 0.028 Bq/kg and 0.034 Bq/kg for ^{239,240}Pu (^{238}Pu) and ^{241}Am respectively. Only for some terrestrial mosses of the Urbino area, where the plutonium and americium concentration as very low, a bigger quantity (16–40 g) of sample had to be analyzed to get the necessary sensitivity.

For more details on the radioanalytical method see also References 5 and 6.

Results

Urbino area

Twelve samples of tree trunk mosses, eight samples of tree trunk lichens and five samples of terrestrial mosses were analyzed.[7] Table 1 summarizes the results showing the concentration ranges and the relevant mean for ^{239,240}Pu, ^{238}Pu and ^{241}Am. By taking into account these results, it appears that the mean concentrations follow the trend: tree trunk mosses ≥ tree trunk lichens > terrestrial mosses. Furthermore these results show that, in all the three analysed groups, ^{239,240}Pu concentration is greater than ^{241}Am concentration and the last one much greater than ^{238}Pu concentration. The ratio ^{239,240}Pu/^{241}Am ranges from 2 to 3, the ratio ^{241}Am/^{238}Pu ranges from 3 to 6 and the ratio ^{239,240}Pu/^{238}Pu ranges from 10 to 15.

Vertical distribution in a "Neckeria Crispa" moss: ^{239,240}Pu, ^{238}Pu and ^{241}Am vertical distributions in a young and high terrestrial moss ("*Neckeria Crispa*") drawn near Urbino were obtained by cutting several 2 cm transversal sections. The results indicate that all three radionuclides are present in the lower part of the plant and that they are undetectable above a 16 cm height (Fig. 2). As expected, the regression curve slope for ^{238}Pu is very close to that of ^{239,240}Pu; on the contrary, the ^{241}Am regression curve shows a lower slope indicating that Am is more mobile within the plant structure than Pu in this kind of moss.

Dolomites area

Vertical distribution of ^{239,240}Pu and ^{241}Am in a "Sphagnum Compactum" moss: ^{239,240}Pu and ^{241}Am vertical distribution in a very peculiar and old terrestrial moss ("*Sphagnum Compactum*") drawn on the Dolomitic Alps (North-east Italy) is shown in Fig. 3. An interesting peak at 5–6 cm from the top is observed for both radionuclides. By dating the different sections through ^{210}Pb method[8] peak was found to correspond to the period 1960–1970 when the fall out depositions were very heavy due to the several nuclear weapon tests in the open air.

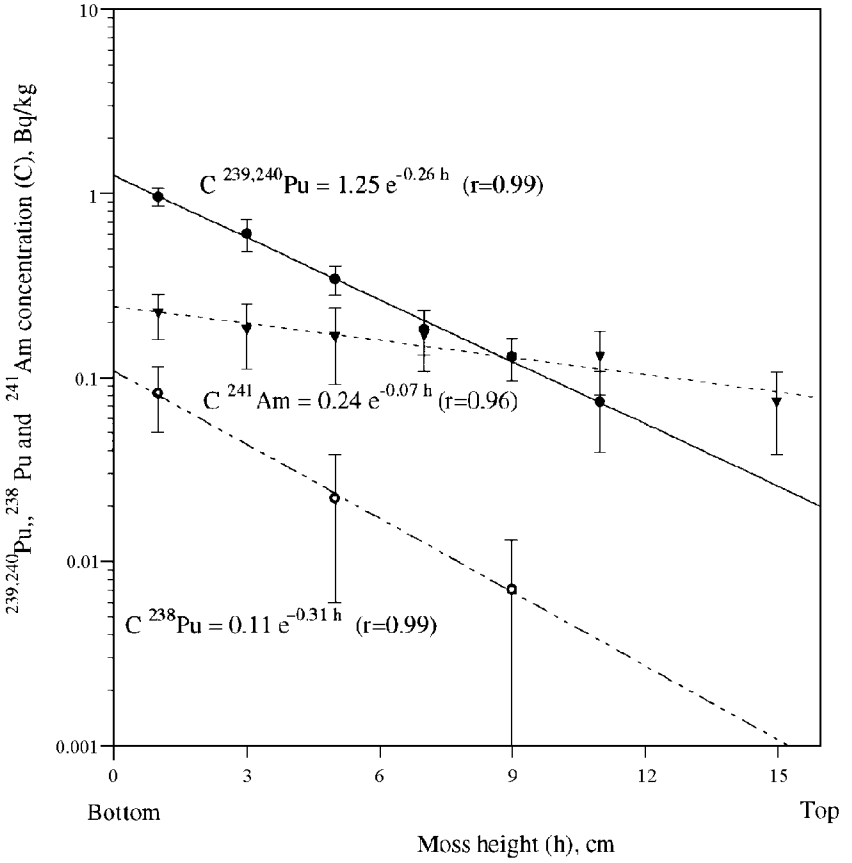

Fig. 2. • ^{239,240}Pu, ○ ^{238}Pu and ▽ ^{241}Am vertical distribution in the terrestrial moss "*Neckeria Crispa*" drawn near Urbino (Central Italy)

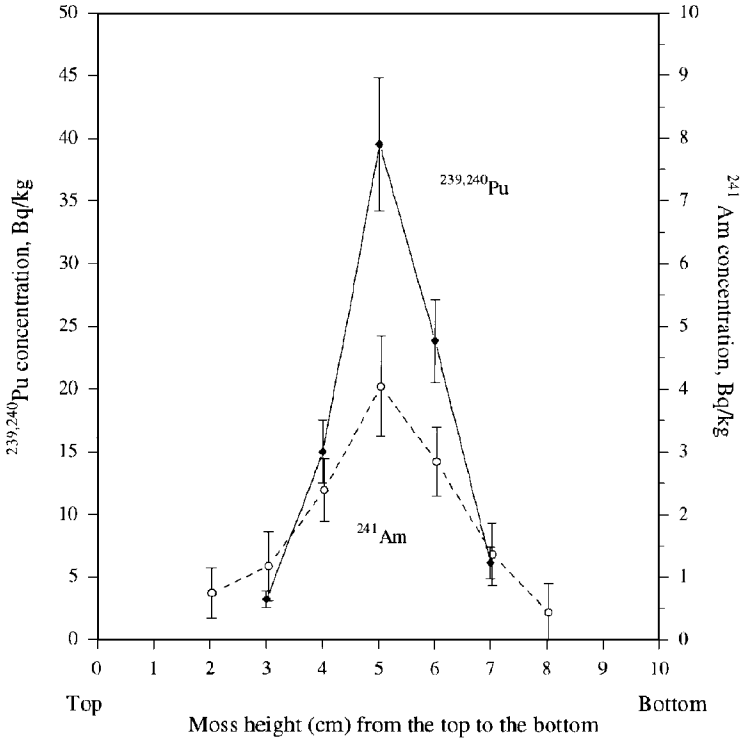

Fig. 3. ^{239,240}Pu and ^{241}Am vertical distribution in a "*Sphagnum Compactum*" moss drawn in a Dolomites area

Conclusions

The results show that both mosses and lichens are very effective accumulators of Pu and Am. They may act as efficient filters in trapping airborne radioactive contaminants as well as natural reservoirs from which these radionuclides are periodically released. Therefore, both mosses and lichens can be recommended as very good biological indicators of Pu and Am fallout caused by nuclear facilities accidents and from nuclear weapon tests. Some long life moss species can play an important role in cycling naturally or artificially enhanced radionuclides in the atmosphere over long time scales. The results also show that ^{241}Am contents in these plants are high enough to be determined by a sensitive radiochemical method.

The studies of these radiotracers are important to predict the level and distribution of the radionuclides in the environment, to understand their biological and ecological behaviour in a given contamination condition.

As far as the very old and peculiar Sphagnum mosses are concerned, the results show that they can keep the "memory" of pollution events occurred even in past times. Therefore by dating the vertical sections through the ^{210}Pb method and by analyzing some non mobile radioactive or stable elements it is possible to have an idea of the period characterized by the relevant pollution.

References

1. E. NIEBOER, D. H. S. RICHARDSON, F. D. TOMASINI, Bryologit, 81 (1978) 226.
2. C. PAPASTEFANOU, M. MANALOPOULOU, T. SAWIDIS, Elsevier Science Publishers Ltd, England, 1989, p. 199.
3. N. S. JOHN, M. ELLIS KATHERINE, Elsevier Science Publishers Ltd, England, 1990, p. 151.
4. M. R. D. SEAWARD, University of Bradford, 1992.
5. D. DESIDERI, M. A. MELI, F. NONNIS MARZANO, C. ROSELLI, C. TESTA, C. TRIULZI, M. VAGHI, Chem. Ecol., 12 (1996) 259.
6. G. JIA, D. DESIDERI, F. GUERRA, M. A. MELI, C. TESTA, J. Radioanal. Nucl. Chem., 220 (1977) 15.
7. G. JIA, D. DESIDERI, F. GUERRA, M. A. MELI, C. TESTA, J. Radioanal. Nucl. Chem., 222 (1997), in press.
8. S. DEGETTO et al., Syntheses and Methodologies in Inorganic Chemistry, 6 (1996) 426.