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**Importance of colloids in the transport within the dissolved phase (<450nm)
of artificial radionuclides from the Rhône River towards the Gulf of Lions
(Mediterranean Sea)**

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ABSTRACT

The significance of colloidal fractions regarding the transport of artificial radionuclides in natural water systems is underlined by using sequential ultrafiltration both in the Rhône freshwater and the marine area under and outside the influence of the river outflow. Indeed, the Rhodanian aquatic system represents an interesting test site as various artificial radionuclides are released into the Rhône River by several nuclear installations. We focused our study on ¹³⁷Cs, ¹⁰⁶Ru, ⁶⁰Co, ²³⁸Pu and ²³⁹⁺²⁴⁰Pu. Our results show that Fe, Al and Organic Carbon (OC) are the main components of colloidal matter. Colloids represent about 15% of dissolved (<450 nm) OC and 25% of dissolved Fe and Al exported towards the sea. Within the dissolved (< 450 nm) phase, these colloidal compounds are shown to account for the transport of 40% for both Co and Ru, 60% for ²³⁸Pu and ²³⁹⁺²⁴⁰Pu and have no significance on ¹³⁷Cs flux.

KEY WORDS: colloids, artificial radionuclides, speciation, Rhône River, estuary, Mediterranean Sea.

INTRODUCTION

Natural waters represent key systems regarding the exportation of numerous elements from the continental area to the ocean. The interactions with suspended and colloidal matter within these systems heavily influence the mobility and redistribution of different elements (Stumm and Morgan, 1981). The colloidal fraction may affect the behaviour of the metals in two ways. In the river, the colloidal fraction may enhance transport by limiting the free ions available for adsorption to solid phases either on the river bed or suspended in the water. On the other hand, metal removal in the low salinity region of the estuary is known to be driven by flocculation of part of the colloidal pool. Besides, trace elements bound in colloidal phases may be unavailable to biological uptake (Rich and Morel 1990; Wells and Mayer 1991), limiting the transport of potentially toxic metals up the aquatic food web. Regarding artificial radionuclides, whilst various studies have investigated their distribution among particulate and dissolved phases, very few studies have been devoted to their transport by colloids with an exception for plutonium isotopes (Nelson et al. 1985; Mitchell et al. 1995; Eyrolle et al 1999, Dai et al. 2001).

The presence of artificial radionuclides in the Rhône River is mainly associated with controlled radioactive releases from several nuclear facilities along the river and its tributaries, but is also linked with other sources including past atmospheric nuclear tests and the Chernobyl fallout. Until 1997, the major part of the industrial releases came from the Marcoule reprocessing plant (Charmasson, 1998). Several research programmes aim at a better understanding of the behavior of riverine inputs and associated contaminants in the Gulf of Lions. In this frame, new dissolved and colloidal phase data for the distribution of ^{137}Cs , ^{106}Ru , ^{60}Co , ^{238}Pu and $^{239+240}\text{Pu}$ from the Rhône River to the marine environment are reported and discussed in this paper by integrating the results reported in Eyrolle et al. (1999) and Eyrolle and Charmasson (2001).

The main objectives of the study were:

- (1) to determine the occurrence and behaviour of colloids (both organic and inorganic) during the freshwater and sea water mixing.

(2) to determine the role of colloids in the transport of artificial radionuclides from the Rhône River towards the Gulf of Lions

This was achieved using a large volume ultrafiltration system as high concentration factors are required for the detection of activities ranging from some mBq m⁻³ for plutonium isotopes to some Bq m⁻³ for ¹³⁷Cs, ¹⁰⁶Ru, ⁶⁰Co within the Rhône aquatic system (Calmet et al 1988; Martin and Thomas; 1990; Charmasson, 1998).

MATERIALS AND METHODS

The study area

Downstream from the city of Arles, the Rhône River divides into two branches, the Grand Rhône and the Petit Rhône, which flow across a large delta (1750 km²) (Figure 1). The former branch drains 90% of the water discharge and is 52 km long whereas the second one carries 10% of the discharge, is 60 km long and is characterized by a meandering laying out. The mixing zone is highly stratified, with a deep salt wedge intrusion into the lower river course and a thin unstable surface plume extending several kilometers seaward dependent on wind stress and river flow rate. Guieu et al. (1991) have estimated that most of the particulate Rhône input settles on a relatively small area close to the river mouth. About two-thirds of the solid discharge accumulate over an area of 4000 km², representing only 17% of the total surface of the continental shelf in the Gulf of Lions, and a large fraction of the remaining input does not significantly leave the shelf area (Monaco et al., 1990). The Rhône is the major River entering the Western Mediterranean Sea and is therefore the main source of sediments and freshwater to this area, with an average water discharge of 1700 m³ s⁻¹ and an annual solid discharge fluctuating between 2.6 and 26.5 million tonnes (Antonelli, 2002).

Sampling and analytical methods

Large water samples (1000 l) were collected from 1996 to 1997 during several cruises at the lower course of the Grand Rhône under various river flow rates, suspended particulate matter (SPM)

concentrations and salinities (*Table 1*). The Rhône River flow rates were recorded in Arles by the Rhône River National Company. The SPM concentrations were measured by standard gravimetry.

Samples were filtered sequentially using 1200 nm and 450 nm cellulose acetate cartridges during collection. Filters with 450 nm pore size are traditionally used to separate the so-called ‘particulate’ phase from the ‘dissolved’ phase (Buffle et al. 1992). These samples were then poisoned with NaN_3 (0.1 g.l^{-1}) and subsequently sequentially ultrafiltered through membranes with size/molecular weight cut-off of 200 nm, 100 000 Da, 20 000 Da and 5 000 Da. No specific calibration for molecular cut-off was done but referring to the manufacturer calibrations we may assume that the 5000 Da membrane retain colloids over 2 nm. All samples were collected in 150 l low density polyethylene containers previously washed 3 times with 10 l of the sample which were discarded. Each ultrafiltration step was proceeded until the retentate volume reached around 5 l. The ratio permeate/retentate was then about 250. The ultrafiltration system, processing procedures, cleaning and handling protocols are reported in detail in Eyrolle and Charmasson (2000 and 2001).

Organic carbon (OC) measurements were performed with a SHIMAZU TOC 5000 analyser. Inorganic element analyses were performed by Atomic Absorption Spectrophotometry or Inductively Coupled Plasma Mass Spectrometry. γ ray emitters (^{137}Cs , ^{60}Co and ^{106}Ru) were concentrated using batch co-precipitation (adsorption) with MnO_2 -KCFC carrier (Helfman and Kalmikov, 1962) and analysed by direct gamma spectrometry with Germanium HP detectors (Bouisset and Calmet, 1997). Plutonium contents were determined after co-precipitation with $\text{Fe}(\text{OH})_3$ and using radiochemical procedures by solvent extraction and/or ionic exchange. Upon radiochemical separation, plutonium alpha sources were prepared by electrolytic plating onto stainless-steel discs. The determination of ^{238}Pu and $^{239+240}\text{Pu}$ activities were achieved as described in Goutelard et al. (1999).

Mass balances or recoveries were calculated according to the expressions reported by Eyrolle et al. (1996). The relative uncertainties are 1.5% for organic carbon concentrations, 1% for volumes, 2% for Mg, 4% for Ca, 5% for Cu and Zn, 10% for Fe, Si and Al, 9% for $^{239+240}\text{Pu}$ and 20% for the other radionuclides.

RESULTS AND DISCUSSION

Ultrafiltration mass recoveries

During fractionation experiments cleaning and handling protocols are critical (Buesseler, 1996). Mass balance provides a first-order check on sorptive losses or contaminations (Carlson et al., 1985; Buesseler, 1996; Eyrolle et al., 1996). If bad recoveries are obtained, no conclusions can be drawn since losses or contaminations during the ultrafiltration experiment may not affect the whole size distribution. This especially concerns the artificial radionuclides occurring at very low levels. Generally, mass recoveries obtained show no significant losses or contaminations during ultrafiltration runs with an exception for Fe at Station 4 (*Table 2*) where the mass recovery is 71 ± 14 %. These results indicate an accurate fractionation of samples. Both the cleaning/conditioning steps and the large volumes used contributed to successful results.

OC, Fe, Al, Mn and Cu concentrations

In the Rhône River (i.e. stations 1 to 3), Fe and Al concentrations in the dissolved phase (<450 nm) seem to be partially flow rate dependent while OC and Si do not show clear trend with respect to this parameter. Cauwet et al. (1990) did not observed a correlation between dissolved organic carbon (DOC) and the Rhône River flow and reported values ranging from 1800 to 3000 $\mu\text{g l}^{-1}$. As underlined by Guieu et al. (1997) dissolved trace metals concentrations in the Rhône River vary greatly and these trends might not be significant owing to the very limited number of data. However the increase in Al concentrations with increasing flow rate can be linked to an increase in clay mineral content during high flow rates.

Al, Fe, Si and DOC do demonstrate a decrease with increasing salinities in the Rhône plume (i.e. station 4 excepted). Chou and Wollast (1997) also reported such a distribution for both Al and Si in the Rhône plume which they link to a simple dilution of the river components by seawater without any significant removal or production during the mixing process. However, their Al dissolved values are

far lower since they are in the range 10.8-16.2 $\mu\text{g l}^{-1}$ in the river freshwater (psu=0, river flow rates not given). Regarding Fe, Guieu et al. (1997) mentioned a mean dissolved value nearing 10 $\mu\text{g l}^{-1}$ for the Rhône River which is the same order of magnitude reported in *Table 2*.

The increase Fe concentrations near the bottom at station 5 (buoy Roustan) might be due to a release from the underlying sediment pore water but can also be linked to the presence of the bottom nepheloid layer. At the same location Cauwet et al. (1990) reported an increase in DOC in samples collected close to the sea bottom and characterized by high turbidity values. These authors also mentioned very high DOC concentrations in the interstitial waters (up to 30 mg l^{-1}).

In the open sea (i.e. station 4) we observed very high contents for both Fe and Al in the surface waters. Despite the fact that we have only one sample, we have to underline that is well known that Mediterranean waters demonstrate high Al content. In the Gulf of Lions, dissolved Al and Fe find their origin in various sources : the riverine inputs and the atmospheric inputs (Guieu et al., 1991), these latter having two main components in this area i.e. an anthropogenic one and a saharian one (Chester et al., 1993; Guieu et al., 1997). Though highly variable, Al and Fe atmospheric inputs are quite high over the Gulf of Lions since Guieu et al. (1997) measured total deposition fluxes reaching respectively 918 (± 734) and 840 (± 600) $\text{kg km}^{-2} \text{ year}^{-1}$. However, the Al value reported on *Table 2* for station 4 is quite high (115 $\mu\text{g l}^{-1}$) compared to usually characterizing Mediterranean water values, i.e. concentrations in the range 0.5 to 4 $\mu\text{g l}^{-1}$ (Chou and Wollast, 1997).

Organic and inorganic colloids

In the Rhône River freshwaters 15% of the DOC is in the form of colloids. At the freshwater-sea water mixing zone a low amount of colloidal organic carbon (4%) was observed for the low salinity waters (station 6) while amounts from 31 to 41% were observed in higher salinity waters. These results are in agreement with those obtained by Dai et al. (1995) as these authors observed that colloidal organic carbon contributes 8-30% of the DOC with a maximum value in the River water.

The offshore seawater sample, station 4, shows little fractionation of DOC with 93% in the true dissolved phase. This finding is consistent with the small amount of high molecular weight material

reported in a compilation of ultrafiltration results from offshore waters (Carlson et al., 1985; Whitehouse et al. 1990).

The amounts of colloidal manganese were less than 7% at any location while the proportions of Fe and Al observed in the colloidal phase reached 42 and 35% within the superficial waters of the mixing area, respectively. These data underscore the widespread importance of Fe oxy-hydroxide/organic colloids, which have long been recognized as the dominant Fe phase in rivers and estuaries (Boyle and Edmond, 1977; Fox, 1988). The colloidal nature of Al is expected as this metal is present in natural aquatic systems both as clays and organic complexes that extend to the colloidal size range (Stumm, 1992). The relatively low proportions of colloidal Fe, Al and OC at station 6 is rather unexpected. Indeed, significant amounts of colloidal material were found at stations 1 and 3 characterized by similar SPM contents and river flow rates. This may first reflect a variability of the colloidal compound pool since sampling was not done simultaneously. This may also reflect a rapid removal of the colloidal materials through flocculation-coagulation processes within the low salinity gradient. Nevertheless, our data do not let us really confirm on this last hypothesis.

Mn exhibits smaller colloidal proportions than Fe and Al and no significant variation among locations. This is consistent with Mn speciation, which favours the free ionic form or small complexes in freshwaters (Sigg 1987). The amounts of colloidal Mn observed in the Rhône system agree with the 5% colloidal Mn found in another French River, the Garonne (Kraepiel et al. 1997).

Within the dissolved phase, Cu is expected to be found in the colloidal size range as it is a strong 'particle' reactive element due to its high affinity with most naturally occurring organic and inorganic ligands. Our data indicate that 4-21% are in the colloidal size range, with the lowest values for the most depleted colloidal Fe-Al-OC samples, i.e. stations 4 and 6.

No clear relation between organic and inorganic colloid amounts and flow rates has been observed as it might be expected regarding the range of SPM contents. This may be partly explained by the variability of SPM and colloid nature and origin as data have been acquired over a large time scale integrating obvious temporal variations.

Our results underline that OC, Fe and Al may represent an important pool of trace metal colloidal reactive compounds in the Rhône system. Unfortunately, our limited number of data do not easily allow to argue on their behaviour at the freshwater-seawater mixing zone.

Radionuclide activities

In spite of the large volume of water fractionated by ultrafiltration, only ^{137}Cs and $^{239+240}\text{Pu}$ were detected in all the samples in which they have been analysed (no Pu analyses at stations 1 and 2).

In the studied area, ^{137}Cs and plutonium isotopes originate both from the weathering of the catchment basin contaminated by atmospheric fallout (from past nuclear tests and the Chernobyl accident) and from the liquid effluents released from the Marcoule reprocessing plant. Regarding the plutonium isotopes the two sources are characterized by very different $^{238}\text{Pu}/^{239+240}\text{Pu}$ activity ratios (AR). Today, the draining waters from the Rhône basin demonstrate ratio bordering 0.025 while Marcoule releases are characterized by a theoretical constant ratio nearing 0.3, i.e. at least ten times higher (Charmasson, 1998). Both ^{106}Ru and ^{60}Co characterize the Marcoule reprocessing plant liquid releases and are thus submitted to dilution processes either by the watershed inputs or the marine waters.

Due to variability both in the releases from the Marcoule plant and the Rhône River flow rate, the radionuclide concentrations at the various locations cannot be compared since the sampling took place at different periods. Nevertheless, we can draw broad trends. Thus, ^{106}Ru and ^{60}Co were only detected in the Rhône River under moderated flow rates and for low salinity waters, i.e. for low dilution conditions (preventing us to follow their speciation in the plume). Dissolved ^{137}Cs concentrations in the river appear also to be clearly flow rate dependent with low activity when the flow rate is high. This is consistent with the results reported by Charmasson (1998). Regarding plutonium, $^{238}\text{Pu}/^{239+240}\text{Pu}$ activity ratios in the river (AR= 0.41 at station 3) clearly demonstrate the influence of the Marcoule discharges while at station 4 in the open sea (AR=0.026) we have clearly the signature of an input from atmospheric fallout

Radionuclide partitioning

Regarding radionuclide transport by the colloidal fractions, it is generally accepted that caesium does not have a tendency to undergo complexation reactions but behaves predominantly as a discrete ionic species. Indeed this element is known to compete with potassium ions regarding its sorption on clay material. However, Hilton et al. (1988) have observed Chernobyl-derived ^{137}Cs within the colloidal size range in surface lake. We did not observe such a behaviour within the Rhône system. This may be due to low proportion and/or bad ^{137}Cs scavenging efficiency of the colloidal clay material drained by the Rhône River.

Contrarily to caesium, significant proportions of ^{106}Ru , ^{60}Co and plutonium isotopes were observed in the colloidal phases when detected. Indeed, in the river from 31 to 40% of ^{106}Ru and ^{60}Co in the dissolved phase were bound to colloids, this proportion was even greater for plutonium since it reaches 59 to 62% at station 3. (at station 6, no significant amount of ^{106}Ru , ^{60}Co or plutonium isotopes within the colloidal size range were observed which is consistent with respect to the very low amounts of colloidal OC-Fe and Al available for adsorption or complexation reactions at this location).

Regarding ^{106}Ru and ^{60}Co , interactions with colloidal compounds from field experiments have been poorly reported in the literature with some exceptions for liquid radioactive wastes (Leonard et al., 1995; Salbu et al., 1987) or ground waters (Buddemeier and Hunt, 1988).

Our results underscore that colloidal compounds in the Rhône waters contribute to the transport of a significant amount of dissolved radioactive cobalt, ruthenium and plutonium isotopes, thus enhancing the transfer efficiency within the River and limiting the amounts of free ions available for biological uptake or for adsorption to solid phases either on the river bed or suspended in the water. Unfortunately, our data do not let us to argue on the behaviour of the radionuclides bound to colloids when entering in the mixing area, except to some extent for plutonium.

At any location, the amounts of plutonium observed in the colloidal phase are similar for $^{239+240}\text{Pu}$ and ^{238}Pu , thus underlying there is no significant isotopic fractionation related to ultrafiltration processing as expected and related elsewhere (Dai et al., 2001). The highest proportions of colloidal plutonium are found in the river (59-62%). At the mouth of the river 17% of dissolved plutonium is still found in

the colloidal phase in the surface plume when this proportion reaches 41% in the bottom water sample. These values are significantly higher compared to what is reported for coastal or open sea waters by Dai et al. (2001) and Mitchell et al. (1995). The $^{238}\text{Pu}/^{239+240}\text{Pu}$ ratios in the river clearly demonstrate the influence of the Marcoule discharges as the observed activity ratio is close to the 0.3 while at station 4, in the open sea, we have clearly the signature of an input from atmospheric fallout. Within the mixing zone, the $^{238}\text{Pu}/^{239+240}\text{Pu}$ ratio is only available for bottom waters. In this last sample (station 5F) the activity ratio demonstrates a small contribution from industrial discharges. Owing to the ratio obtained it appears that only 3 to 18% of the Pu originate from the Marcoule discharges. The salinity indicates an even lower river contribution indicating the Mediterranean as the main Pu source in this sample. This can be linked to particle-bound plutonium released into the bottom nepheloid layer following the degradation of such particles and/or to an input via the underlying pore-water sediment partly contaminated by the Marcoule discharges. We are not aware of any data concerning Pu concentration in the pore water near the Rhône mouth. The potential for geochemical remobilisation of Pu from the sediment in response to various changes (redox, microbiological activity, ...) has been addressed periodically without any clear conclusion about the importance of this phenomenon or about the main driving factors (e.g. Sholkovitz, 1983; Malcolm et al., 1990; Morris et al., 2001). However, in the open sea the increase of dissolved Pu concentrations near the bottom has been widely reported (e.g. Nelson et al., 1984; Cochran et al., 1987; Mitchell et al., 1995).

CONCLUSIONS

The lack of size partitioning data concerning artificial radionuclides in the literature is primarily associated with the extremely low concentrations generally encountered in the field and even lower concentrations in the fractionated samples. Due to this constraint our data set is very limited. This point is especially critical in the Rhône River system which is characterized by a large variability (Martin et Thomas, 1990; Guieu et al., 1997).

In our samples, colloidal populations in the Rhône system are mainly represented by OC, Fe and Al. These compounds may then represent an important pool for trace element sorption and greatly

influence the behaviour of trace elements either in river or at the mixing zone. Our data demonstrate that within the dissolved phase (< 450 nm) colloidal compounds in the Rhône contribute to the transport from 40 to 60% of radioactive cobalt, ruthenium and plutonium isotopes. However, as underlined by Martin and Thomas (1990), the suspended mater is the major vector for most of the radionuclides discharged into the Rhône River system. Indeed, in the River freshwaters our data (not given) indicate that up to 86% of ^{137}Cs , 43% of ^{106}Ru , 54% of ^{60}Co and 85% of Pu isotopes were bound to particles (> 450 nm).

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TABLE AND FIGURE CAPTIONS

Table 1: Sample locations and characteristics.

Table 2: Concentrations ($\mu\text{g l}^{-1}$) or activities (Bq m^{-3}) of organic carbon (OC), stable and artificial elements in the dissolved phase ($< 450 \text{ nm}$), distributions (%) between the 'true' dissolved fraction ($< 2 \text{ nm}$) and the colloidal one ($2\text{-}450 \text{ nm}$) and mass balance recoveries (%) after fractionation runs. DL: Detection Limit, NM: not measured; ND: Not Determined; NP: partition Not Possible due to a bad mass recovery; RU_c : relative uncertainty on concentrations or activities; RU_r : Relative Uncertainty on distributions and RU_m : relative uncertainty on mass recoveries.

Figure 1: Sampling map.

Station	Date of collection	location	Depth of sampling/total depth (m)	Rhône flow rate (m ³ s ⁻¹)	SPM (mg l ⁻¹)	Salinity (PSU)	Radionuclides analyses	references
1	09-21-1996	Rhône River; Arles 43°40,83N 04°36,50E	0.5/2	800	5.52	0	Gamma spectrometry	Eyrolle and Charmasson 2000; Eyrolle and Charmasson 2001
2	12-02-1996	Rhône River; Esquineau	0.5/5	4500	140.10	0	Gamma spectrometry	Eyrolle and Charmasson 2000
3	05-13-1997	Rhône River; Arles 43°40,83N 04°36,50E	0.5/2	1700	14.25	0	Gamma spectrometry, ²³⁸ Pu and ²³⁹⁺²⁴⁰ Pu	Eyrolle and Charmasson 2000; Eyrolle et al. 1999
4	04-10-1997	Golf of Lion Off shore	10/-	-	0.89	37.332	Gamma spectrometry	Eyrolle and Charmasson 2000
5S	09-09-1997	43°18,788N 04°51,250E	2/29	1050	1.13	31.309	Gamma spectrometry, ²³⁸ Pu and ²³⁹⁺²⁴⁰ Pu	-
5F	09-09-1997	43°18,788N 04°51,250E	25/29	1050	0.99	37.918	Gamma spectrometry, ²³⁸ Pu and ²³⁹⁺²⁴⁰ Pu	-
6	09-14-1997	43°22,96N 04°48,60E	3.5/4.0	1230	50.23	3.956	Gamma spectrometry, ²³⁸ Pu and ²³⁹⁺²⁴⁰ Pu	-

Table 1

	OC	Mn	Fe	Al	Cu	¹³⁷ Cs	¹⁰⁶ Ru	⁶⁰ Co	²³⁸ Pu	²³⁹⁺²⁴⁰ Pu	
1	Conc. <450 nm ($\mu\text{g l}^{-1}$ or Bq m^{-3})	2604	NM	4	18	2.5	4.9	23	1	ND	ND
	% True dissolved	83	NM	77	82	81	91	60	61	ND	ND
	% Colloidal	17	NM	23	18	19	9	40	39	ND	ND
	<i>Mass Recovery (%)</i>	<i>91</i>	<i>NM</i>	<i>104</i>	<i>87</i>	<i>91</i>	<i>NM</i>	<i>NM</i>	<i>NM</i>	<i>NM</i>	<i>NM</i>
3	Conc. <450 nm ($\mu\text{g l}^{-1}$ or Bq m^{-3})	2200	4.0	17	65	2	0.6	43	0.16	0.00272	0.00667
	% True dissolved	88	93	92	99	89	99	69	62	38	41
	% Colloidal	12	7	8	1	11	1	31	38	62	59
	<i>Mass Recovery (%)</i>	<i>100</i>	<i>85</i>	<i>108</i>	<i>85</i>	<i>101</i>	<i>100</i>	<i>102</i>	<i>88</i>	<i>98</i>	<i>107</i>
2	Conc. <450 nm ($\mu\text{g l}^{-1}$ or Bq m^{-3})	3510	6.1	14.1	82.5	2.4	0.32	<DL	<DL	ND	ND
	% True dissolved	85	99	75	71	90	<DL	<DL	<DL	ND	ND
	% Colloidal	15	1	25	29	10	<DL	<DL	<DL	ND	ND
	<i>Mass Recovery (%)</i>	<i>103</i>	<i>86</i>	<i>87</i>	<i>102</i>	<i>101</i>	<i>NM</i>	<i>NM</i>	<i>NM</i>	<i>NM</i>	<i>NM</i>
6	Conc. <450 nm ($\mu\text{g l}^{-1}$ or Bq m^{-3})	889	282	24	17	11.6	0.9	6.9	0.29	0.00301	0.00626
	% True dissolved	96	96	100	91	96	100	98	97	92	100
	% Colloidal	4	4	0	9	4	0	2	3	8	0
	<i>Mass Recovery (%)</i>	<i>96</i>	<i>98</i>	<i>105</i>	<i>91</i>	<i>94</i>	<i>98</i>	<i>96</i>	<i>88</i>	<i>102</i>	<i>109</i>
5S	Conc. <450 nm ($\mu\text{g l}^{-1}$ or Bq m^{-3})	628	56	34	10	2.5	1.85	<DL	<DL	<DL	0.00992
	% True dissolved	60	95	58	65	86	100	<DL	<DL	<DL	83
	% Colloidal	40	5	42	35	14	0	<DL	<DL	<DL	17
	<i>Mass Recovery (%)</i>	<i>89</i>	<i>95</i>	<i>93</i>	<i>103</i>	<i>107</i>	<i>106</i>	<i>NM</i>	<i>NM</i>	<i>NM</i>	<i>98</i>
5 F	Conc. <450 nm ($\mu\text{g l}^{-1}$ or Bq m^{-3})	178	19	225	4	1.4	2.4	<DL	<DL	0.00074	0.01
	% True dissolved	69	96	77	87	79	100	<DL	<DL	62	59
	% Colloidal	31	4	23	13	21	0	<DL	<DL	38	41
	<i>Mass Recovery (%)</i>	<i>95</i>	<i>97</i>	<i>102</i>	<i>98</i>	<i>106</i>	<i>86</i>	<i>NM</i>	<i>NM</i>	<i>92</i>	<i>103</i>
4	Conc. <450 nm ($\mu\text{g l}^{-1}$ or Bq m^{-3})	1010	6.2	163	115	1	2.3	<DL	<DL	0.00018	0.007
	% True dissolved	93	98	NP	95	94	93	<DL	<DL	NM	NM
	% Colloidal	7	2	NP	5	6	7	<DL	<DL	NM	NM
	<i>Mass Recovery (%)</i>	<i>87</i>	<i>87</i>	<i>71</i>	<i>81</i>	<i>88</i>	<i>96</i>	<i>NM</i>	<i>NM</i>	<i>NM</i>	<i>NM</i>
RU_c (%)	2	10	10	10	10	12	12	13	9	9	
RU_d or RU_m (%)	4	20	20	20	20	20	20	20	20	20	

Table 2

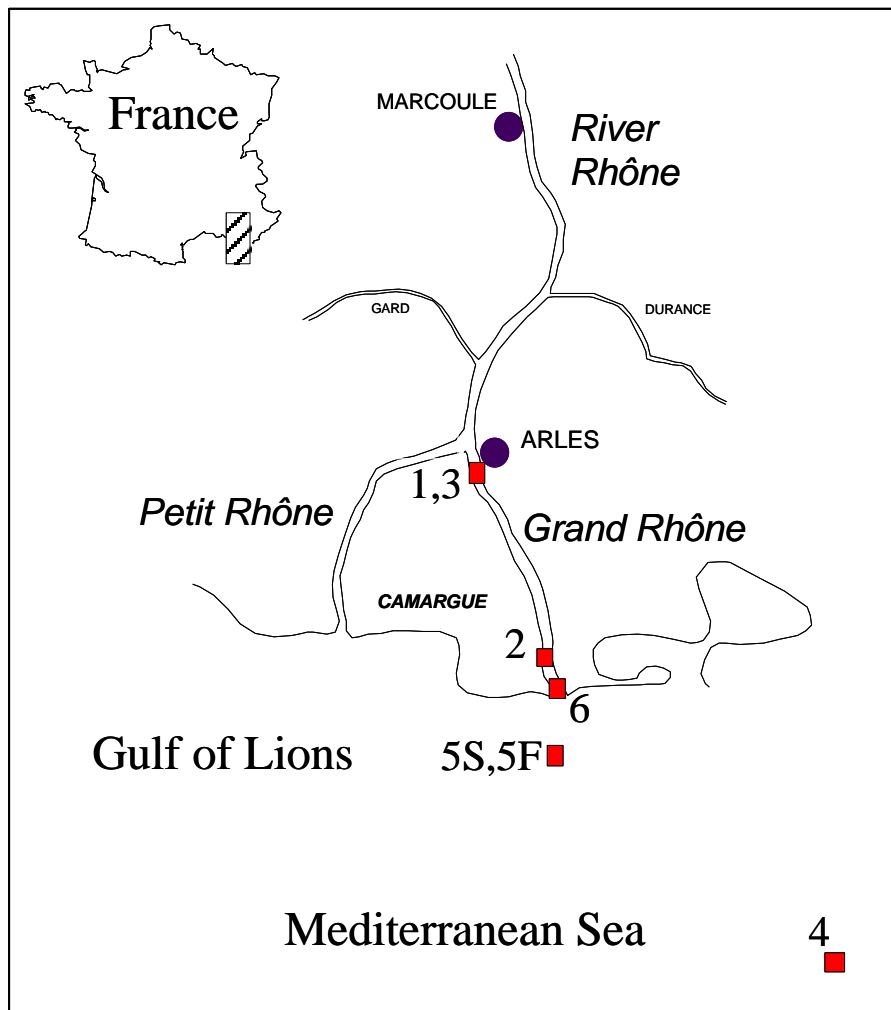


Figure 1