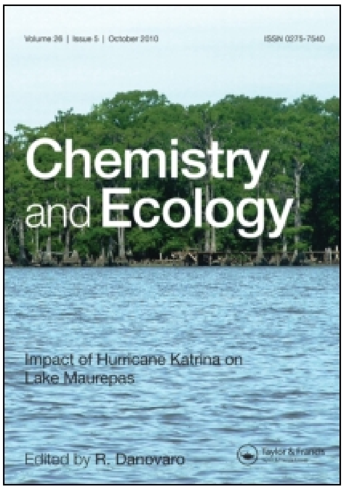

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Horizontal Distribution of Some Artificial Radionuclides in Sediments Collected Off the Rhone Mouth (Gulf of Lions-Mediterranean Sea)

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HORIZONTAL DISTRIBUTION OF SOME ARTIFICIAL RADIONUCLIDES IN SEDIMENTS COLLECTED OFF THE RHONE MOUTH (GULF OF LIONS-MEDITERRANEAN SEA)

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The Rhône delta is a typical microtidal estuary. River runoff spreads out into the Gulf of Lions as a surface plume influenced by the prevailing N or NW wind and the Liguro-Provençal-Catalan current. In the prodelta, sediments form a filter that traps radionuclides released by the different nuclear installations on the Rhône River and which are associated with fine particles. The horizontal distribution of various artificial radionuclides shows the highest concentrations at the Rhône River mouth. There, ^{137}Cs reaches 86.6 and 94.4 $Bq \cdot kg^{-1}$ dry wt. and is still present 10 miles away (32.5 $Bq \cdot kg^{-1}$ dry wt.) in the southwestward direction (influence of the general circulation). Data obtained in the sediment at Roustan station between 1980 and 1990 show that in 1990, ^{137}Cs levels were similar to those found previous to the Chernobyl accident.

KEY WORDS: Horizontal distribution, radionuclides, sediment, Gulf of Lions, Mediterranean Sea.

INTRODUCTION

Since the time the Aswan dam was built on the Nile, the Rhône River has taken on greater importance in the delivery of sediment to the Mediterranean Sea. Compared to the Ebro (200 $m^3 \cdot s^{-1}$), the Arno (103 $m^3 \cdot s^{-1}$), the Tiber (234 $m^3 \cdot s^{-1}$) and the Po rivers (1500 $m^3 \cdot s^{-1}$), the Rhône River has the largest average discharge (1700 $m^3 \cdot s^{-1}$). Its discharge varies considerably due to the complicated regimes of its tributaries, some of which have a glacial source (e.g Isére, Drôme), whereas the others are fed by rainfall in the Mediterranean region (Durance, Gard.). The Rhône River, 812 km long, drains a basin of 97800 km^2. The supply of sediment to the Mediterranean Sea is of two main origins: fluvial and aeolian. Eighty percent, i.e. 2 to $19 \cdot 10^6\, t \cdot y^{-1}$, of the fluvial supply comes from the Rhône River (Roditis, 1993), the outflow of which can reach $55 \cdot 10^9\, m^3 \cdot y^{-1}$. This load has decreased substantially since hydroelectric development in the Rhône River watershed, for example on the Durance River. Only part of this sediment is deposited at the Rhône mouth and in adjacent nearshore and coastal areas; much of the silts and clays reach the deep basins of the Mediterranean Sea. The Rhône valley is part of the North Sea/Mediterranean Sea link, and represents a region of important economic potential. For environmental and energy delivery reasons, nuclear plants are located along the river. During regular operation, they are allowed to release liquid and gaseous effluents composed of artificial radionuclides. These releases can be used as tracers of the effects of the river on the Mediterranean Sea and of sedimentary phenomena at the river mouth.

MATERIALS AND METHOD

In November 1990, a sediment sampling cruise took place, on the R/V "Georges Petit", in the Gulf of Lions. The sampling device was a "Fluchat" equipment enabling a 728 cm^2 section to a depth of about 30 cm to be taken. As this sampling method does not compress the first few centimetres, subsampling was carried out on a centimetre surface grid. The sediments were then dried at a temperature of 40 °C until a constant weight was obtained. They were then crushed, homogenized and measured by direct gamma ray spectrometry with Type N Germanium high purity detectors (20% efficiency and 1.7 keV resolution at 1.332 MeV). We present here the data obtained on the surficial sediments of the Gulf of Lions delta area, as found in 12 box-cores, as well as a brief review of data obtained previously at the Roustan station, located off the Grand Rhône river mouth.

RESULTS AND DISCUSSION

The suspended materials originating from the Rhône River discharged to the sea are subjected to different processes, such as the influence of currents and numerous other phenomena involving adsorption, desorption and particulate-dissolved phase transfer. Previous studies (Calmet and Fernandez, 1990; Calmet *et al.*, 1992) have shown that ^{137}Cs and ^{134}Cs are good tracers for studying sedimentary phenomena near the river mouth. The shape of the horizontal distribution of ^{137}Cs concentrations in the first five centimetres of sediments (Fig. 1) is similar to those found in the above water mass (Calmet *et al.*, 1992). At 1 and 2 nautical miles away from the Grand Rhône mouth, the BFRoustan (30 m depth) and BF14 (65 m) stations present the highest ^{137}Cs concentrations: 86.6 and 94.4 $Bq \cdot kg^{-1}$ dry wt., respectively. Along this transect off the Rhône river, the ^{137}Cs activity decreases rapidly, as we head offshore, from 71.8 $Bq \cdot kg^{-1}$ dry wt. at BF9 station (95 m), down to 35.2 $Bq \cdot kg^{-1}$ dry wt., 2 nautical miles further (BF8) (Table I). BF10 and BF13 stations, located west of the river mouth, show similar ^{137}Cs contents (71.8 and 76.6 $Bq \cdot kg^{-1}$ dry wt.) from those at BF9 station. It is believed that these three stations are significantly affected by the Rhône River plume. At BF11 and BF3B stations, the average ^{137}Cs concentrations are 33.6 and 32.8 $Bq \cdot kg^{-1}$ dry wt., respectively, i.e. close to the concentration at BF8. Thus, the shape of the 72 $Bq \cdot kg^{-1}$ dry wt. and 34 $Bq \cdot kg^{-1}$ dry wt. isoconcentration curves are nearly the same. This distribution of the ^{137}Cs in the surficial sediments is asymmetric and therefore enables us to define the westward preferential spreading of the Rhône waters. This is related to the influence of both the Rhône River flow, upon entering the sea, drawing along river waters and carrying them southward, and of the Liguro-Provencal-Catalan current flowing southwestward along the continental shelf. The flow of this

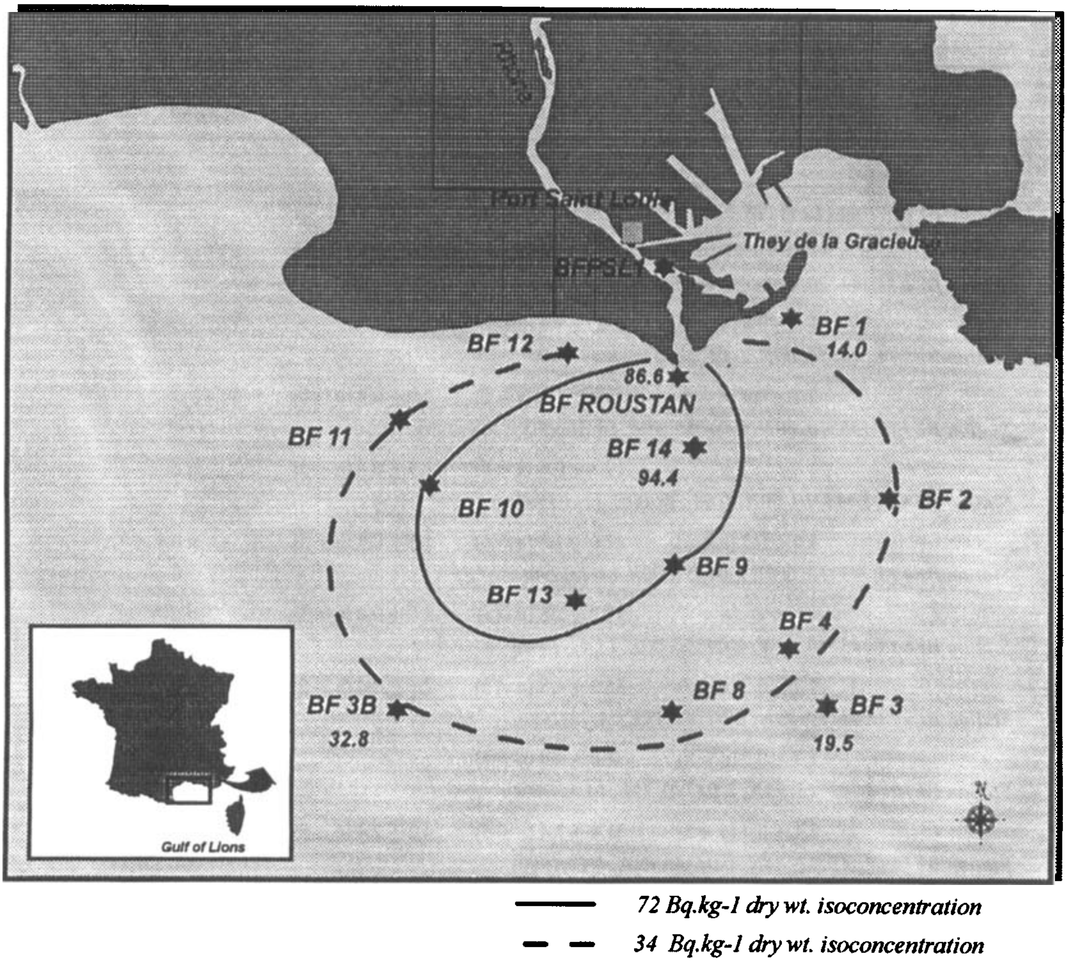

Figure 1 Stations sampled in November 1990 with a "Fluchart" box-corer and ^{137}Cs mean concentrations in the first five centimetres of the sediment.

current is driven along the coast by Coriolis forces but is modified when reaching the NE part of the continental shelf in the Gulf of Lions, where the waters above 100 m depth tend to flow over the shelf (Millot, 1990). This circulation, along with the input of the Rhône, governs sedimentation, resuspension and transport of suspended particles (Courp, 1990). Even if the granulometric profiles found at BF12 and BF1 stations, located on either side of the river mouth at 20 m depth, are similar and show the predominance of a coarse fraction ($> 96\,\mu m$) of fine sands, the preferential westward direction of the Rhône River plume is confirmed by ^{137}Cs data, respectively 33.5 and 14.8 $Bq \cdot kg^{-1}$ dry wt. (Table I). The detection of radionuclides at BF1 station can be explained by the presence of a secondary northeastward flow along the coast to They de la Gracieuse.

Table I Artificial radionuclides, mean concentrations, in the first five centimeters of sediment sampled off the Rhône mouth in the Gulf of Lions (France).

^{106}Ru	^{134}Cs	^{137}Cs	^{241}Am	Size $<4\,\mu m$ (%)
		(Bq·kg^{-1} dry wt.)		
Station BFRoustan (L:43°18, 62N; l: 04°50, 52E, 7/11/1990)				
32.2 ± 9.2 ($n = 5$)	10 ± 2.6 ($n = 5$)	86.6 ± 4.2 ($n = 5$)	1.6 ± 0.4 ($n = 5$)	13.3 ($n = 5$)
Station BF1 (L:43°20, 17N; l: 04°55, 07E, 18/11/1990)				
–	1.3 ± 0.4 ($n = 4$)	14.0 ± 1.8 ($n = 5$)	–	17.2 ($n = 4$)
Station BF2 (L:43°16, 00N; l: 04°58, 52E, 8/11/1990)				
–	2.1 ± 0.4 ($n = 5$)	33.8 ± 1.6 ($n = 5$)	2.0 ± 0.4 ($n = 5$)	32.4 ($n = 5$)
Station BF3 (L:43°10, 00N; l: 04°56, 00E, 13/3/1991)				
	–	19.5 ± 0.9 ($n = 5$)	1.3 ± 0.3 ($n = 5$)	n.d
Station BF3B (L:43°10, 00N; l: 04°39, 00E, 15/3/1991)				
–	1.4 ± 0.2 ($n = 5$)	32.8 ± 1.8 ($n = 5$)	1.5 ± 0.3 ($n = 5$)	n.d
Station BF4 (L:43°12, 00N; l: 04°54, 00E, 8/11/1990)				
–	–	37.2 ± 1.8 ($n = 5$)	2.6 ± 0.4 ($n = 5$)	n.d
Station BF8 (L:43°10, 07N; l: 04°50, 00E, 13/11/1990)				
–	1.2 ± 0.3 ($n = 5$)	35.2 ± 1.8 ($n = 5$)	2.1 ± 0.4 ($n = 5$)	n.d
Station BF9 (L:43°13, 95N; l: 04°49, 99E, 11/11/1990)				
–	1.7 ± 0.5 ($n = 5$)	71.8 ± 4.0 ($n = 5$)	–	29.6 ($n = 5$)
Station BF10 (L:43°15, 99N; l: 04°40, 79E, 14/11/1990)				
–	3.1 ± 0.4 ($n = 5$)	71.8 ± 3.4 ($n = 5$)	4.0 ± 2.4 ($n = 5$)	23.5 ($n = 5$)
Station BF11 (L:43°17, 84N; l: 04°38, 73E, 7/11/1990)				
–	1.9 ± 0.3 ($n = 5$)	33.6 ± 2 ($n = 5$)	2.3 ± 0.5 ($n = 5$)	16.2 ($n = 5$)
Station BF12 (L:43°19, 08N; l: 04°46, 05E, 12/11/1990)				
–	2.2 ± 0.3 ($n = 5$)	33.5 ± 1.8 ($n = 5$)	2.0 ± 0.3 ($n = 5$)	9.8 ($n = 5$)
Station BF13 (L:43°13, 04N; l: 04°45, 91E, 12/11/1990)				
–	2.6 ± 0.3 ($n = 5$)	76.6 ± 3.2 ($n = 5$)	3.4 ± 0.4 ($n = 5$)	33.3 ($n = 5$)
Station BF14 (L:43°18, 02N; l: 04°52, 62E, 8/11/1990)				
40.4 ± 11.8 ($n = 5$)	8.1 ± 0.5 ($n = 5$)	94.4 ± 3.8 ($n = 5$)	2.3 ± 0.4 ($n = 4$)	18.8 ($n = 5$)

Another radionuclide of value in assessing the influence of the Rhône river plume, is ^{134}Cs. This is a fission product with a half-life of 2.1 years that is released by nuclear plants in lesser amounts than ^{137}Cs. While detected at all the stations in our study, its average content does not exceed 10 Bq·kg^{-1} dry wt. As observed for ^{137}Cs, the closer the station to the river mouth, the greater the influence of the Rhône-derived sediments. With a concentration of 1.2 Bq·kg^{-1} dry wt, BF8 station marks the proximity of the southern boundary of this delta area. Later study of the vertical distribution of caesium isotopes, ^{210}Pb and of the ^{137}Cs/^{134}Cs ratio for all the stations will enable us to assess the rates of sedimentation in this high deposition rate area. The isotope ^{106}Ru is mainly released by the reprocessing plant in Marcoule. Its half life is only one year, and it is detected in the Rhône river only at BFPSL1 station and in the River mouth area at BFRoustan and BF14 stations, with average concentrations of 32.2 and 40.4 $Bq \cdot kg^{-1}$ dry wt. (Table I), respectively. These results are similar to those of Martin and Thomas (1990). This area marked with ^{106}Ru is very limited, due to the fact that ^{106}Ru is released under quickly adsorbed polymerized forms and both that its K_d is less than that of ^{137}Cs and that most of the sediments are deposited in the coastal zone. At most of the sampled stations, direct gamma ray spectrometry has also revealed the presence of the isotope ^{241}Am. This isotope is a long-lived radionuclide with a half-life of 432 years, usually measured by alpha ray spectrometry. Derived from ^{239}Pu, ^{241}Am activity also depends on the ^{241}Pu disintegration. Mainly fixed on fine particles, ^{241}Am has a very low desorption capability. It is detected at average concentrations of, respectively, 4 and 3.4 $Bq \cdot kg^{-1}$ dry wt. at BF10 and BF13 stations located within the Rhône dilution zone where sedimentation is low and the mean particle size is < 16 µm.

Among the sedimentary identification criteria, granulometry is a relevant indicator of sedimentary dynamics. Thus, the distribution of fractions (< 4 µm) within the upper 5 cm shows that the finest particles are indeed carried offshore by the surface nepheloid layer, the distance depending on both the intensity and volume of sediment transported by the Rhône River. As a matter of fact, this transport of fine particulate material to offshore areas and to the seafloor on the shelf, takes place via a multilayer system (Aloisi *et al.*, 1982) revealed in the littoral area off the Rhône River mouth. Upstream from the river mouth, where fresh and salt waters meet, electrochemical flocculation phenomena and organo-mineral aggregation lead to the formation of a turbid layer. At the river mouth, the turbid load is distributed among several nepheloid layers (Aloisi, 1986). In addition to the localized influence of the Rhône River that continuously supplies considerable amounts of particles on which radionuclides are adsorbed to the Mediterranean Sea, previous campaigns provide evidence of variations in ^{137}Cs, ^{134}Cs, ^{106}Ru contents in the first five centimetres of sediment. Study of these variations allowed us to identify atmospheric fallout related to the Chernobyl accident. Indeed, at the Roustan station which was sampled with the same box-corer in 1980, 1984, 1986 and 1990, a peak for these three radionuclides was observed during the November 1986 cruise (Table II). The observed $^{137}Cs/^{134}Cs$ ratios in the five first centimetres of these sediments chronologically are 5.2; 9.8; 2.9;

Table II ^{137}Cs, ^{134}Cs and ^{106}Ru concentrations in the first five centimetres at Roustan station for various years.

Roustan Station 0–5 cm	^{134}Cs	^{137}Cs	^{106}Ru
	($Bq \cdot kg^{-1}$ *dry wt.*)		
Nov-80	24.7 ± 3.1	129.6 ± 15.3	57.2 ± 11.2
Nov-84	16.6 ± 16.6	162.8 ± 67.5	56.5 ± 26.5
Nov-86	135.5 ± 31.8	393.8 ± 82.1	195.6 ± 53.3
Nov-90	10.0 ± 3.7	86.6 ± 23.4	32.2 ± 18.7

8.7. The $^{137}Cs/^{134}Cs$ ratio characterizing the Chernobyl accident in 1986 should be 2. Therefore the ratio found in 1986, i.e. 2.9, defined the effects of radioactive atmospheric fallouts at the river mouth from the time of the accident, derived both from direct fallouts on the Mediterranean occidental basin and from indirect fallouts by the washout of the catchment area of the Rhône basin. The other ratios are comparable with the $^{137}Cs/^{134}Cs$ ratio observed in the Rhône River waters at Fourques (close to Arles, 40 km from the river mouth) which has been varying in the range 7 to 14 since 1983, except in 1986 (2.2) (Calmet *et al.*, 1992).

CONCLUSION

These preliminary results complement a large national programme designed to study sedimentary processes in the Gulf of Lions. Within the context of this programme, other parameters have been studied, such as magnetic susceptibility, heavy metals contents, and ^{210}Pb concentration. When complemented with ^{137}Cs contents, these studies will allow a better estimate of the influence of the Rhône River in this area and a better understanding of the behaviour of rhodanian solid loads. In addition to the former samples obtained in November 1990, a deeper and more spatially intensive sampling programme was carried out in March 1991 at the river mouth using a Kullenberg corer that enabled the deeper layer of unconsolidated sediment of this area to be sampled. When all the samples are analysed, we shall be able to assess the volume of radioactive labelled sediments carried down to the continental shelf by the Rhône River since 1958, date of the first nuclear developments.

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