Radionuclides
Suspended matter
Estuary
St. Lawrence river

Radioéléments
Suspensions
Estuaire
Fleuve Saint-Laurent

Distribution of some radionuclides in the St. Lawrence estuary, Quebec, Canada

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ABSTRACT

The distribution of γ-emitting radionuclides in the St. Lawrence estuary was studied in 1978 and 1979, by means of double sampling and the flocculation and centrifugation of very large volumes of water. Eleven radionuclides were detected, originating from a variety of sources, including soil erosion and nuclear weapons testing. The concentrations measured in 1979 were higher than those of 1978; the 21st Chinese nuclear test could be responsible for the increase of some radionuclides. Concentrations decrease markedly from the freshwater part to the marine region of the estuary. Dilution by oceanic waters, relative affinity with suspended matter and radioactive decay are the principal mechanisms involved in the distribution patterns. Cesium-137, ^{144}Ce, ^{226}Ra, ^{228} Ra and ^{228}Th are strongly associated with suspended matter, while about two thirds of ^{7}Be, ^{106}Ru and ^{235}U are present in the liquid phase. Results suggest that ^{235}U is released from sediments in the maximum turbidity zone.

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RÉSUMÉ

Distribution de quelques radioéléments dans l'estuaire du Saint-Laurent, Québec, Canada

La présence et la distribution de certains radioéléments dans l'estuaire du fleuve Saint-Laurent (Québec, Canada) ont été étudiées en 1978 et 1979 à partir d'un double échantillonnage par floculation et centrifugation de grands volumes d'eau. Onze éléments provenant de sources diverses ont été mesurés, et les concentrations en 1979 étaient généralement supérieures à celles de 1978. La 21e explosion nucléaire chinoise pourrait être responsable de l'augmentation des teneurs de certains éléments. On remarque généralement une diminution des concentrations depuis l'eau douce tout au long de l'estuaire. La dilution par les eaux océaniques, l'affinité de certains éléments pour la matière solide en suspension et la désintégration sont les principaux mécanismes régissant la distribution des radioéléments. Le césium-137, le ^{144}Ce, le ^{226}Ra, le ^{228}Ra et le ^{228}Th sont fortement associés à la matière particulaire en suspension, tandis que les deux tiers environ du ^{7}Be, du ^{106}Ru et de l'^{235}U sont dans la phase aqueuse. Les résultats montrent une libération de l'uranium-235 à partir des sédiments, dans le bouchon de turbidité.

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INTRODUCTION

The St. Lawrence river has a drainage basin of 1.32×10^6 km^2. The mean annual runoff is 11×10^6 m^3.sec.$^{-1}$, with a flood peak in April and May contributing 23% of the yearly drainage (d'Anglejan, 1981). The suspended matter load is estimated at about 5-10 million tons per year, the major part occurring in spring (Sérodes, 1980).

In the middle estuary, two channels, respectively along the north and south shore, control the general circulation of waters, which is cyclonic. Tidal inflow is dominant in the north channel, which is deep near Ile aux Lièvres but decreases in depth from there on to Ile d'Orléans. The river discharge preferentially follows the shallow south channel, which exhibits lower surface salinities and higher turbidity (d'Anglejan, 1981). In the lower estuary, the beginning of the Laurentian channel forms a deep central trough flanked by shallow shelf areas along the shores.

Sea water intrudes into the estuary up to the vicinity of Ile d'Orléans (Fig. 1). The middle estuary is considered as partially mixed, while the lower estuary is characterized by a three-layer stratification (Kranck, 1979). Surface salinities are highly variable in the maximum turbidity zone, depending on tide and freshwater discharge fluctuations. Downstream from Cap-aux-Oies, where it varies between 14 and 19‰, the surface water salinity increases gradually, reaching 26-30‰ near Rimouski.

Several studies have described the space and time distribution of suspended matter in the estuary (Sérodes, 1975; d'Anglejan, Ingram, 1976; Kranck, 1979; Silverberg, Sundby, 1979; d'Anglejan, 1981). Compared to other major rivers of the world, the concentrations of suspended matter in the St. Lawrence river are very low (5 to 20 mg.l^{-1} at Quebec city), except in April and May when they are higher. During our sampling, we measured 7 and 16 mg.l^{-1} respectively in November 1978 and September 1979 at Saint-Jean (station 1), which is located 45 km downstream from Quebec city, a few kilometres before the maximum turbidity zone. In this latter region, suspended matter concentrations are very high (15 to >200 mg.l^{-1}) and turbidity extends along the south shore as far as Cap-aux-Oies, where concentrations decrease to about 10 mg.l^{-1}. The main particularity of the sedimentological regime of the middle estuary is an intense accretion of mud on some tidal flats during the summer months (Sérodes, 1980). The lower estuary is characterized by particle-poor surface waters (<1-3 mg.l^{-1}).

There is no study dealing with radionuclides in the estuary itself; in recent years, however, Roy *et al.* (1979; 1981 *a*; 1981 *b*) have obtained numerous results on the radioactivity of the St. Lawrence river and its tributaries upstream of Quebec City by using the sludge produced by municipal water filtration plants. The object of this paper is to present and discuss the distribution of certain radionuclides (Table 1) from fresh water to saline water along a 420 km zone of the St. Lawrence estuary, and to evaluate their respective affinities for the solid and liquid phases.

EXPERIMENTAL METHODS

As radionuclide concentrations are very low in surface water and even more so in saline water, their measurement requires the sampling of very large volumes of water. Consequently, 1 300 l were sampled for high turbidity water and 1 800 l for fresh water (Saint-Jean, station 1) and highly saline water (Ile-aux-Lièvres and Rimouski, stations 4 and 5). All samples were pumped at low tide from surface water. During the filling of the tanks, three one-litre aliquots were taken at regular intervals to determine the amount

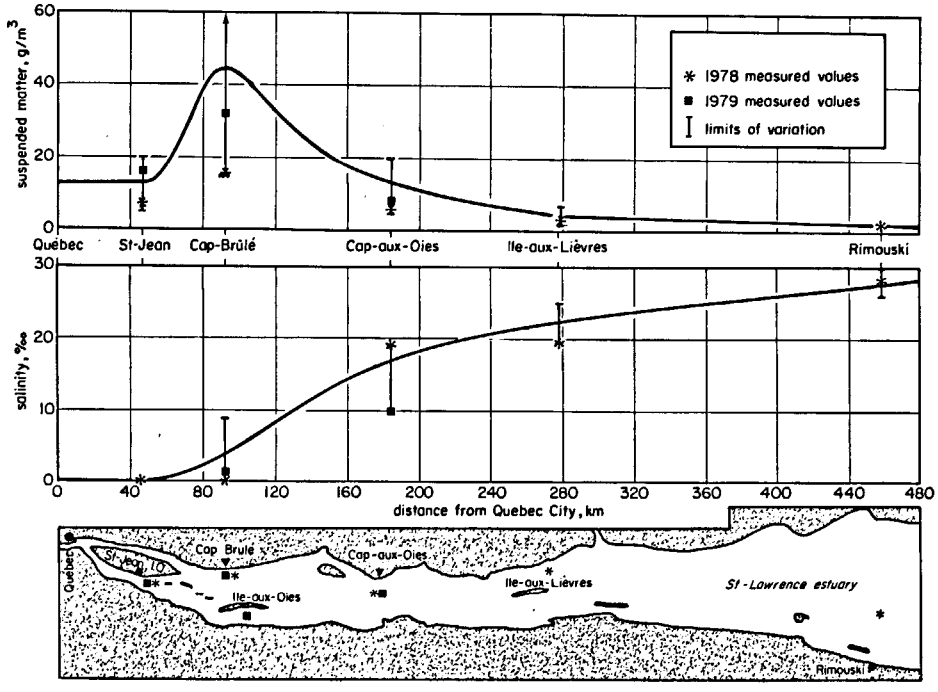

Figure 1

Longitudinal variations of suspended matter and salinity in the St. Lawrence estuary.

Variations de la salinité et de la matière en suspension le long de l'estuaire du Saint-Laurent.

Table 1
Nuclear properties required for the measurement of radionuclides and γ-emitters.
Caractéristiques des radioéléments et émetteurs γ mesurés.

Radionuclides detected	γ-emitter detected	γ-emitter energy (keV)	Abundance of the γ-ray (%)	Half-lives of radionuclides detected
^{7}Be	id	477	10.3	53 *d*
^{95}Zr	id	724; 757	44; 55	65 *d*
^{95}Nb	id	766	100	35 *d*
^{106}Ru	^{106}Rh	622	9.8	367 *d*
^{125}Sb	id	428	30	2,7 *y*
^{137}Cs	id	662	84.6	30 *y*
^{144}Ce	id	134	11.1	284 *d*
^{226}Ra	^{214}Pb	352	37	$1,6 \times 10^{3}$ *y*
^{228}Ra	^{228}Ac	909	27	5,8 *y*
^{228}Th	^{212}Pb; ^{208}Tl	239; 583	47; 31	1,91 *y*
^{235}U	id	185,7	54	7×10^{8} *y*

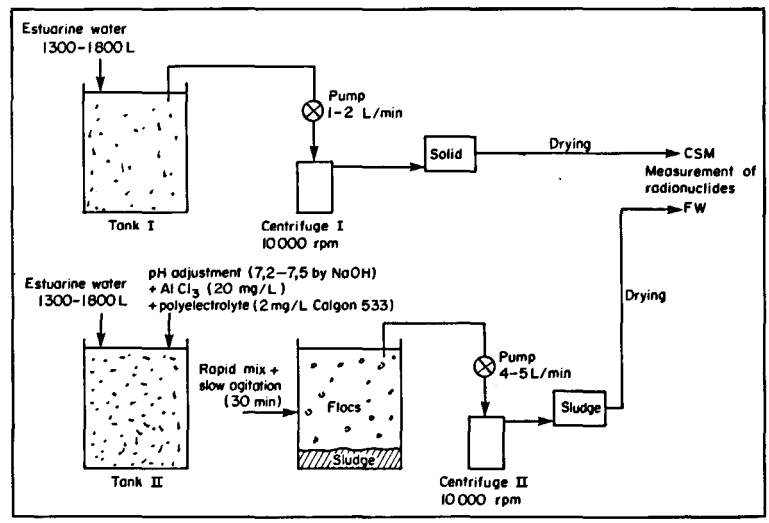

Figure 2
Treatment of samples by centrifugation and flocculation.
Méthode de traitement des échantillons d'eau par centrifugation et floculation.

of suspended matter. The filling operation lasted about 15 minutes.

In 1978, samples were gathered on August 13 (station 5) and on November 15-17 (stations 1, 2N, 3 and 4). The radionuclides were collected by a treatment based on the flocculation of raw water with an aluminum salt (Fig. 2). First, a 50% solution of NaOH was used to maintain the pH between 7.2 and 7.5 after the addition of $AlCl_3$ to a concentration of 20 $mg.l^{-1}$. A cationic polyelectrolyte (calgon 533) was also added (2 $mg.l^{-1}$), to improve the settlement of flocs. The flocculation aid was applied in a much higher concentration than in municipal water filtration plants, in order to get big flocs settling rapidly, even though the boat was in constant motion.

Once all the chemicals were added, the solution was rapidly agitated for one to two minutes, followed by a slow agitation for 30 min., after which most of the big flocs were deposited at the bottom of the tank. The supernatant was then centrifuged with a Wesfalia centrifuge running at 10000 rpm at a rate of 4 to 5 $l.min^{-1}$, in order to recover the small flocs. The solids recovered by centrifugation were combined with those settled at the bottom of the tanks, placed in plastic bags and brought to the laboratory. The time required for a complete sampling was of the order of 6 to 7 hours. In the laboratory, the sludge samples were centrifuged, air-dried at 30°C, weighed and analysed for their radioactivity.

The analysis of the results obtained in 1978 showed that it was necessary to know the amount of radionuclides specifically associated with the suspended matter in order to obtain a better understanding of their behaviour. Therefore, in 1979, double samplings were taken between September 9 to 12 at stations 1, 2N, 2S and 3 (*see* Fig. 1). Sampling was carried out as before at low tide with surface water, which was pumped into two different tanks. As shown schematically in Figure 2, one sample was flocculated as previously described and the other was used to collect the suspended matter by centrifugation; the sludge and suspended matter samples are identified respectively as FW and CSM. A very efficient recovery of the suspended matter would have required a water centrifugal output much less than the rate of 1 to 2 $l.min.^{-1}$ which was adopted in order to limit the centrifugation time to less than 24 hours. Under these conditions, 60% of the suspended matter was recovered on the average as compared with the amount retained on a 0.45 μm Millipore filter paper; the suspended matter concentrations are those which were effectively recovered by centrifugation (CSM), and are not the expression of suspended matter measured by filtration.

The radioactivity measurements were carried out with a Ge(Li) coaxial detector interfaced with an Inotech 5200 nuclear spectrometer. The samples, weighing between 10 and 60 g, were packed in aluminum cups, placed on the top of the detector and counted in a fixed

geometry for about 70 hours. The spectra in the energy range of 100 to 1 300 keV were accumulated over 1 024 channels. The detector was standardized for various peak heights by using two certified reference materials obtained from the Canada Centre for Mineral and Energy Technology (Ingles *et al.*, 1977; Faye *et al.*, 1979), namely the radioactive ores DH-1 and BL-5; DH-1 contained 0.177% uranium and 0.105% thorium, while BL-5 contained 7.09% uranium. The DH-1 reference material was used as such, whereas the BL-5 was diluted with four parts of Al_2O_3. The samples were counted several weeks after sampling, so that secular equilibrium was reached in the case of the ^{214}Pb-^{226}Ra and the ^{212}Pb-^{228}Th pairs. The counting rates were calculated by subtracting the background from the photopeak areas. They were corrected for the counting efficiency, the abundance of the γ-ray and the decay time between counting and sampling. The results are given for the sampling date. The nuclear properties relative to the measurements of the radionuclides reported in this study are given in Table 1. In the case of the 185.7 keV γ-ray used to calculate the amount of ^{235}U, the contribution of 186 keV γ-ray due to ^{226}Ra was subtracted; this was done by using the counting rate of the 352 keV γ-ray due to the ^{214}Pb-^{226}Ra pair and in taking into account the respective abundance and counting efficiencies of the 186 and 352 keV γ-rays.

RESULT AND DISCUSSION

The concentrations of eleven radionuclides detected at different stations in flocculated water samples in 1978 are given in Table 2. The same radionuclides, as detected in both flocculated water and centrifuged suspended matter in 1979, are given in Table 3.

Table 2

Concentrations of radionuclides in the St. Lawrence estuary, 1978, expressed in pCi.m^{-3} of flocculated water.

Concentrations des radioéléments dans l'estuaire du Saint-Laurent en 1978, exprimées en pCi.m^{-3} d'eau floculée.

	Location				
Radionuclides	1 St-Jean	2N Cap Brûlé	3 Cap-aux-Oies	4 Ile-aux-Lièvres	5 Rimouski
^{7}Be	266	110	81	56	10
^{95}Nb	8	6	4	1	1
^{95}Zr	4	4	ND	ND	ND
^{106}Ru	67	54	26	6	6
^{125}Sb	8	4	5	ND	1
^{137}Cs	35	35	13	3	2
^{144}Ce	29	39	21	8	2
^{226}Ra	10	12	3	4	3
^{228}Ra	20	21	15	9	3
^{228}Th	16	29	13	8	3
^{235}U	4	9	15	6	3

ND = not detected.

Table 3

Concentrations of radionuclides in centrifugated suspended matter (CSM) and flocculated water (FW) of the St. Lawrence estuary, 1979, expressed in pCi.m^{-3} of raw water.

Concentrations des radioéléments dans la matière en suspension obtenue par centrifugation (CSM) et dans l'eau floculée (FW) de l'estuaire du Saint-Laurent en 1979, exprimées en pCi.m^{-3} d'eau.

	Location							
	1 St-Jean		2N Cap Brûlé		2S Ile-aux-Oies		3 Cap-aux-Oies	
Radionuclides	CSM	FW	CSM	FW	CSM	FW	CSM	FW
^{7}Be	166	693	46	381	308	418	96	182
^{95}Nb	6	24	ND	23	ND	44	ND	3
^{95}Zr	4	23	ND	9	ND	63	ND	9
^{106}Ru	12	61	2	76	35	96	8	34
^{125}Sb	1	9	ND	3	5	6	1	4
^{137}Cs	36	49	30	96	80	124	22	26
^{144}Ce	10	15	12	33	25	49	12	18
^{226}Ra	11	13	8	28	32	35	7	8
^{228}Ra	17	26	15	39	39	53	6	8
^{228}Th	21	32	21	86	68	89	23	30
^{235}U	1	5	1	6	2	8	1	19

N and S refer to the north and south channel positions of station 2.

Beryllium-7 is a fallout cosmic-ray produced radionuclide. The ^{95}Zr-^{95}Nb pair, ^{106}Ru, ^{125}Sb, ^{137}Cs and ^{144}Ce are fallout fission products resulting from nuclear weapons testing (Roy *et al.*, 1979; 1981 *a*). Radium-226, ^{228}Ra, ^{228}Th and ^{235}U occur as a result of the presence of natural radioactivity in the environment, and find their way in the water by numerous processes such as soil erosion, resuspension of bottom sediments and sediment-water interactions in zones of highly variable salinity. At similar dates, in the St. Lawrence river at the Quebec City level, a few activation products like ^{46}Sc, ^{134}Cs and ^{54}Mn were detected in addition to those observed in the estuary (Roy *et al.*, 1981 *b*). These activation products come from the operation of nuclear installations and partly from weapon testing fallout, in the case of ^{54}Mn.

Variations in the concentrations of the radionuclides

Comparison of the concentrations of the FW samples from the 1979 and 1978 cruises shows that, except for ^{144}Ce, concentrations are generally higher in 1979 than in 1978 (*see* Tables 2 and 3). The main factors affecting the variation of the concentrations are the amount of the suspended matter, an increase in fallout, the decay rate of the radionuclides and the retention time in the St. Lawrence watershed.

Beryllium-7

This cosmic-ray produced radionuclide is generated at a nearly constant rate in the atmosphere.

Its concentration might thus be expected to be independent of its decay rate and of its retention time in the river watershed, but to be strongly influenced by the amount of precipitation. It would therefore appear that the increase in rainfall during August-September 1979 accounts for the large increase of ^{7}Be fallout in 1979 as compared with 1978.

Cesium-137, ^{228}Th, ^{228}Ra, ^{226}Ra

These radionuclides are introduced continuously in the aquatic system by soil erosion or from bottom sediments; the long-lived ^{137}Cs is considered to behave in the same manner as naturally occurring radionuclides. For these radionuclides, the variation in concentrations should be related to the variations in the amount of the suspended matter. And, indeed, the concentrations of suspended matter were higher in 1979 than in 1978 (Fig. 1).

Zirconium-95-^{95}Nb pair, ^{106}Ru, ^{125}Sb, ^{144}Ce

For these radionuclides, the variation of the concentrations can be summarized as follows: a fairly large increase in the ^{95}Zr-^{95}Nb pair; a small variation for ^{106}Ru and ^{125}Sb; and a definite decrease for ^{144}Ce. It should be noted that a similar behaviour was observed upstream (Roy *et al.*, unpublished results). Because the increase in the amount of suspended matter in 1979 should have influenced all these radionuclides in the same manner, it is necessary to look elsewhere for an explanation of the three different patterns observed. The origin of the radionuclides and their retention time in the watershed could be part of the explanation. Fallout following Chinese nuclear weapons testing is responsible for the presence of these five fission products, and the 18th, 19th, 20th and 21st tests, the characteristics of which are given below, have contributed to some extent to their concentration.

Chinese tests	Year	Date	Approximate yield (kilotons)
18th	1976	November 17	4000
19th	1977	September 17	20
20th	1978	March 14	20
21st	1978	December 14	< 20

The fallout of the 21st test would have been deposited on snow-covered ground, and would have spread over the very large St. Lawrence watershed. Snow melting in spring would have carried the greater part of this fallout into the tributaries of the St. Lawrence, so that the peak concentrations should have occurred late in summer in the estuary. This could explain the increase in the concentration of the ^{95}Zr-^{95}Nb pair. The 2,7 *y* ^{125}Sb would be due mostly to fallout from the powerful 18th test; the decrease in fallout between 1978 and 1979 would have been compensated by an increase in the amount of suspended matter in 1979, so that the concentration of ^{125}Sb remained about the same in both years. The variation in the concentrations of the 367 *d* ^{106}Ru could be explained by the fact that the fraction which disappeared through decay was replenished by fallout from the 21st explosion, with the result that in 1979 about half of the ^{106}Ru came from the 21st explosion, while the remainder came from the preceding tests. The decrease in the concentrations of the 284 *d* ^{144}Ce could be explained partly by the fact that the large fraction which disappeared by decay was not fully replaced by the fallout from the 21st explosion.

Geographic distribution

The geographic variations of radionuclide concentrations along the estuary are more adequately described using the 1978 results (Table 2) because the region investigated was larger than in 1979, when attention was focussed on the distribution of radionuclides between suspended matter and water. The first point to be noted is that concentrations are always higher in the freshwater part of the estuary (Saint-Jean, station 1) than in the lower estuary (Rimouski, station 5). This confirms the fact that the origin of radionuclides is the drainage basin and not the ocean. The second important point is that all the radionuclides do not exhibit their maximum concentration at Saint-Jean. As far as longitudinal variations are concerned, only the eight principal radionuclides have been considered: ^{95}Nb, ^{95}Zr, ^{125}Sb are excluded, either because they are not detected at all stations or because they are present in very low concentrations. Five radionuclides (^{137}Cs, ^{144}Ce, ^{226}Ra, ^{228}Ra, ^{228}Th) present a maximum value at Cap-Brûlé (station 2N) in a zone of very turbid water. Both ^{7}Be and ^{106}Ru show a continuous decrease, which is steeper in the case of ^{7}Be. Finally, ^{235}U exists in very similar concentrations in fresh water and in highly

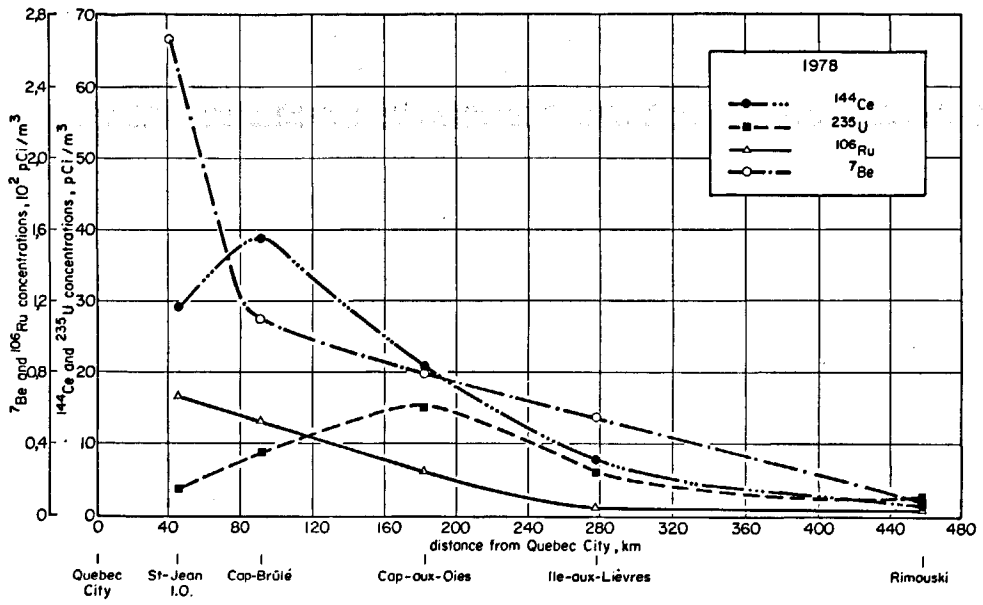

Figure 3
Distribution patterns of radionuclides along the St. Lawrence estuary.
Distribution des radioéléments le long de l'estuaire du Saint-Laurent.

saline water but exhibits a maximum at the limit of the high turbidity zone in median saline water (station 3, Cap-aux-Oies). These characteristics are illustrated in Figure 3, where the first group of radionuclides is represented by ^{144}Ce. It should be noted that the same tendencies are found in the 1979 results, except for ^{106}Ru which behaves like the ^{144}Ce group.

Three basic mechanisms could help to explain the geographic distribution of radionuclides in the estuary: the increasing dilution of fresh water by less radioactive saline waters; the relative affinity for suspended matter coupled with variations of suspended solids concentrations and salinity; and finally the decay of radionuclides during the residence time of water and solids between the stations.

The five elements represented by ^{144}Ce in Figure 3 follow closely the pattern of turbidity in the estuary as shown in Figure 1. With the exception of ^{144}Ce, the four others have long half-lives; consequently, only the first two mechanims should be considered. The strong association of ^{137}Cs with solid matter in freshwater systems being well-known (Tamura, 1972; Francis, Brinkley, 1976; Jackson, Inch, 1980), it could be inferred that ^{144}Ce, ^{137}Cs, ^{226}Ra and ^{228}Th are preferentially present as adsorbed forms. Then, their concentrations should be proportional to suspended solid values, even though the use of flocculation to collect the radionuclides does not differentiate the part associated with suspended matter from the amount extracted as dissolved species by adsorption on precipitating alumina. This is confirmed by a regression analysis made with the four CSM samples of 1979, which shows a good correlation between suspended matter and radionuclide concentrations (coefficient r^2 varying from 0.71 for ^{226}Ra to 0.84 for ^{144}Ce).

Ruthenium-106 does not follow the pattern of turbidity and presents a continuous decrease from Saint-Jean to Rimouski. In this case, dilution seems to be the most important mechanism, but the retention time of waters along the estuary could account for a small part of the decrease because ^{106}Ru has a half-life of 367 days.

Beryllium-7 is the most abundant γ-emitting radionuclide at any station in the estuary. The gradual decreases of concentrations do not follow the corresponding dilutions along the estuary. For example, the dilution between Cap Brûlé (station 2N) and Ile-aux-Lièvres (station 3) is much more important than the dilution between Saint-Jean (station 1) and Cap Brûlé, even though the concentration ratios of ^{7}Be are quite similar. Consequently, other mechanisms must explain the observed variations. It is difficult to evaluate the influence of suspended matter, because the decrease in ^{7}Be concentrations between Saint-Jean clear waters and Cap Brûlé very turbid waters is too high. It would appear that the decreases in ^{7}Be concentrations are due mainly to its decay and partly to dilution or variations in suspended matter.

The behaviour of ^{235}U is difficult to explain. This element of natural origin has a very long half-life; in consequence, only dilution and turbidity differences should be responsible for the observed pattern. Dilution is certainly not an important factor, at least in the upper part of the estuary, because ^{235}U concentrations increase by a factor of 2 from station 1 to station 2N and from station 2N to station 3. A similar longitudinal distribution is observed in 1979 (FW samples in Table 3). A portion of the first increase is probably due to the presence of more suspended matter at Cap Brûlé, but this is not the case for the second increase at Cap-aux-Oies, where turbidity is much lower than at Cap Brûlé. A possible explanation of this phenomenon could be a "source" of ^{235}U between the end of the freshwater zone and the end of the turbid zone, 150 km downstream. Since the St. Lawrence does not have important tributaries in this region, the source must be in the estuary itself, and could be explained by two mechanisms: the segregation of uranium-rich particles along the maximum turbidity zone, or the release of ^{235}U from suspended or bottom sediments to the water column. The former hypothesis is highly improbable, and is set aside by 1979 results (CSM in Table 3). Consequently, it should be admitted that there is a

transfer of ^{235}U from the solid phase to the water phase. This release of uranium was observed by Thomson *et al.* (1973) in Long Island Sound on bottom sediment cores, as a result of oxidation at the sediment-water interface. This transformation probably concerns some other radionuclides, but is more difficult to detect among the numerous mechanisms involved. The desorption of ^{235}U is also made noticeable because this element has very low concentrations in freshwater samples.

From all the results of the 1978 cruise, the necessity of evaluating the relative amount of radionuclides associated with particulate matter and with water became evident; this fact was at the origin of the double sampling technique used in 1979.

The relative affinity of radionuclides for water and for suspended matter

The 1979 cruise was limited to the maximum turbidity zone of the estuary. The concentrations of radionuclides in centrifuged suspended matter (CSM) and flocculated water (FW) are given in Table 3. At all locations and for all the radionuclides, FW values are higher than CSM values. This is the result of the adsorption of dissolved species on freshly precipitated aluminium trihydroxide and also of a better recovery of suspended matter by flocculation compared to centrifugation which retains only 60% of the solids as already mentioned.

In order to evaluate the amount of suspended solids trapped by flocs, a basic assumption is necessary, namely that ^{137}Cs scavenged by flocculation of raw water is only the part which is associated with the suspended matter, and that no dissolved ^{137}Cs is extracted by this process. This hypothesis has been confirmed by laboratory experiments conducted on highly concentrated solutions of ^{137}Cs (up to 67 500 $pCi.l^{-1}$) submitted to flocculation with aluminium sulphate, which showed no extraction of cesium by flocs.

Thus, the FW: CSM ratio of ^{137}Cs concentrations gives, for the different sampling stations, the proportion of suspended matter recovered by flocculation (FSM) compared with centrifugation (Table 4). The ratios of suspended matter recovered by flocculation and centrifugation vary from 1.2 in saline water to 3.2 at Cap Brûlé in highly turbid water. This latter value is very high compared to the others, and could be due to a bad performance of the centrifugation at this station.

Using the suspended matter ratios, the part of the radionuclide concentrations related to suspended matter in FW samples can be calculated by multiplying the ratio with the CSM value. These calculations are then related to FW values, and expressed as percentages of radionuclides associated with suspended matter (Table 5). In some cases, the calculated percentages are higher than 100 and are expressed as >100%. In freshwater samples, ^{144}Ce, ^{226}Ra, ^{228}Ra and ^{228}Th are highly associated with suspended matter, and this property is maintained in saline water. This confirms the results of 1978 already described (Table 2). Beryllium-7, ^{106}Ru and ^{235}U present various percentages from one station to another, but in fresh water about two-thirds of their total concentration is under dissolved or colloidal forms. The proportion of ^{235}U associated with suspended matter varies from 28% in clear fresh water to 41-48% in turbid fresh water, and then drops to 4% in saline water. This illustrates the results already shown in Figure 3, and supports the release of ^{235}U from bottom or suspended sediments to the water column by oxidation processes, as proposed by Thomson *et al.* (1973).

Table 4

Characteristics of centrifuged and flocculated water samples of the St. Lawrence estuary, 1979.

Caractéristiques des échantillons d'eau de l'estuaire du Saint-Laurent (1979) soumis à la floculation et à la centrifugation.

	Location			
	1 St Jean	2N Cap Brûlé	2S Ile-aux-Oies	3 Cap-aux-Oies
Salinity (‰)	0	0	0	10
Centrifuged SM (CSM, $g.m^{-3}$)	17.4	31.9	46.7	20.3
Flocculated SM (*) (FSM, $g.m^{-3}$)	23.5	102.2	72.4	24.5
FSM/CSM	1.35	3.20	1.55	1.20

(*) Calculated from ^{137}Cs data.

Table 5

Percentages of radionuclides associated with suspended matter in flocculated water samples of the St. Lawrence estuary, 1979.

Pourcentage des teneurs en radioéléments associés à la matière en suspension dans des échantillons d'eau floculée de l'estuaire du Saint-Laurent (1979).

	Location			
Radionuclides	1 St-Jean	2N Cap Brûlé	2S Ile-aux-Oies	3 Cap-aux-Oies
^{7}Be	32	39	> 100	63
^{106}Ru	27	8	56	28
^{125}Sb	17	—	> 100	39
^{137}Cs(*)	100	100	100	100
^{144}Ce	93	> 100	78	85
^{226}Ra	> 100	97	> 100	96
^{228}Ra	88	> 100	> 100	87
^{228}Th	89	78	> 100	92
^{235}U	28	41	48	4

(*) Hypothesis of calculations.

CONCLUSION

Even though the flocculation and centrifugation of very large volumes of water are constraining techniques, they constitute valuable means of collecting radionuclides present in very dilute concentrations. The nuclides detected in the St. Lawrence estuary come from the watershed and are of geological or atmospheric fallout origin. Various mechanisms are involved in the explanation of the temporal and geographic evolution of the concentrations, dilution and suspended matter abundance being the most important; decay is, however, the leading factor for ^{7}Be, while release from the sediments plays a major role in the case of ^{235}U. A number of applications could be foreseen from these results, such as the dating of sediment deposits or the evaluation of residence time of sediments or water. In that latter case, the use of the short-lived ^{7}Be seems to be the most appropriate, because it is very abundant; unfortunately, its natural atmospheric origin makes it highly variable, and it will be necessary to measure its input fluctuations at the same time.

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