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^{137}Cs inventory in sediment near the Rhone mouth: role played by different sources

Inventaires de ^{137}Cs dans les sédiments à l'embouchure du Rhône: contribution des différentes sources

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Abstract

The low tidal range of the Mediterranean Sea and the high sediment load of the Rhone induce the formation of an important submarine delta. The ^{137}Cs inventory in sediment on a 480 km² area near the Rhone mouth reached 19.6 TBq in 1990. The spatial distribution of both sediment accumulation rates and ^{137}Cs concentrations in this area confirm the existence of a spatially well-delimited zone, i.e. the prodelta, where both parameters are the highest. Thus, 40% of the total amount of ^{137}Cs stored in this 480 km² area was found within the prodelta (30 km²) in the close vicinity of the river mouth. This attests to the efficiency of the ^{137}Cs trapping by the prodelta sediments. A study on the part the different ^{137}Cs sources in the Mediterranean Sea play in contributing to this inventory has been carried out. These sources are (i) direct deposition from both global fallout and the Chernobyl accident, (ii) indirect inputs arising from the erosion of surface soils of the Rhone catchment area that were also contaminated by the previous fallout, (iii) liquid effluents from the nuclear industry into the Rhone waters. Assuming that direct deposition is evenly distributed over the study area (480 km²) and that the particulate ^{137}Cs input from the Rhone is entirely trapped in this same area, these sources account for about 50% of the inventory. These hypotheses are unlikely in a coastal area subject to various physical disturbances but they are conservative enough to assess the lower limit of the nuclear liquid releases contribution to this inventory to be 50%.

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Résumé

Le faible marnage en mer Méditerranée et les apports solides élevés du Rhône contribuent à la formation d'un important delta sous-marin. L'inventaire en ^{137}Cs dans les sédiments sur une aire de 480 km² près de l'embouchure du Rhône atteignait 19,6 TBq en 1990. Les distributions spatiales des taux d'accumulation sédimentaire ainsi que des concentrations en ^{137}Cs confirment l'existence d'une zone bien délimitée, le prodelta, où ces deux paramètres sont les plus élevés. Ainsi, 40% de la quantité totale de ^{137}Cs stockée dans cette zone de 480 km² sont trouvés dans le prodelta (30 km²) au voisinage immédiat de l'embouchure. Ceci atteste de l'efficacité du piégeage du ^{137}Cs dans les sédiments du prodelta. Une étude sur la contribution à cet inventaire des différentes sources de ^{137}Cs en mer Méditerranée est réalisée. Ces sources sont (i) les dépôts directs dus aux retombées globales ainsi qu'aux retombées de l'accident de Tchernobyl, (ii) les apports indirects par le drainage du bassin versant marqué également par ces deux types de retombées, (iii) les rejets liquides des diverses installations nucléaires rhodaniennes. Si l'on suppose une distribution homogène des dépôts directs sur l'aire étudiée (480 km²) et que les apports en ^{137}Cs particulaires par le Rhône sont restés confinés dans cette même zone, ces sources contribuent pour 50% à l'inventaire. Ces hypothèses sont improbables dans une aire côtière soumise à des perturbations variées mais elles sont suffisamment majorantes pour évaluer la contribution minimale des rejets liquides des installations nucléaires à cet inventaire, à savoir 50%.

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1. Introduction

With a catchment area of about 98 800 km^2, a mean annual liquid flow of 54 billion m^3, and a mean annual solid discharge of several millions of tons, the Rhone River is one of the main sources of terrigeneous inputs to the western Mediterranean Sea.

In the Mediterranean Sea, areas of fine-grained deposits, i.e. the prodeltas, are found off the mouth of the main rivers (Aloïsi et al., 1975; Monaco, 1975; Boldrin et al., 1988; Durrieu de Madron et al., 2000). These deposits, related to river discharges, are characterized by high accumulation rates, and are thus efficient temporary storage of riverborne substances such as radionuclides (e.g. Martin and Thomas, 1990; Delfanti et al., 1997; Gulin et al., 1997; Radakovitch et al., 1999).

Caesium isotopes are known to present high affinity for clay particles in freshwater environments and a much lower affinity for particles in saline environments primarily because of competitive inhibition of sorption by other cations at high ionic strength. However, it is unlikely that a complete desorption of radiocaesium fixed onto particles within the Rhone river freshwaters occurs in the estuary before the fast and massive trapping of labelled particles in the prodeltas by flocculation and aggregation mechanisms. In addition, part of caesium cannot desorb in saline waters since it is strongly fixed in clay lattices by substituting for K+. Thus, caesium appears to be a good tracer to determine the behaviour of particle inputs from the Rhone river. Vertical distribution of caesium isotopes in sediment close to the Rhone mouth has allowed to evaluate sedimentation rates of several tenth of centimetres per year (Badie et al., 1984; Calmet and Fernandez, 1990; Charmasson et al., 1998).

In the Rhone area ^{137}Cs has several origins, i.e. atmospheric fallout from both past nuclear bomb testings and the Chernobyl accident, wash-out of the river catchment basin contaminated with these fallout, and liquid effluents from the nuclear industry. Indeed, several nuclear power plants and one spent fuel reprocessing plant are authorized to discharge low-level radioactive liquid effluents into the Rhone river system (Charmasson et al., 1994). Until 1997, year of the break in the reprocessing operations in Marcoule, the major part of the industrial releases came from this installation (Charmasson, 1998).

In this paper, the ^{137}Cs inventory at the Rhone river mouth and the relative contributions from these various sources are assessed and discussed.

2. Sampling and analyses

Sediment samples from the Rhone submarine delta were collected in thirteen locations by Fluchat box-corer and in two locations by Kullenberg corers in November 1990 and March 1991 (Fig. 1). Water depth extended from 20 m in locations near the coastline to 110 m near the shelf-break.

Cores were cut on board into 1 cm (box corer) or 10 cm sections (piston corer). Samples were then dried at 60 °C to constant weight, then homogenized and packaged into 200 or 350 cm^3 geometry to undergo a non-destructive gamma spectrometry. These analyses were performed for 24–48 h using N-type hyper pure germanium detectors with a beryllium window 0.5 mm thick. Detectors were located underground under a 3-m slab of concrete, in a room shielded with 10 cm low activity lead and 5 mm electrolytic copper. Activities expressed as Bq kg^{-1} dry weight were decay corrected to the date of sample collection.

3. Results and discussion

3.1. Inventory

In an area with a significant spatial variability in sediment deposition, such as the area near the Rhone mouth, it is difficult to interpret concentration data, since a 1-cm section of sediment core does not represent the same period of sediment accumulation and therefore of nuclear effluent integration at every site. Indeed, accumulation rates greater than 20 cm year^{-1} were reported in the prodelta area, (Calmet and Fernandez, 1990; Charmasson et al., 1998; Radakovitch et al., 1999), while on the Gulf of Lion's shelf these rates varied between 0.1 and 0.3 cm year^{-1} depending on their location with respect to the Rhone mouth (Zuo et al., 1991, 1997; Abassi, 1998; Radakovitch et al., 1999). It thus seems more appropriate to deal with inventories, i.e. with the total ^{137}Cs activity per unit surface present at a given location.

^{137}Cs inventories are presented in Table 1. Vertical ^{137}Cs profiles at these locations were reported in Charmasson et al. (1998) and Radakovitch et al. (1999). The inventories were calculated by summing the ^{137}Cs activity per unit surface in each layer of the core. Values for cores BFRous and BF14 in the prodelta area could not be used to determine accurate inventories in the corresponding locations since the thickness of the sediment column labelled by ^{137}Cs was greater than the length of the core. In these locations inventories in Kullenberg corers (KL) were used instead.

Data from Table 1 and Fig. 2 clearly demonstrate a sharp decrease in inventory with distance from the river mouth and reflect the spatial distribution previously reported in the same area for ^{137}Cs and various trace metals (Span, 1984; Fernandez et al., 1991). The total amount of ^{137}Cs stored in this area, estimated by numerical methods using Surfer® software, is 19.6 TBq. More than 90% of this amount were found within an area of 160 km^2 near the mouth and about 40% in the prodelta area which is around 30 km^2.

3.2. Contribution of different ^{137}Cs sources

Atmospheric testings of nuclear devices have been conducted between 1945 and 1980 with heavy test periods in 1952–1954, 1957–1958 and particularly 1961–1962 giving rise to what is commonly called global or worldwide fallout. Data regarding fallout deposition at the surface of the oceans
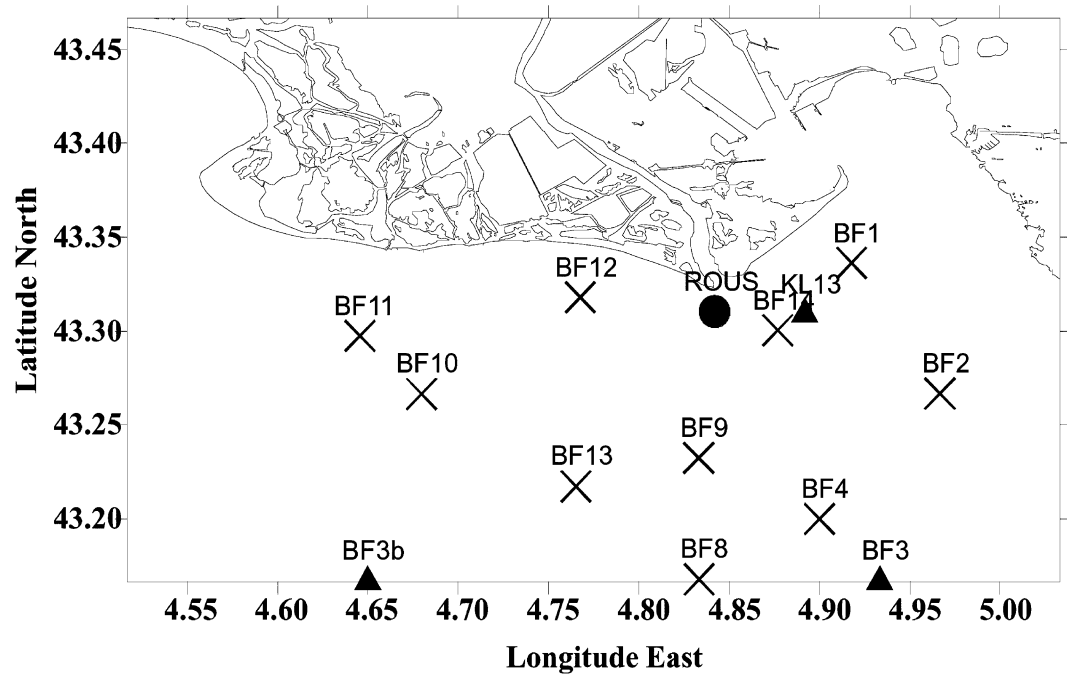
Fig. 1. Locations of the sediment sampling stations off the Rhone river mouth (north western Mediterranean Sea) in November 1990 (crosses) and March 1991 (solid triangles). BF, box-corer; KL, Kullenberg piston corer. The Roustan site (Rous) was sampled at both periods and with both corers.

Table 1
^{137}Cs inventories (kBq m^{-2}) in sediment sampled in November 1990 and March 1991 ([*]) off the Rhône mouth. BF, Fluchat box-corer; KL, Kullenberg corer

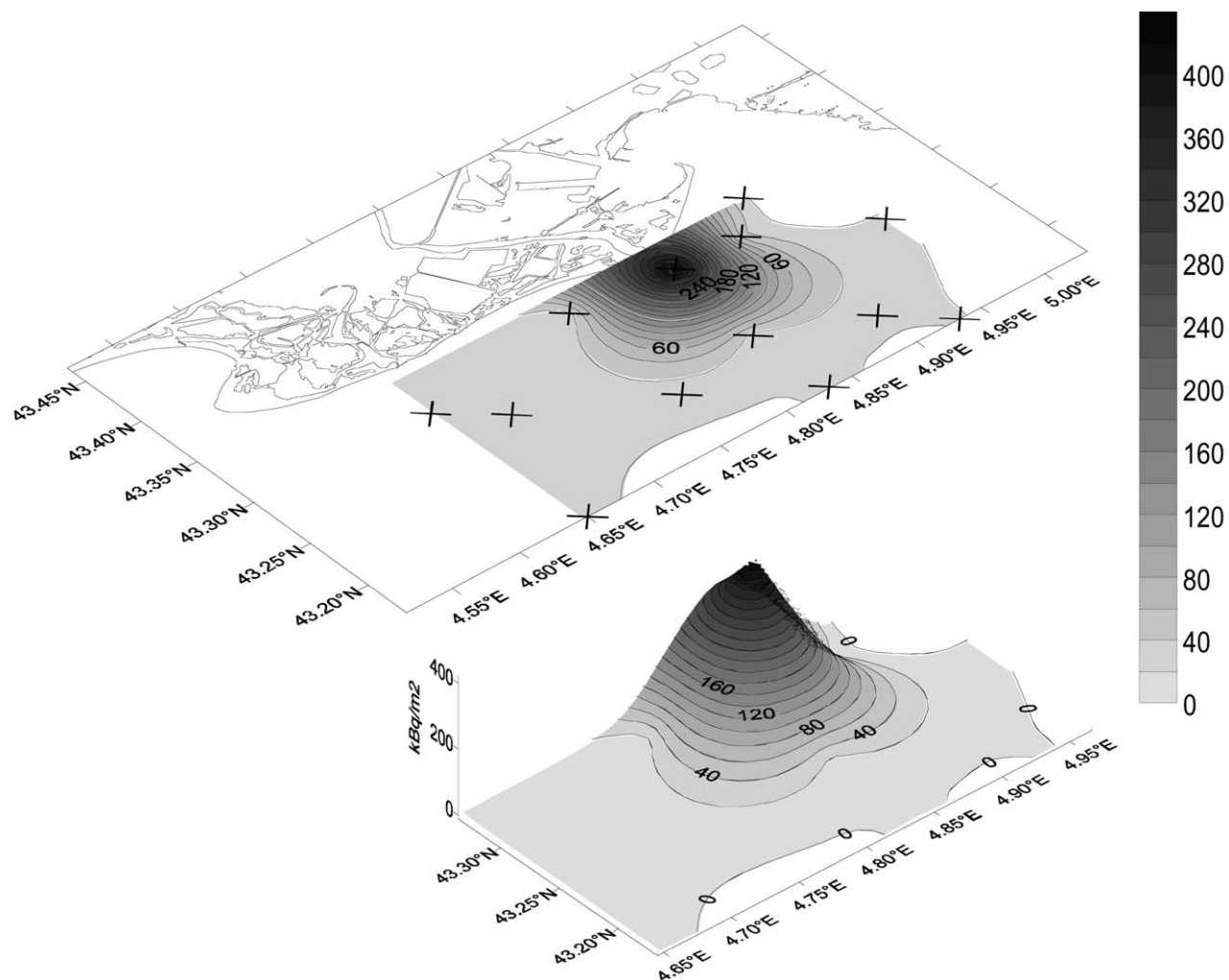| Core	^{137}Cs inventory (kBq m^{-2})	Sampling depth (m)	Accumulation rates (cm year^{-1}) from Charmasson et al. (1998) and Radakovitch et al. (1999)
BF1	1.64	20	–
BF2	2.55	95	0.36
BF4	2.69	104	0.26
BF8	3.79	102	0.29
BF9	9.17	95	0.54
BF10	7.71	56	0.65
BF11	3.96	30	0.25
BF12	8.85	23	0.25
BF13	8.92	89	0.42
BF3[*]	0.84	110	0.21
BF3b[*]	1.91	80	–
KLRous[*]	434	30	37–48
KL13[*] (east BF14)	27.33	60	–

–, Not determined.

Fig. 2. Iso-value of ^{137}Cs inventory in sediment off the Grand Rhone mouth in 1990. Kriging in 2D and 3D (*x*, latitude; *y*, longitude; *z*, inventory in kBq m^{-2}).

are not available and therefore fallout deposition over land is used instead. In order to determine the cumulative deposit of ^{137}Cs fallout over the study area, we used the ^{137}Cs annual deposition given by Thomas (1988) and derived from measurements carried out at Milford Haven and Chilton (UK). The ^{137}Cs cumulative deposit (i.e. the activity present in a specific area at a given time, which is the result of past depositions and radioactive decay) from weapon fallout is estimated to reach 2.7 kBq m^{-2} in 1990 in the Rhone area.

As regards the fallout from the Chernobyl accident (April 1986), it is difficult to find a single value characterizing the environment near the Rhone mouth due to the patchiness of the deposition pattern. UNSCEAR (1988) gives a mean value of 3.2 kBq m^{-2} in 1986 for the south eastern part of France, which has been especially contaminated. It is likely that deposition on the Rhone mouth area was lower at the time of the accident. However, the value of 3.2 kBq m^{-2} has been considered for establishing budgets because, due to the general water circulation in this area, a certain amount of ^{137}Cs has certainly been rapidly advected from the more contaminated eastern areas. Atmospheric deposition after 1986 was considered to decrease exponentially with a half-time of

about 300 d in agreement with several observations in Europe (Hotzl et al., 1989; Calmet et al., 1994; Spezzano et al., 1994). This led to a ^{137}Cs cumulative deposit due to Chernobyl of 5.1 kBq m^{-2} at the end of 1990.

Since the cores have been sampled in shallow waters, one can consider that direct deposition arising from these fallout has reached the seabed which thus should be characterized by ^{137}Cs inventories around 7.8 kBq m^{-2}. However, one must keep in mind that more than 90% of global fallout were deposited prior to 1970 and radionuclides entering the marine environment are redistributed by different physical, chemical and biological processes.

Data in Table 1 show that at stations BF1, BF2, BF3, BF3B, BF4, BF8, BF11 the inventories are lower than the value characterizing fallout from both global fallout and the Chernobyl accident (7.8 kBq m^{-2}) when inventories in BF9, BF10, BF12 and BF13 are similar or slightly higher. Only the Roustan and 14 locations present inventories well in excess. This distribution among the cores reflects mainly the Rhone inputs and it is obvious that most of the cores presenting inventories in excess of fallout values are located in areas with a strong Rhone influence.
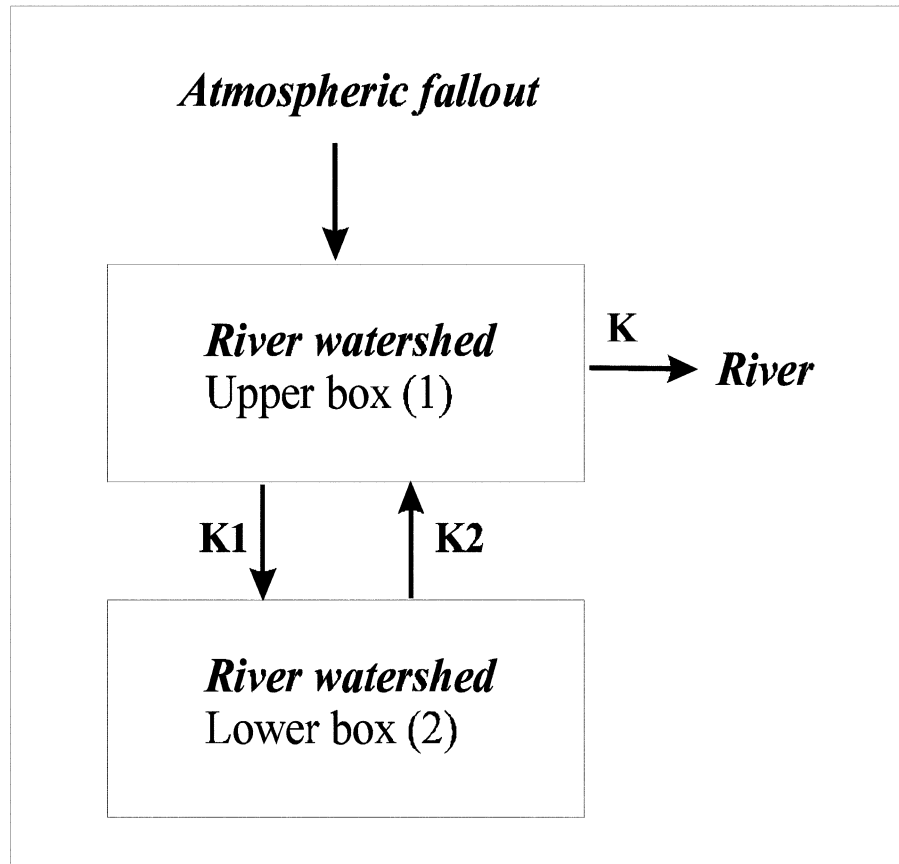
Fig. 3. Two-box model of the Rhone river watershed from Thomas (1988).

In addition to direct fallout deposition, this area received radionuclides transported from land to sea by river runoff. Indeed, erosion of surface soils from the Rhone watershed, which also received fallout from atmospheric weapon testings and the Chernobyl accident, has led to ^{137}Cs delivery to the Rhone waters and therefore the Mediterranean Sea. For determining these inputs, it would be necessary to have ^{137}Cs temporal series on both dissolved and particulate phases in the Rhone waters since the beginning of atmospheric nuclear testings. Such data are lacking and a model presented by Thomas (1988) was used to determine the ^{137}Cs output from the Rhone watershed over the period 1954–1990. Considering the Rhone watershed as a black box, this model links quantitatively the outputs to the inputs without describing the processes governing the behaviour and distribution of ^{137}Cs within the soil. This model considers two soil subsystems in the watershed (Fig. 3). One upper box (box 1) receives the atmospheric fallout and is the source of ^{137}Cs (both particulate and dissolved) to the river, the lower box (box 2) receives ^{137}Cs by exchanges with the previous box. Exchanges between these two boxes follow a first-order kinetic, i.e. exchanges are proportional to the amount accumulated within the box. The output flux (particulate and dissolved) is given by $K * M_1$ (M_1 being the content of the upper box in GBq km^{-2} year^{-1}). The particulate flux is then derived by taking into account this output flux, the surface of the watershed (km^2) and the particulate fraction of the riverine transport.

This latter parameter is calculated from ^{137}Cs Kd value and both solid and water discharges of the Rhone river. Full details on the model are given in Thomas (1988, 1997).

In our case, atmospheric inputs are annual ^{137}Cs fallout deposition (from weapon testings and the Chernobyl accident) on the Rhone watershed from 1954 to 1990 (see above).

Thomas (1997) gives for the Rhone system the following values: 0.0045 for K (the fraction of box 1 content that escapes to the river), 0.60 for K_1 (the fraction from box 1 that is transferred to box 2) and 0.042 for K_2 (the fraction from box 2 that is transferred to box 1) These parameters show that fallout-derived radionuclides are rapidly stored within the watershed soil, which thus represents an important reservoir, of which the annual output towards the river, and thus the coastal zone of the Mediterranean Sea, is low.

Taking into account the ^{137}Cs partition coefficient within the Rhone river (Kd 30 800 given by Thomas, 1997), the Rhone watershed surface (98 800 km^2) and the ratio solid discharge/liquid flow (3.6×10^6 t year^{-1}/5.34×10^{10} m^3 year^{-1}), the amount of particulate ^{137}Cs transported by the river from the catchment area to the Mediterranean Sea from 1954 to 1990 was estimated to reach 5.6 TBq (amount decay corrected to 1990).

Assuming that this amount is entirely trapped in the study area (480 km^2) and that direct deposition from global fallout and the Chernobyl accident are evenly distributed over the same area (3.7 TBq; 7.8 kBq m^{-2} * 480 km^2), these sources

account for about 50% of the ^{137}Cs accumulated in the delta sediment whose total inventory is about 19.6 TBq. These assumptions about ^{137}Cs distribution are unlikely in a coastal area subject to different disturbances (hydrodynamical, sedimentary, and biological). However, they are conservative enough to allow the determination of the lower limit of the nuclear liquid discharges contribution to this inventory. This contribution is thus at least 50% (i.e. 10 TBq). This amount represents about 20% of the cumulative discharges by the various nuclear installations between 1961 and 1990 in the Rhone river. Indeed, releases from nuclear power plants and the Marcoule reprocessing plant reached 51.4 TBq (amount decay corrected to 1990).

4. Conclusions

This work represents an attempt to determine the relative importance of a number of potential sources to the ^{137}Cs inventory in marine sediments in the area under the influence of the Rhone river. This inventory is certainly to be refined since it is based on few samples, particularly in the prodelta area. However, this study shows that the sediment compartment is of prime importance in the frame of research on the dispersion of contaminants introduced in the Mediterranean Sea by the Rhone river.

Even if sediment appears as an effective and important sink for particle-reactive elements transported by rivers, this storage is partly transitory. Indeed, storms induce resuspension phenomena, especially within the prodeltaic areas, which feed the benthic nepheloid layer (Courp and Monaco, 1990; Delfanti et al., 1997). Besides, high flow periods of the Rhone river appear to be at the origin of strong disturbances in sediment records as it has been underlined for the 1994 floods (Charmasson, 1998) preventing us from having a clear relationship between ^{137}Cs vertical profile and its rate of supply from the various sources.

In addition, the fact that reprocessing operations of spent nuclear fuel ceased in 1997 at Marcoule represents a strong signal and it is of high interest to follow the environmental response to such an event. Indeed, desorption of ^{137}Cs from sediment towards the water column has been observed in the Irish Sea after an important decrease in the releases from the Sellafield reprocessing plant (Poole et al., 1997).

It thus appears especially interesting to follow the temporal evolution of ^{137}Cs stocks at the Rhone river mouth and to study on one hand the chemical remobilization processes mainly to be related to the break in the reprocessing of spent fuel in Marcoule and on the other hand the physical remobilization processes due to resuspension phenomena upon natural (storms, floods, etc.) or man-induced (dredging, fishing, etc.) activities. Several programmes (both national and international) are currently dealing with these aspects in the Gulf of Lion. They should improve our knowledge and allow the quantifying of these processes in this area.

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