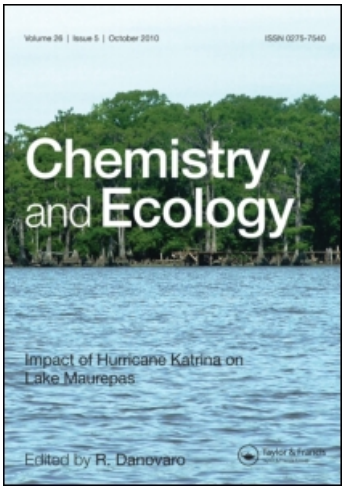

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RADIOACTIVE ISOTOPES OF STRONTIUM, CAESIUM AND PLUTONIUM IN SEDIMENTS OF THE NORTHERN ADRIATIC SEA

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Sediment samples of different strata (0–3 and 12–15 cm) were collected between 1990 and 1992 during seasonal cruises in the Northern Adriatic Sea. A complete mapping of the Sr-90, Cs-137, Pu-238 and Pu-239(240) concentrations was obtained for samples covering a wide area, stretching from the Gulf of Trieste towards the Ancona shoreline. Sr-90 concentrations varied between 1.5 and 6.5 Bq kg^{-1} dw, Cs-137 was in the range 0.9–38.9 Bq kg^{-1} dw, Pu-239(240) in the range 0.08–1.5 Bq kg^{-1} dw and Pu-238 around 0.03 Bq kg^{-1} dw. Special reference was also put on the comparison between the off-shore environment and data obtained from samples collected inside the Po river delta. In spite of the major accumulation of Cs-137 inside the estuarine environment, higher concentrations of strontium and plutonium isotopes were detected in the offshore environment.

KEY WORDS: Artificial radioisotopes, caesium, plutonium, strontium, Adriatic Sea, Po delta.

INTRODUCTION

A radioecological assessment of the Northern Adriatic sediment layer was performed in the framework of a collaboration between the Radioecology Laboratory of Parma University and the Institute of General Chemistry of Urbino University. This paper is part of a wider work carried out by both research groups concerning the investigation of several pollutants in the marine environment.

In the past, data about different radionuclides detected in biotic and abiotic specimens and their cycling within the Adriatic ecosystem have already been presented (Nonnis Marzano and Triulzi, 1994). These studies pointed out the importance of the Adriatic sediment compartment as a final point of radiopollutant accumulation. For this reason, the investigations in this study were aimed only to the determination of the highly persistent radionuclides Sr-90, Cs-137, Pu-238 and Pu-239(240) in different sediment samples, with special emphasis on the mapping of their concentrations in an area stretching from the Gulf of Trieste towards the Ancona shoreline.

Particular attention was also given to the comparison between the radioactivity concentration determined in the offshore area and that of the Po river delta. In a recent work, Bondavalli (1995) reported this estuarine environment as an area of strong Cs-137 accumulation. Our previous papers (Nonnis Marzano and Triulzi, 1994) illustrated that the river has strongly influenced the depositional pattern of radionuclides in the open sea after the Chernobyl event. Furthermore, the importance of this kind of radioecological investigations in the Adriatic Sea is supported by the intense commercial fishing and increasing sea-farming activities in the area.

SAMPLING AND METHODS

Sediment samples of different strata (mainly 0–3 cm and 12–15 cm) were collected in 1990 by means of a box corer in sites near the main Adriatic cities. The sampling was performed at the border of the Italian and Croatian international waters (Fig. 1). In addition, surface sediment samples were collected in two stations of the Goro lagoon (Po river delta) in 1992 to compare different environments: one sampling station located in the middle of the lagoon (St. 4), influenced by the tidal current, with sediment composed mainly of clay and silt and a more protected station (St. 5/8) with sandy sediment less influenced by the river outputs and the river-sea exchanges (Fig. 1).

After drying and grinding the samples, the Cs-137 concentrations (0.662 MeV) were determined through direct gamma spectrometry by means of a PGT Silena germanium detector. The data were elaborated with an IBM PC equipped with an EG&G Ortec software programme.

Strontium-90 and plutonium isotopes determinations were carried out after specific radiochemical separations (Delle Site *et al.*, 1980). Plutonium-242 (4 dpm) as internal tracer was added to the ashed sediment which was subject to HNO_3 attack. Columns of Microthene-TOPO (tri-n-octylphosphine oxide) followed by a classical electrodeposition on stainless steel disks were then carried out.

The sources were hence counted by alpha spectrometry with a EG&G-Ortec silicon chamber for the identification of Pu-238 (5.5 MeV), Pu-239(240) (5.1 MeV) and Pu-242 (4.9 MeV). The data were elaborated with the EG&G-Ortec programme above reported.

Strontium was determined from the eluate resulting from the plutonium extraction. By this technique (Testa *et al.*, 1990) Y-90 (Sr-90 descendant) was extracted with di-(2-ethylhexyl)-phosphate (HDEHP) and precipitated with oxalic acid. Y-90 was then measured by means of a ASPN low background beta counter.

RESULTS AND DISCUSSION

Caesium-137 was the most persistent and abundant radionuclide detected in fallout samples of the Po plain during pre and post-Chernobyl periods (Nonnis and Triulzi 1994). The contribution of the Chernobyl event to the Sr-90 and plutonium isotopes environmental contamination was negligible (ENEA, 1986). The presence

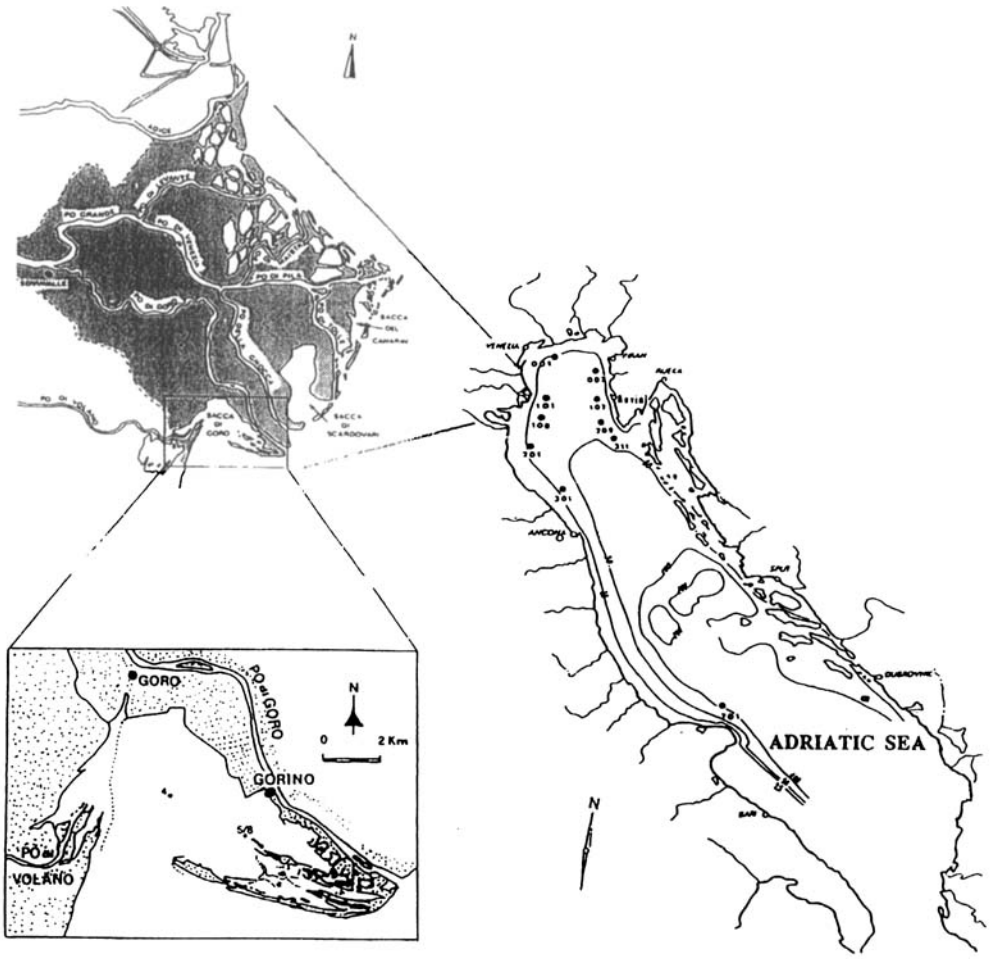

Figure 1 Study area in the Adriatic Sea (right) and Po river delta (left).

of plutonium and strontium over the Mediterranean environment was therefore mostly related to the nuclear atmospheric testing of the sixties and seventies. In spite of the long time elapsed, their long half-lives make these fission and transuranic products still detectable in the Northern Adriatic Sea.

The distribution of Cs-137 in the offshore environment partly reflected the different Chernobyl deposition over the area. Higher concentrations were generally detected along the Italian coastline with a north-south gradient, the highest value (18 Bq kg^{-1}*) being determined in front of the Po river delta (St. 108).

Concentrations of Sr-90 varied between the detection limit (1.5 Bq kg^{-1}) and 6.4 Bq kg^{-1} for the top layer and between <1.5 and 6.5 Bq kg^{-1} for the underlying strata.

Lower values were observed for the plutonium isotopes. The concentrations of Pu-239(240) ranged between 0.21 and 1.23 Bq kg^{-1} for the top strata and 0.08–1.47

*Different concentrations are given as dry weights.

$Bq\,kg^{-1}$ for the ones underneath. Concentrations of Pu-238 were generally low and very close to the detection limits: an average value of 0.03 $Bq\,kg^{-1}$ being the most common result.

A complete representation of the different concentrations detected is reported in Table I.

For the Sr-90 and plutonium concentrations, the values detected in the two samples of the Sacca di Goro (estuarine ecosystem) were lower than those determined in the open Adriatic. Strontium-90 was in the range 1.9–2.8 $Bq\,kg^{-1}$, Pu-239(240) variable between 0.05 and 0.15 $Bq\,kg^{-1}$ and Pu-238 lower than the detection limits (<0.01 $Bq\,kg^{-1}$).

On the contrary, the behaviour of Cs-137 had an opposite trend. Concentrations as high as 39 $Bq\,kg^{-1}$ were measured in St. 4 (central station) strongly influenced by the tidal current and lower values outside the river mouth. Moreover, the river seemed to have largely influenced the Cs-137 depositional pattern even in the open sea area. In fact, the Italian coastline showed higher radioactivity levels along the river plume (which stretches approximately until station 301) where the sediment composition is mostly made of clay and silt.

Figure 2 presents a comparison between concentrations of Sr-90, Cs-137 and Pu-239(240) detected in three representative stations (one inside the delta and two representing the opposite sides of a transect facing the river mouth). It is

Table I Concentrations ($Bq\,kg^{-1}$ dw) of Sr-90, Cs-137, Pu-239(240) and Pu-238 in estuarine and marine sediment samples. (Sample A&B are shown below.)

Sampling station	*Sr-90* $Bqkg^{-1}$ *dw*	*Cs-137* $Bqkg^{-1}$ *dw*	*Pu-239 (240)* $Bqkg^{-1}$ *dw*	*Pu-238* $Bqkg^{-1}$ *dw*
Open sea-international waters				
001 Venice A	6.44 ± 0.54	2.94 ± 0.20	0.50 ± 0.05	0.03 ± 0.01
Venice B	<1.5	1.49 ± 0.32	0.53 ± 0.06	0.02 ± 0.01
007 Porec A	1.50 ± 0.16	2.51 ± 0.30	0.24 ± 0.02	<0.05
101 Po delta A	5.50 ± 0.22	8.76 ± 0.65	0.44 ± 0.03	0.03 ± 0.01
Po delta B	2.87 ± 0.24	1.51 ± 0.30	0.21 ± 0.03	<0.06
107 Rovinj A	5.76 ± 0.48	2.55 ± 0.30	0.47 ± 0.05	0.02 ± 0.01
Rovinj B	2.13 ± 0.24	0.87 ± 0.20	0.08 ± 0.01	0.002 ± 0.001
108 Po delta A	4.90 ± 0.50	18.1 ± 0.50	0.80 ± 0.09	0.03 ± 0.01
Po delta B	6.50 ± 0.70	8.42 ± 0.60	1.47 ± 0.15	0.03 ± 0.01
201 P. Corsini A	5.80 ± 0.70	17.1 ± 0.60	1.23 ± 0.13	0.02 ± 0.01
P. Corsini B	5.00 ± 0.70	6.87 ± 0.30	0.74 ± 0.10	0.02 ± 0.01
209 Pula A	4.78 ± 0.37	2.08 ± 0.24	0.32 ± 0.10	0.03 ± 0.01
Pula B	<1.50	1.60 ± 0.20	0.35 ± 0.04	0.02 ± 0.01
301 Riccione A	<1.50	8.93 ± 0.60	0.61 ± 0.10	0.03 ± 0.01
311 Porer Island A	<1.50	1.07 ± 0.20	0.21 ± 0.05	0.02 ± 0.01
701 Gargano A	–	4.35 ± 0.70	0.42 ± 0.10	<0.06
Sacca di Goro-river estuary				
St. 4 River mouth	2.80 ± 0.60	38.9 ± 0.71	0.15 ± 0.02	<0.01
St. 5/8 Internal	1.90 ± 0.50	6.77 ± 0.23	0.05 ± 0.01	<0.01

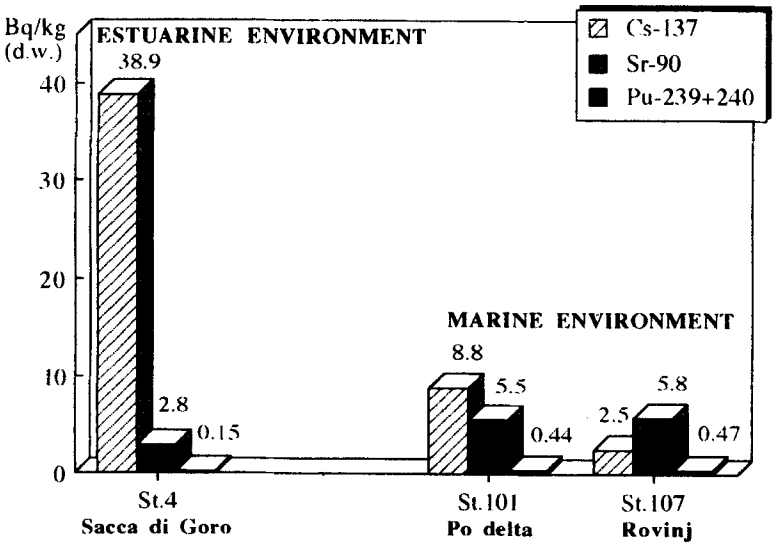

Figure 2 Comparison between concentrations (Bq kg^{-1} dw) of the different radioisotopes detected in significant stations of the estuarine and marine environments.

quite clear the difference between levels of the three radionuclides in the two environments.

The ratios Pu-238/Pu-239(240) and Sr-90/Pu-239(240) (respectively around 0.02–0.06 and 5–26) were in good agreeement with values determined by other authors (Pentreath, 1987; Delfanti *et al.*, 1994) in marine environments not influenced by nuclear power plants.

It is well known that the Chernobyl event has scarcely influenced the input of strontium and plutonium in the Mediterranean environment. Their ground deposition after the Chernobyl event has been marginal and negligible in comparison to the fallout environmental input of radiocaesium (Enea, 1986).

The ratio Cs-137/Sr-90 ranged between 1 and 4 in the marine environment and around 25 inside the Po river delta; the ratios Cs-137/Pu-239(240) resulted within the ranges 5–25 in the marine environment and it was much higher (466–600) in the estuarine ecosystem. These results confirmed that the Po delta is a real Chernobyl reservoir with particular reference to the Cs-137 accumulation. In fact, the sedimentation of Cs-137 at the estuarine bottom was generally higher than that determined in the open sea. On the contrary, concentrations of Sr-90 and plutonium isotopes were lower in the estuarine area (Fig. 2).

It will be helpful to readers that the Po river delta is an area with very shallow waters (depth 0.5–1.5 m) and high physical and biological perturbation. For such reasons, the sediment mixing and resuspension, together with the complex geochemistry of the area probably play a major role in influencing the distribution of the caesium, plutonium and strontium isotopes inside the lagoon and their transport to the open Adriatic Sea.

Acknowledgements

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