ATMOSPHERIC INPUT OF RADIONUCLIDES TO THE NORTHWESTERN MEDITERRANEAN SEA

Sang-Han Lee*, Pavel P. Povinec, Mai K. Pham

International Atomic Energy Agency, Marine Environment Laboratory, Monaco - S.Lee@iaea.org

Abstract

^{137}Caesium, ^{238}Pu, ^{239,240}Pu and ^{241}Am were determined in total monthly deposition (wet + dry) to elucidate the delivery behaviour of anthropogenic radionuclides into the northwestern Mediterranean Sea. The results show that the concentrations of ^{239,240}Pu in precipitation were similar to those found in Mediterranean surface water while the concentrations of ^{241}Am in precipitation were higher. The annual contribution of transuranics from the atmosphere to northwestern Mediterranean was much lower than their mean annual loss. However, many particles loaded in precipitation in February and August probably played important roles in enhancing the scavenging of transuranics in the northwestern Mediterranean Sea.

Keywords: radionuclides, precipitation, atmosphere, Mediterranean Sea

Introduction

Atmospheric input of anthropogenic radionuclides to the ocean has been regarded as one of the most important of the various input sources such as river input, discharges from nuclear industrial activities and accidents, etc. For this reason, in order to study the fluxes of anthropogenic radionuclides, their biogeochemical behaviour and the high scavenging rates of transuranics in the Mediterranean Sea, as well as for monitoring possible accidental discharges of radionuclides to the sea, we regularly collected rainwater (wet and dry deposition) to estimate the amounts of delivery and to measure the concentrations of anthropogenic radionuclides.

Methods and Materials

Precipitation was collected monthly in a 2.35 m^2 plastic container placed on the roof of the building which houses IAEA-MEL in Monaco. Preconcentration of radionuclides in rainwater, radiochemical separation, purification steps and α-spectrometry measurements have been described elsewhere [1].

Results and discussion

Radionuclide concentrations in precipitation and monthly deposition rates are presented in Table 1 and Figure 1, respectively. The monthly deposition of radionuclides into the northwestern Mediterranean Sea shows that a significant amount was delivered from September to October when high precipitation rates were observed. A strong spring peak observed in previous studies [2] was not observed in the recent study; only a small sub-peak was noted at this time. The annual delivery of anthropogenic radionuclides into the Sea was calculated to be 0.51 ± 0.06 mBq m^{-2} for ^{238}Pu, 16.5 ± 0.4 mBq m^{-2} for ^{239,240}Pu, 6.8 ± 0.4 mBq m^{-2} for ^{241}Am and for ^{137}Cs 1.17 ± 0.12 Bq m^{-2}. These amounts correspond to 8% for ^{238}Pu, 6% for ^{239,240}Pu and 32% for ^{241}Am of those measured previously by Thein *et al.* [2] in Monaco during 1978-79.

These results demonstrate that the transport of anthropogenic radionuclides from the stratosphere to the troposphere and then to the sea, which was observed previously, has been replaced by resuspension of radionuclides from soil and their wash-out from air to seawater. Therefore, at present, delivery patterns of radionuclides are dependent on the amounts of precipitation. The recent annual discharges of radionuclides to the Mediterranean Sea through the adjacent rivers are poorly known and only approximate data and gross activities have been reported [3, 4]. If we assume that the same amounts of radionuclides as estimated by Fukai *et al.* [3] are discharged into the Mediterranean Sea (in reality, the present amounts should be lower), the observed deposition amounts of ^{239,240}Pu and ^{241}Am are slightly higher than annual discharges by rivers. The observed deposition amounts are only 4% for ^{239,240}Pu and 5% for ^{241}Am of the mean annual transuranic loss in surface waters measured in the northwestern Mediterranean [5]. Therefore, the total inputs from both the atmosphere and rivers are less than 10% of the annual loss in surface water of the northwestern Mediterranean.

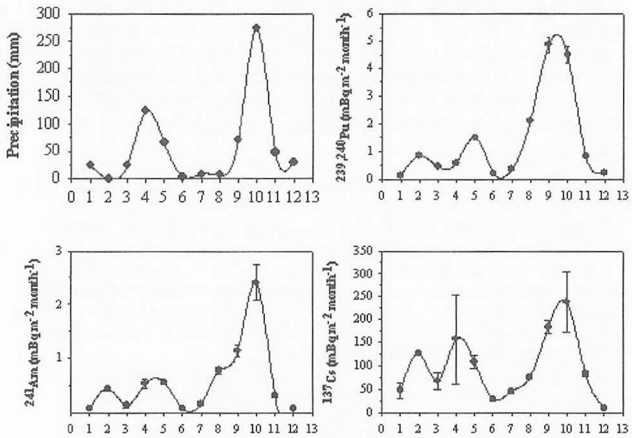

Fig. 1. Monthly deposition (mBq m^{-2} month^{-1}) of radionuclides and precipitation (fallout) into northwestern Mediterranean Sea, 1999.

Table 1. Concentrations and activity ratios of anthropogenic radionuclides in precipitation over Monaco, 1999.

Month	Precipitation (mm)	^{238}Pu (µBq L^{-1})	^{239,240}Pu (µBq L^{-1})	^{241}Am (µBq L^{-1})	^{137}Cs (µBq L^{-1})
January	25	1.4 ± 0.3	4.9 ± 0.7	3.6 ± 0.7	1.9 ± 0.7
*February	-	116 ± 31	3300 ± 180	1780 ± 130	126 ± 1
March	25	2.1 ± 0.6	18 ± 2	5.8 ± 1.6	2.7 ± 0.7
April	123	0.4 ± 0.2	4.7 ± 0.7	4.4 ± 0.7	1.3 ± 0.8
May	65	0.5 ± 0.1	23 ± 1	8.7 ± 0.7	1.7 ± 0.2
June	3.8	2.3 ± 0.6	530 ± 30	23 ± 2	7.4 ± 1.0
July	8.3	1.3 ± 0.3	42 ± 2	21 ± 2	5.6 ± 0.5
August	7.5	7.4 ± 1.5	280 ± 10	105 ± 8	10 ± 1
September	70	1.4 ± 0.6	70 ± 4	16 ± 1	2.6 ± 0.2
October	275	-	16 ± 1	8.8 ± 1.2	0.9 ± 0.2
November	48	2.6 ± 0.4	17 ± 1	6.7 ± 0.8	1.8 ± 0.1
December	30	0.2 ± 0.1	7.0 ± 0.7	2.8 ± 0.3	0.29 ± 0.04

Month	Precipitation (mm)	^{238}Pu/^{239,240}Pu	^{241}Am/^{239,240}Pu	(^{239,240}Pu/^{137}Cs)* 1000
January	25	0.286 ± 0.081	0.73 ± 0.17	2.6 ± 1.0
*February	-	0.035 ± 0.010	0.54 ± 0.05	26.3 ± 1.5
March	25	0.119 ± 0.036	0.33 ± 0.10	6.6 ± 1.9
April	123	0.089 ± 0.046	0.93 ± 0.20	3.7 ± 2.4
May	65	0.023 ± 0.005	0.38 ± 0.03	14 ± 2.0
June	3.8	0.044 ± 0.011	0.44 ± 0.04	7.1 ± 1.0
July	8.3	0.031 ± 0.008	0.49 ± 0.04	7.5 ± 0.7
August	7.5	0.026 ± 0.006	0.37 ± 0.03	28 ± 2
September	70	0.021 ± 0.009	0.23 ± 0.02	26 ± 2
October	275	-	0.54 ± 0.08	19 ± 5
November	48	0.155 ± 0.025	0.39 ± 0.05	10 ± 1
December	30	0.034 ± 0.017	0.40 ± 0.06	24 ± 4

** Dry deposition (no rain): values are in µBq*

The activity ratios of ^{238}Pu/^{239,240}Pu show that plutonium in total deposition originates mainly from global fallout. Noticeable exceptions are in January, March and November and the high deviations from the global fallout ratio (0.024) for these months will require careful interpretation when additional results become available. The ^{241}Am/^{239,240}Pu ratios (weighted mean 0.38), do not reveal any signature of a special input source compared to that of global fallout (0.37). The observed mean activity ratio of ^{239,240}Pu/^{137}Cs is 0.015. When taking decay correction into account, our result is in good agreement with that (0.023) of Thein *et al.* [2].

References

[1] - La Rosa J., Burnett W., Lee S-H., Levy I., Gastaud J. and Povinec P.P., (in print). Separation of actinides, caesium and strontium from marine samples using extraction chromatography and sorbents. *J. Radional. Nucl. Chem.*

[2] - Thein M., Ballestra S., Yamato A. and Fukai R., 1980. Delivery of transuranic elements by rain to the Mediterranean Sea. *Geochim et Cosmochimica Acta.*, 44: 1091-1097.

[3] - Fukai R., Ballestra, S., Thein M. and Guion, J., 1981. Input of transuranic elements through rivers into the Mediterranean Sea. Report SM-248/125, *In* : Impact of radionuclide release into the marine environment. IAEA, Vienna, 3-14.

[4] - Martin J. M., and Thomas A. J., 1990. Origins, concentrations and distributions of artificial radionuclides discharged by the Rhone river to the Mediterranean Sea. *J. Environ. Radioactivity*, 11: 105-139.

[5] - Fowler S. W., Noshkin V. E., La Rosa J. and Gastaud, J., 2000. Temporal variations in plutonium and americium inventories and their relation to vertical transport in the northwestern Mediterranean Sea. *Limnol. Oceanogr.*, 45(2) 446-458.