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Determination of the residence time of suspended particles in the turbidity maximum of the Loire estuary by ^{7}Be analysis

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Abstract

The aim of the present work was to evaluate the half life of suspended particles in the Loire estuarine turbidity maximum by analysis of ^{7}Be budgets. The methodology was based on in situ sampling and further measurements aiming at quantifying ^{7}Be sources (atmospheric deposition and river inputs) and ^{7}Be stock in the water column of the turbidity maximum. ^{7}Be river inputs were determined by monthly ^{7}Be measurements performed upstream of the estuary. ^{7}Be atmospheric deposition was estimated by using an empirical relation between ^{7}Be deposition and rainfall. ^{7}Be in particles of the estuarine turbidity maximum was measured at eight different dates corresponding to different tidal and hydrological conditions. ^{7}Be sources and stocks thus determined have been compared to a mathematical model. Results allow to quantify the 'standard half life' of suspended particles in the Loire estuarine turbidity maximum and show that it depends on the season (6–10 months in summer and about 0.7 month during flood periods). Furthermore, a rather good linear correlation was observed between the standard half life of particles and the sum of flow rates in the Loire river during 60 days before each sampling date. The kinetic evolution of the mass of particles within the turbidity maximum could be estimated by this method and appeared to be consistent with previous studies. Moreover, the method proposed in this study could presumably be used for estimating ^{60}Co concentrations in the estuarine turbidity maximum.

Keywords: beryllium; Loire estuary; suspended particulate matter; residence time

1. Introduction

Macrotidal estuaries are generally characterised by a 'turbidity maximum' zone. For example, in the Loire estuary, concentrations of suspended particulate matter (SPM) in the turbidity maximum (up to $>3000\,mg\,l^{-1}$) are much higher than those measured upriver (ca. 20–$100\,mg\,l^{-1}$). The estuary turbidity maximum may be explained by several physical or chemical processes. These are effect of asymmetric tides in the deposition and resuspension of sediments (Uncles & Stephens, 1989), effect of stratification on turbulence which could promote particle accumulation (Geyer, 1993) and formation of new particles at low–high salinity contact by flocculation (Aloïsi et al., 1982; Martin et al., 1994; Sholkovitz, 1976, 1978).

Suspended solids in estuaries play a major role in the transport of pollutants from rivers to seawater because they may react as traps for trace elements (Morris et al., 1986; Pham et al., 1997; Turner & Millward, 1994). Consequently, it is important to know the mean residence time of the particulate matter pool in the water column of the turbidity maximum, that carries anthropogenic or natural sources of pollutants from the riverine watershed. Natural radionuclides have often been used as time tracers of particles in the environment (Walling & Woodward, 1992). In particular, ^{7}Be has been used for studies in watersheds and aquatic environments (Bonté et al., 2000; Canuel, Martens, &

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Benninger, 1990; Dibb & Rice, 1989; Dominik, Burrus, & Vernet, 1989; Feng, Cochran, & Hirschberg, 1999; Martin, Mouchel, & Thomas, 1986; Olsen, Larsen, Lowry, Cutshall, & Nicholls, 1986; Olsen et al., 1986). ^{7}Be (half life = 53 days) is a cosmogenic radionuclide supplied to estuarine waters by both direct atmospheric deposition and upstream river inputs. Its atmospheric deposition is mainly governed by rainfall scavenging. ^{7}Be may be a useful tracer to evaluate particulate transport in an estuary because it satisfies several conditions: (1) because of its abundance, it is easily measurable in water and in particles by γ spectrometry; (2) ^{7}Be has a great affinity for particles, as shown by high average distribution coefficients (ratio particulate activity/dissolved activity) (in the range 10^{4}–10^{5} l kg^{-1}) (Dibb & Rice, 1989; Olsen et al., 1986; Thomas, 1988); and (3) its half life is in the same order of magnitude, or lower, than the expected particles transit time in an estuary (some months).

The aim of the present work was to evaluate the mean residence time of suspended particles in the Loire estuarine turbidity maximum by analysis of ^{7}Be budgets. The methodology is based on measurements aiming at quantifying ^{7}Be sources (atmospheric deposition and river inputs) and ^{7}Be stock in the turbidity maximum. ^{7}Be sources and stocks were then compared using a mathematical model.

2. Experimental section and methods

2.1. Study area

The Loire estuary is situated in Southern Brittany (France) and is 80 km long. The Loire river has an irregular flow rate varying from 80 to 5500 m^{3} s^{-1} with an average of 800 m^{3} s^{-1}. Solid load input in the estuary is about 10^{9} kg year^{-1} (Figueres, Martin, Meybeck, & Setler, 1985). The Loire estuary is characterised by a turbidity maximum zone bounded by lower SPM concentrations both landward and seaward and representing a total mass of particles of about 500,000 t (Migniot, 1993). Part of the particulate matter pool of the turbidity maximum is expelled into the open sea during high winter river flows.

2.2. Sampling and ^{7}Be analysis

Water and SPM samples were collected in river, estuary and rainfall in order to evaluate ^{7}Be sources and stocks. Locations of the sampling points in the Loire estuary are indicated in Fig. 1.

2.2.1. SPM sampling in the Loire river

SPM are collected monthly along the Loire river by Office de Protection des Rayonnements Ionisants

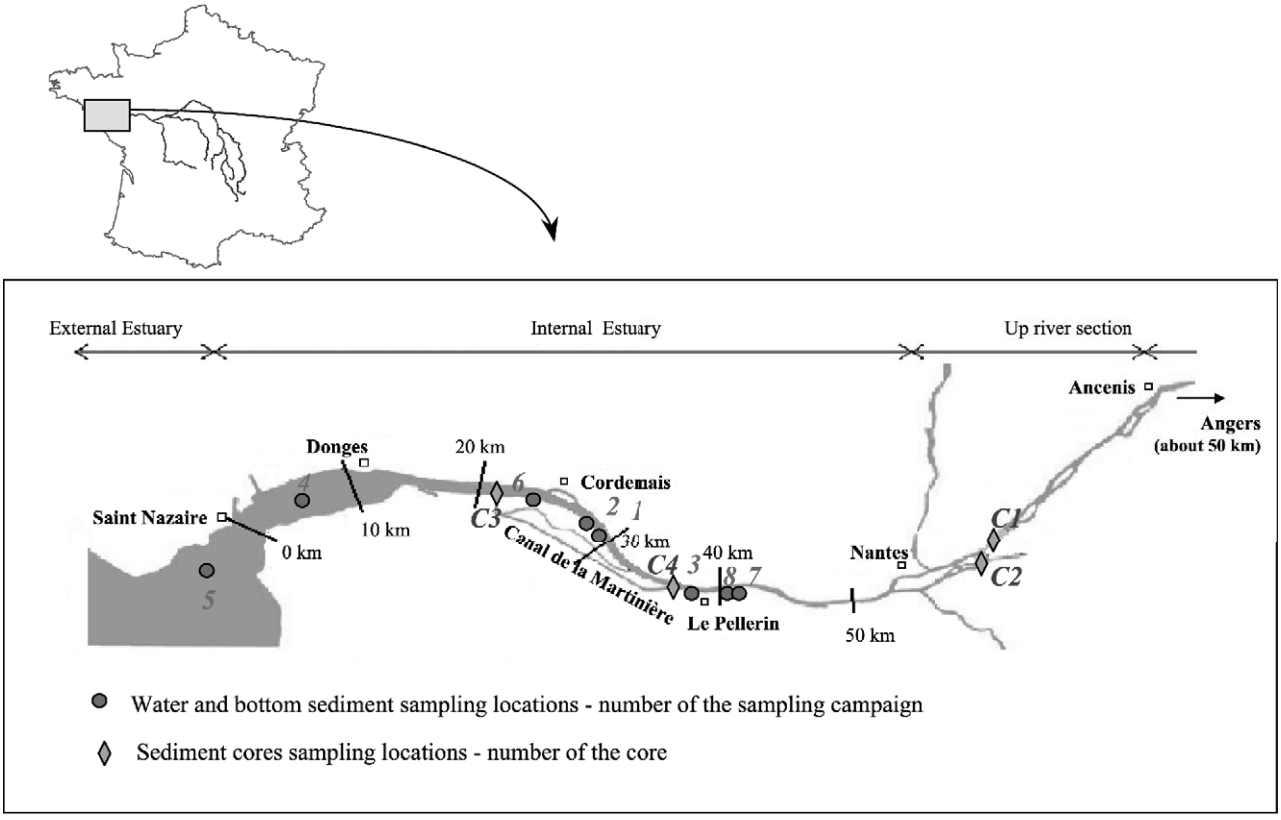

Fig. 1. Location map of the Loire estuary and sampling stations.

(OPRI) to monitor radioactive levels at five different stations. To collect a mean SPM sample each month, water is continuously pumped from the river and passes through a decanter. Particles deposited in the bottom of the decanter at the end of each sampling period are collected and analysed by γ spectrometry. Results are produced as monthly reports edited by OPRI. It may be noted that such a decanter for the sampling of suspended particles in river was compared with other techniques such as centrifugation and cartridge filtration during an intercomparison programme. It was shown that the concentrations of metals in samples collected by decantation, centrifugation or cartridge filtration, respectively, were generally consistent (Ciffroy et al., 1999). Furthermore, samples collected in river by decantation and in the Loire estuary by centrifugation, respectively, may be compared in terms of granulometry: median diameters of SPM collected in river for the intercomparison exercise previously mentioned were in the range 10–15 μm (Ciffroy et al., 1999); the mean value of median diameters for SPM collected in the Loire estuary by centrifugation was 16.6 μm. Consequently, such experimental results suggest that riverine and estuarine samples may be properly compared. For this study, we used 50 monthly ^{7}Be measurements performed at the Angers station, situated just upstream of the Loire estuary, during the period 01/1997–05/2001.

2.2.2. Rainfall sampling

Rain samples are collected monthly by OPRI at the Angers meteorological station. Results are produced as monthly reports edited by OPRI. For this study, we used monthly ^{7}Be measurements performed at the Angers station during the period 01/1997–02/2000.

2.2.3. SPM sampling in the estuarine turbidity maximum (see Fig. 1)

SPM was sampled in the zone of estuarine turbidity maximum at eight different dates corresponding to different tidal and hydrological conditions (Table 1). For each sampling campaign, the zone of estuarine turbidity maximum was localised by turbidity measurements and echo sounding, allowing to detect the presence of mud at the sediment–water column interface. For each date, raw water samples (more than 1000 l) were collected at three different water depths and centrifuged immediately after collection (CEPA Z61 centrifuge). SPM thus collected were afterwards treated as follows: SPM samples were oven-dried overnight at 110 °C and stored in polypropylene tubes ready for non-destructive gamma spectrometric investigations. Counting was conducted at the Laboratoire Souterrain de Modane (L.S.M., CNRS-CEA) in the French Alps using a very low background, high efficiency well-type Ge detector with crystal volume of 430 cm^3 (Reyss, Schmidt, Legeleux, & Bonté, 1995). This big crystal allows the measurement of samples of about 14 cm^3 within the hole of the detector.

2.2.4. SPM sampling in the sea

In February 1999, SPM was collected by centrifugation at a marine station out of influence of the estuary (Le Croisic, France). The procedure of treatment of the sample was the same as previously described for estuarine SPM.

2.2.5. Bottom sediment sampling

For the sampling campaigns performed in the estuary in July 1998 and 1999 and in February 1999, bottom sediments were collected by an Ekman sediment sampler. Furthermore, sediment cores were sampled in May or September 1998 in locations where sedimentation could be expected (out of the main river channel in full current) (see Fig. 1). The procedure of treatment of the samples was the same as previously described for estuarine SPM.

2.2.6. Water sampling in the estuarine turbidity maximum (see Fig. 1)

For sampling campaigns 1–4, water was collected after elimination of SPM to measure ^{7}Be in the dissolved phase. The sampling and treatment procedure is as follows: collection of 50 l at the output of the centrifuge; filtration through a 0.5-μm cartridge (Millipore Milligard CW 06 01S); collection of filtered water; and addition of Fe as carrier in the acidified water together with ^{229}Th spike. $Fe(OH)_3$ precipitates were measured by γ spectrometry under the same conditions as for SPM activities determination. $Be(OH)_3$ and $AlCl_3$ were added as carriers to insure the hydroxide coprecipitation of beryllium with iron. Chemical efficiency for the recovery of Th and Be was measured to be similar by cross-checking the recovery of a ^{229}Th spike and that of Be measured by atomic absorption during preliminary experiments. The recovery efficiency was measured to be better than 80% and often near 95%. Gamma-ray counting efficiencies were determined using a calibration curve obtained as described in Reyss et al. (1995). Results were corrected to take into account decay during the time elapsed between the sampling and counting dates.

3. Experimental results

3.1. ^{7}Be sources

3.1.1. Atmospheric deposition

^{7}Be atmospheric deposition and its relationship with rainfall measured in Angers (expressed in GBq km^{-2} month^{-1}) is presented in Fig. 2. The correlation

Table 1
SPM sampling in the Loire estuarine turbidity maximum and in the sea

Date	Loire river flow rate ($m^3 s^{-1}$)	Sample reference	Salinity ($g l^{-1}$)	Depth (m)	Mean SPM concentration over depth (SPM concentration for surface sampling depth) ($g l^{-1}$)	Particulate ^{7}Be ($Bq kg^{-1}$)	Dissolved ^{7}Be ($Bq m^{-3}$)	^{7}Be distribution coefficient ($l kg^{-1}$)
03/07/1998	292	1HS	7.2	1	1.3 (0.112)	41 ± 2	0.78 ± 0.07	53,000
		1HH	18.4	6		40 ± 1	0.61 ± 0.09	66,000
		1HM	5.7	9.5		30 ± 1	0.48 ± 0.1	63,000
		1HB				20 ± 1		
08/07/1998	270	2HS	7.9	1	1.9 (0.088)	26 ± 1		
		2HH	16.4	7		27 ± 1	0.48 ± 0.08	56,000
		2HM	4.2	11.5		29 ± 1		
		2HB				27 ± 3		
		2ES	3.8	1		35 ± 1	0.73 ± 0.08	48,000
		2EH	4.5	6.5		34 ± 1	0.55 ± 0.09	62,000
		2EM	4.1	10.5		28 ± 1		
		2EB				20 ± 1		
		2LS	0.3	1	1.3 (0.209)	26 ± 2	0.64 ± 0.09	41,000
		2LH	0.1	4		36 ± 2	0.65 ± 0.12	55,000
		2LM	8.5	7.5		25 ± 1		
		2LB				27 ± 1		
		2FH	4.6	5.5		35 ± 1	0.66 ± 0.11	53,000
		2FM	8.8	11.5		25 ± 1		
		2FB				24 ± 2		
		2H*S	10.3	1	4.6 (0.252)	26 ± 2	0.48 ± 0.09	54,000
		2H*H	17.7	6.5		30 ± 2	0.37 ± 0.08	81,000
		2H*M	6.6	11		23 ± 2		
		2H*B				24 ± 2		
15/09/1998	225	3HS	1.1	0.5	3.1 (0.232)	24 ± 2	0.57 ± 0.1	42,000
		3HH	1.6	5		19 ± 1	0.51 ± 0.1	37,000
		3HM	0.6	10		15 ± 1	0.31 ± 0.1	41,000
05/02/1999	1310	4HS	5.9	1	3.5 (0.119)>	38 ± 2	0.49 ± 0.09	78,000
		4HH		8.5		40 ± 2		
		4HM		12.5				
		4ES	1.9	1		46 ± 2		
		4EH	4.7	5.5		43 ± 2		
		4EM		12		37 ± 2		
		4LH		5.5		36 ± 2		
		4FS		1.2		38 ± 2		
		4FH		5		38 ± 2		
		4H*S	4.4	0.1	2.9 (0.117)	41 ± 2		
		4H*H	24	7.1		50 ± 2		
		4H*M		12.5		39 ± 2		
25/02/1999	3070	5HS		2.1		102 ± 2		
		5ES		1		91 ± 2		
		5HM		14		37 ± 2		
		5HB				27 ± 1		
17/05/1999	663	6HS		1.4	2.9 (0.215)			
		6HH	0.3	7				
		6HM		9.5		40 ± 3		
		6FH		5.7		32 ± 3		
		6FM		8.5		36 ± 1		
12/07/1999	267	7HS		1	2.1 (0.132)			
		7HH		5				
		7HM		10.5		25 ± 1		
		7HB				24 ± 1		
		7FS	3.7	2.4		25 ± 2		
		7FH				21 ± 2		
		7FM				24 ± 8		
28/09/1999	354	8LM				23 ± 1		
		8FS		1		41 ± 1		
		8FH	0.2	4.5		27 ± 1		
03/02/1999		Sea	35		0.021	108 ± 3		

Samples are referred to as follows: numbers 1–8, number of sampling date; first letter, tide period; H, high tide; E, ebb tide; L, low tide; F, flood tide; H*, following high tide; second letter, sampling depth; S, surface; H, half of total depth; M, mud zone; B, bottom sediments.

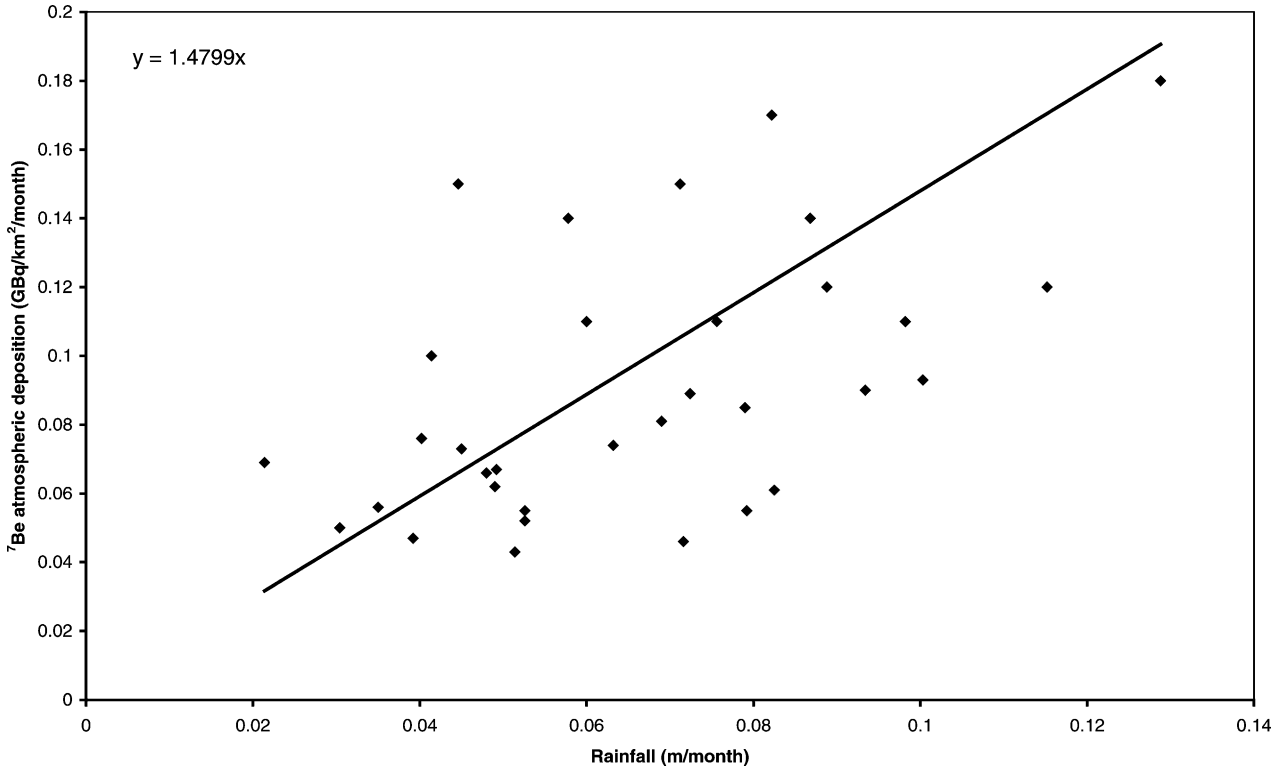

Fig. 2. Relationship between ^{7}Be atmospheric deposition and rainfall in Angers.

between ^{7}Be deposition (D, GBq km^{-2} month^{-1}) and rainfall (R, m month^{-1}) is:

$$D = 1.48R(r^2 = 0.65). \quad (1)$$

Such correlation coefficients have been commonly observed and explained by the fact that rainfall constitutes the major depositional pathway of ^{7}Be (Bonté et al., 2000; Caillet, Arpagaus, Monna, & Dominik, 2001; Olsen et al., 1985; Turekian, Benninger, & Dion, 1983). Some scattering of data points along the regression line could be the result of other processes, such as the season and time elapsed between consecutive rain events (Caillet et al., 2001). Although such processes were not modelled in this study, the correlation coefficient r^2 has been considered high enough to use this relationship for further applications. Consequently, in further modelling calculations, daily ^{7}Be inputs from atmospheric deposition will be calculated using Eq. (1) previously defined.

3.1.2. Riverine inputs

Because of the sampling system used in monitoring programmes (continuous decantation over 1 month) and because of the short ^{7}Be half life, ^{7}Be concentrations measured in SPM collected at Angers were corrected to take into account the radioactive decay during the sampling period, assuming that particulate ^{7}Be activity is constant over this period.

Although ^{7}Be flux to earth is known to be mainly governed by rainfall scavenging (Olsen et al., 1985; Turekian et al., 1983), no significant relationship was found between ^{7}Be in SPM and rainfall in Angers (results not shown); this could be explained by the fact that particulate ^{7}Be in Angers is controlled not only by local rainfall in Angers, but also by meteorological conditions on the Loire watershed. A simple statistical analysis of the 50 available data was performed. The mean particulate ^{7}Be activity at Angers is equal to 137 Bq kg^{-1} and the standard deviation is equal to 50 Bq kg^{-1}. As it was not possible to predict ^{7}Be riverine inputs by a relation 'particulate ^{7}Be = f(rainfall)', it was considered in further calculations that ^{7}Be riverine inputs are constant and equal to the mean value (i.e. 137 Bq kg^{-1}).

3.2. ^{7}Be concentrations in the turbidity maximum, bottom sediments and the sea

^{7}Be measurements on SPM collected in the estuarine turbidity maximum, in the sea and on bottom sediments are reported in Table 1. Experimental results show that:

- ^{7}Be activities in SPM collected in the turbidity maximum are much lower than those measured in SPM collected upstream of the estuary. Indeed, mean ^{7}Be activity in the freshwater solid phase is 137 Bq kg^{-1}, while particulate ^{7}Be activity measured in the estuary is in the range of 15–50 Bq kg^{-1}

(except for the experiment performed during a flood (25/02/1999), where the ^{7}Be activity was close to 100 Bq kg^{-1}). This experimental observation could be explained by different factors: (i) the transit time of particles through the estuary may be long enough to lead to an observable ^{7}Be radioactive decay; (ii) old unlabelled particles may be supplied to the turbidity maximum from the seaward delta front; (iii) desorption of ^{7}Be from particles along the salinity gradient could be a loss process in the mass balance; and (iv) a significant part of dissolved ^{7}Be may be removed from the estuary on the ebb tide.

- For a given sampling point, even if a high SPM gradient is observed over depth, no significant differences were observed between ^{7}Be measurements performed at different depths in the water column (except for the experiment performed during a flood (25/02/1999)). This observation could result from a homogeneous redistribution of particles over depth during successive deposition–resuspension cycles; the quantity of SPM showed a gradient over depth, but their quality could be homogeneously distributed.
- During a tidal cycle (08/07/1998 and 05/02/1999 experiments), ^{7}Be activities in SPM collected in the turbidity maximum do not significantly change.
- ^{7}Be activity in SPM sampled in the sea showed a lower value than those generally measured in SPM sampled in freshwater (108 Bq kg^{-1} in seawater and mean value of 137 Bq kg^{-1} in freshwater) and a higher value than those generally measured in SPM sampled in estuarine water.
- ^{7}Be activities in bottom sediments collected by the Ekman sediment sampler in the main river channel are generally close to those measured in the water column (except for the experiment performed during a flood (25/02/1999)). This result suggests that rapid deposition–resuspension cycles occur in such zones of the estuary, leading to a homogeneous ^{7}Be activity in SPM and in bottom sediments, respectively. In such a high-energy environment, pronounced sediment resuspension probably reintroduces deposited ^{7}Be back into the water column. In cores sampled in locations where sedimentation could be expected (out of the main river channel in full current), ^{7}Be was detectable only in the first 5 cm of each core. Sedimentation rates were estimated by measurements of ^{210}Pb$_{xs}$. Sedimentation rates were found to be in the range 0.4–4.2 cm year^{-1} (Fig. 3). Taking into account the radioactive half life of ^{7}Be, it is normal that ^{7}Be was detectable only in the first 5 cm of each core.

For sampling points at which filtered water was also collected, simultaneous measurements of ^{7}Be in the particulate and the dissolved phases, respectively, allow calculation of the distribution coefficient K_d (i.e. ratio between ^{7}Be activity in the particulate and the dissolved

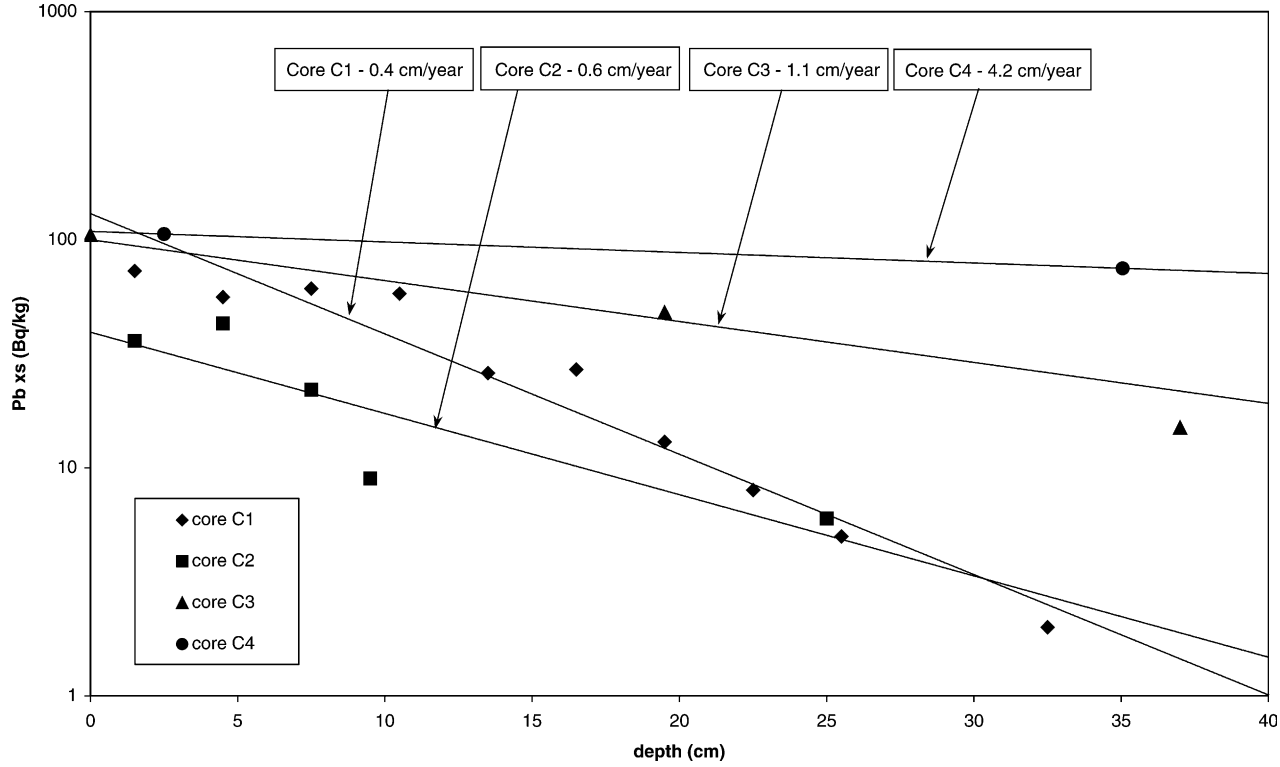

Fig. 3. ^{210}Pb$_{xs}$ in sediment cores sampled in the Loire estuary.

phases, respectively) (see Table 1). Experimental results give the following mean value: $K_d = 55{,}000\,l\,kg^{-1}$ (range 37,000–81,000 $l\,kg^{-1}$), which is coherent with other values reported in literature (Dibb & Rice, 1989; Olsen et al., 1986; Thomas, 1988).

4. Modeling section

4.1. Identification and selection of processes

Experimental results showed that ^{7}Be activities in SPM collected in the turbidity maximum are much lower than those measured in SPM collected upstream of the estuary. As mentioned previously, several processes could theoretically influence the activity of SPM in the water column of the turbidity maximum and contribute to explain experimental results.

4.1.1. ^{7}Be dilution from sea inputs

Theoretically, old unlabelled particles could be supplied to the turbidity maximum from the seaward delta front, and in such a case, ^{7}Be concentration in estuarine particles would decrease. However, some experimental observations suggest that such a process is not predominant: (i) ^{7}Be activity in SPM sampled in the sea (station out of influence of the estuary) showed higher value than those generally measured in SPM sampled in the turbidity maximum (108 Bq kg^{-1} in seawater and values ranging from 15 to 50 Bq kg^{-1} in estuarine water); (ii) according to previous studies (Migniot, 1993; Negrel, 1997), inputs of SPM from the sea to the estuary are much lower than those from the Loire river to the estuary. In particular, Negrel (1997) has investigated the chemical composition of bed sediments in the Loire estuary and their evolution between landward and seaward; according to Ca composition of estuarine sediments, he concluded that input of marine particles is not predominant. Consequently, in the present modelling work, it was considered that particulate fluxes of ^{7}Be from the sea to the estuary are negligible compared to those from the river. However, in future investigations, it would be useful to confirm such a hypothesis, in particular, by extending data sets on ^{7}Be activity in SPM sampled in the sea.

4.1.2. ^{7}Be sorption–desorption along the salinity gradient

Desorption of ^{7}Be from particles along the salinity gradient could theoretically be a loss process in the mass balance. However, some observations may suggest that such a process is not predominant. They are outlined as follows. (i) For the sampling campaigns occurring in summer 1998, K_d values were determined under various salinity conditions (from 0.1 to 17.7 $g\,l^{-1}$). Experimental results showed that K_d values remain in the same order of magnitude over a wide range of salinity and that they are not clearly related to salinity. Consequently, it may be suggested that salinity does not significantly influence the exchange processes of ^{7}Be at the water–particle interface. (ii) Batch experiments performed by Dibb and Rice (1989) to study the sorption kinetics of ^{7}Be onto estuarine particles did not show any clear relationship between K_d values and salinity. Consequently, in the frame of this study, it was assumed that no significant desorption occurs along the salinity gradient. However, further investigations should be necessary to confirm such a hypothesis, in particular, by performing batch experiments aiming at studying ^{7}Be sorption–desorption kinetics in controlled conditions, as it was done for some metals (Ciffroy, Garnier, & Benyahya, 2003).

4.1.3. Loss of dissolved ^{7}Be from the estuary

As hydrodynamics of water and particles are different in the estuary, a significant part of dissolved ^{7}Be may be removed from the estuary on the ebb tide. However, because of high K_d and SPM values in the estuary, ^{7}Be is predominantly present under particulate forms. Indeed, when calculations are performed with the mean K_d and SPM values ($K_d = 55{,}000\,l\,kg^{-1}$ and $SPM = 2.6\,g\,l^{-1}$), it can be shown that dissolved forms represent only 0.7% of total ^{7}Be. Consequently, losses of dissolved ^{7}Be from the estuary to the sea were neglected in the mass balance calculations. In future works, it will be useful to verify such a hypothesis by measuring ^{7}Be in water downstream of the estuary.

4.1.4. Residence time of particles in the turbidity maximum

It is well known that residence time of water and particles in macrotidal estuaries are very different. Consequently, the residence time of particles in the turbidity maximum could be long enough to lead to an observable ^{7}Be radioactive decay.

In summary, it has to be underlined that, in the model presented in this paper, some potential processes such as ^{7}Be dilution from sea inputs, ^{7}Be sorption–desorption along the salinity gradient and loss of dissolved ^{7}Be from the estuary were neglected. Even if some experimental data could justify such assumptions, it will be useful in future works to undertake specific measurements such as the following to better investigate these assumptions: batch experiments in controlled conditions to study the influence of salinity on ^{7}Be sorption–desorption; more extended quantification of ^{7}Be activity in sea particles and estimation of possible dilution effect; measurements of dissolved ^{7}Be expelled to the sea. However, considering available data sets, it was assumed in the present work that the behaviour of ^{7}Be in the turbidity maximum is predominantly governed by residence time of particles in this estuarine zone. Consequently, a model was developed to verify whether such an assumption can give consistent estimations of residence times of particles in the estuary.

4.2. Description of the model

The model considers that ^{7}Be in SPM of the turbidity maximum originates from:

- ^{7}Be particulate inputs from the river, and
- ^{7}Be deposited from rainfall. It is assumed that, after deposition, ^{7}Be originating from rainfall is homogeneously distributed over depth and that sorption may be described by an equilibrium distribution coefficient K_d.

In the model described subsequently, two associated times were defined for each particle: t_i is the date the particle entered the estuarine turbidity maximum and t_0 is the present date (more precisely, the sampling date). To simplify the further discussion, the difference $\theta_i = (t_0 - t_i)$ is arbitrarily called 'estuarine age' of the particle. ^{7}Be activity of a particle whose estuarine age is θ_i can be modelled as follows:

$$A_0(\theta_i) = A_{river}(\theta_i)\, e^{-\lambda\theta_i} + \sum_{t_i}^{t_0} A_{rain}(t) \times \frac{K_d}{h(1+K_d\overline{SPM})} e^{-\lambda(t_0-t)}\Delta t, \quad (2)$$

where

- $A_0(\theta_i)$ is ^{7}Be activity at instantaneous time t_0 of a particle whose age is θ_i (Bq kg^{-1}),
- $A_{river}(\theta_i)$ the ^{7}Be activity at time t_i of a particle whose age is θ_i (Bq kg^{-1}) (i.e. ^{7}Be activity when the particle entered the estuarine turbidity maximum),
- λ the ^{7}Be radioactive decay constant (j^{-1}),
- $A_{rain}(t)$ the ^{7}Be rainfall activity at time t (Bq m^{-2} day^{-1}) (here $t_i < t < t_0$),
- K_d the ^{7}Be distribution coefficient (m^3 kg^{-1}) (mean value = 55,0001 kg^{-1}),
- h the water depth at the sampling point (m) (mean value = 10 m),
- $\overline{SPM}$ the SPM concentration (averaged over depth) in the turbidity maximum (kg m^{-3}) (mean value = 2.6 kg m^{-3})
- Δt is the calculation time step (here $\Delta t = 1$ day).

The different terms of Eq. (2) describe the following processes:

- $A_{river}(\theta_i)\, e^{-\lambda\theta_i}$ describes the loss of ^{7}Be initially associated with the particle by radioactive decay (as mentioned previously, losses by desorption were neglected);
- $A_{rain}(t)(K_d/h(1 + K_d\overline{SPM}))e^{-\lambda(t_0-t)}$ describes sorption of ^{7}Beoriginating from rainfall onto the particle and continuous loss by radioactive decay.

Because of high values of K_d and SPM parameters, the expression $K_d/1 + K_d\overline{SPM}$ in Eq. (2) may be simplified in $1/\overline{SPM}$.

Furthermore, the repartition of particles over estuarine age classes is determined taking the following assumptions into consideration: probability for a particle to be evacuated from the estuary to the sea or to bottom sediment increases with particle estuarine age. Consequently, the number of particles in a given estuarine age class decreases with age. The only probability distribution function which allows us to obtain such conditions is an exponential distribution, this means that the ratio SPM_{θ_i}/SPM_{tot} is governed by the following relationship:

$$\frac{SPM_{\theta_i}}{SPM_{tot}} = \frac{e^{(-\theta_i/\beta)}}{\beta}, \quad (3)$$

where SPM_{θ_i} is the concentration of particles whose estuarine age is θ_i (kg m^{-3}) and SPM_{tot} the total SPM concentration (kg m^{-3}).

It has to be remembered that such a probability distribution implicitly simulates losses of particles and associated ^{7}Be from the water column of the turbidity maximum to bottom sediment and/or to the sea. In particular, the parameter β may be interpreted as follows. If constant SPM inputs from the river are assumed, the ratio $SPM_{\theta_i}/SPM_{\theta_i=0} = e^{(-\theta_i/\beta)}$ represents the fraction of SPM evacuated from the turbidity maximum by expulsion to sea or sedimentation during a period θ_i. Under these conditions, the half life of SPM in the turbidity maximum $T_{1/2}$ is the time θ_i for which $SPM_{\theta_i}/SPM_{\theta_i=0} = 1/2$. Consequently, it may be demonstrated that $T_{1/2} = \beta \ln 2$. Therefore, β may be assimilated to a half life in standard conditions. In the further discussion, results will be discussed in term of 'standard half life' $T_{1/2}$.

According to Eqs. (2) and (3), mean ^{7}Be particulate activity in a sample collected at t_0 can be estimated by the following relation:

$$A_0 = \sum_{\theta_i=0}^{\theta_i=\infty} \frac{\ln 2\, e^{(-\ln 2\theta_i/T_{1/2})}}{T_{1/2}} \times \left[A_{river}(\theta_i)\, e^{-\lambda\theta_i} + \sum_{t_0-\theta_i}^{t_0} A_{rain}(t) \frac{1}{h.\overline{SPM}} e^{-\lambda(t_0-t)}\Delta t \right]. \quad (4)$$

4.3. Input data of the model

To estimate the standard half life $T_{1/2}$ of particles in the turbidity maximum, it is necessary to know the following input data:

- The mean depth h in the zone investigated. In situ measurements reported in Table 1 show that maximum depth at each sampling point in the zone investigated is about 10 m (value chosen for further calculations).
- A mean SPM concentration in the turbidity maximum (noted $\overline{SPM}$ in Eq. (4)). For determining

a mean SPM concentration over depth, the relationship proposed by Migniot (1993) and describing the SPM gradient over depth in the Loire estuary is used. This relationship may be written as follows:

$$\mathrm{SPM}(h) = \mathrm{SPM}(h_0)\, e^{-K(h-h_0)}, \quad (5)$$

where

- SPM(h) is the SPM concentration at height h from the bottom ($g\,l^{-1}$),
- SPM(h_0) the SPM concentration at a reference height h_0 from the bottom ($g\,l^{-1}$)
- K is a constant (m^{-1}). When mud is present at the bottom interface, $K = 0.4\ m^{-1}$.

Consequently, the mean SPM concentration over depth is given by:

$$\mathrm{SPM} = \frac{\mathrm{SPM}(H)}{KH}(e^{KH} - 1), \quad (6)$$

where H is the total height from the bottom at the sampling point (m).

Surface samples have been chosen as reference levels for SPM concentration (i.e. $h_0 = H$). Furthermore, only samples collected during high or low tides are taken into account because mud is present at the bottom interface during theses tidal phases, and consequently, the value chosen for the constant K may be considered valid. On the other hand, during ebb and flood tides, a significant resuspension of mud is generally observed and the Eqs. (5) and (6) cannot be applied. Results reported in Table 1 allow estimation of a mean SPM concentration over depth: $\overline{\mathrm{SPM}} = 2.6\ g\,l^{-1}$ (range 1.3–4.6 $g\,l^{-1}$).

- ^{7}Be activity associated with particles entering the estuary (i.e. series of $A_{river}(\theta_i)$). As mentioned previously, it was considered that ^{7}Be riverine inputs are constant and equal to the mean value calculated taking into account 50 monthly measurements (i.e. 137 $Bq\,kg^{-1}$).
- ^{7}Be activity in rainfall (i.e. series of $A_{rain}(t)$). As mentioned previously, daily ^{7}Be inputs from atmospheric deposition were calculated using Eq. (1) previously defined and describing the correlation between ^{7}Be deposition and rainfall.

5. Results and discussion

5.1. Standard half life of particles in the turbidity maximum

Theoretical mean ^{7}Be activity in the particulate pool of each sample (i.e. $\overline{A_0}$) is calculated using Eq. (4); the "standard half" life $T_{1/2}$ is thus calibrated by comparison between the theoretical activity $\overline{A_0}$ and the activity actually measured (Table 2). Results show the following points:

Table 2
Standard half life of particles in the turbidity maximum

Date	$T_{1/2}$ (months)
03/07/1998	4.4 ± 1.2
08/07/1998	6.1 ± 1.1
15/09/1998	10.5 ± 3
05/02/1999	3.8 ± 0.6
25/02/1999 (surface)	0.7 ± 0.15
17/05/1999	4.7 ± 0.9
12/07/1999	8.4 ± 1
28/09/1999	6.8 ± 2.7

- The "standard half" life $T_{1/2}$ depends on the season. For summer conditions, it is generally situated in the range 6–10 months. For winter or spring conditions (05/02/1999 and 17/05/1999 experiments), it is significantly lower (in the range 4–5 months).
- For the experiment performed in a flood period (25/02/1999; flow rate = 3070 $m^3\,s^{-1}$), the standard half life $T_{1/2}$ of particles present at the water surface is very low (about 0.7 month for samples 5HS and 5ES). Consequently, it may be suggested that a great part of SPM present in the water column of the turbidity maximum have been expelled to the sea. However, the standard half life $T_{1/2}$ of particles present in the mud zone (5HM sample) is higher and shows that some particles remain trapped at the bottom interface.

5.2. Standard half life of particles in the turbidity maximum and flow rates

As mentioned previously, standard half life of particles in the turbidity maximum depends on the season, and as a consequence, probably on flow rates. To verify that standard half life of particles is actually related to flow rates, linear correlation coefficients between $T_{1/2}$ and the term $\sum_{i=1}^{i=n_i} Q_i$ were calculated, where Q_i is the Loire flow rate at the date corresponding to i days before sampling ($m^3\,s^{-1}$) and n_i is the number of days chosen for the calculation.

Relationships determined for $n_i = 30$, 45, 60 and 90 days are presented in Fig. 4. A rather good linear correlation was observed between the standard half life of particles $T_{1/2}$ and the sum of flow rates in the Loire river during 60 days before each sampling date. Consequently, it may be suggested that dynamics of particles within the Loire estuary is predominantly controlled by hydrological events occurring during 2-month periods.

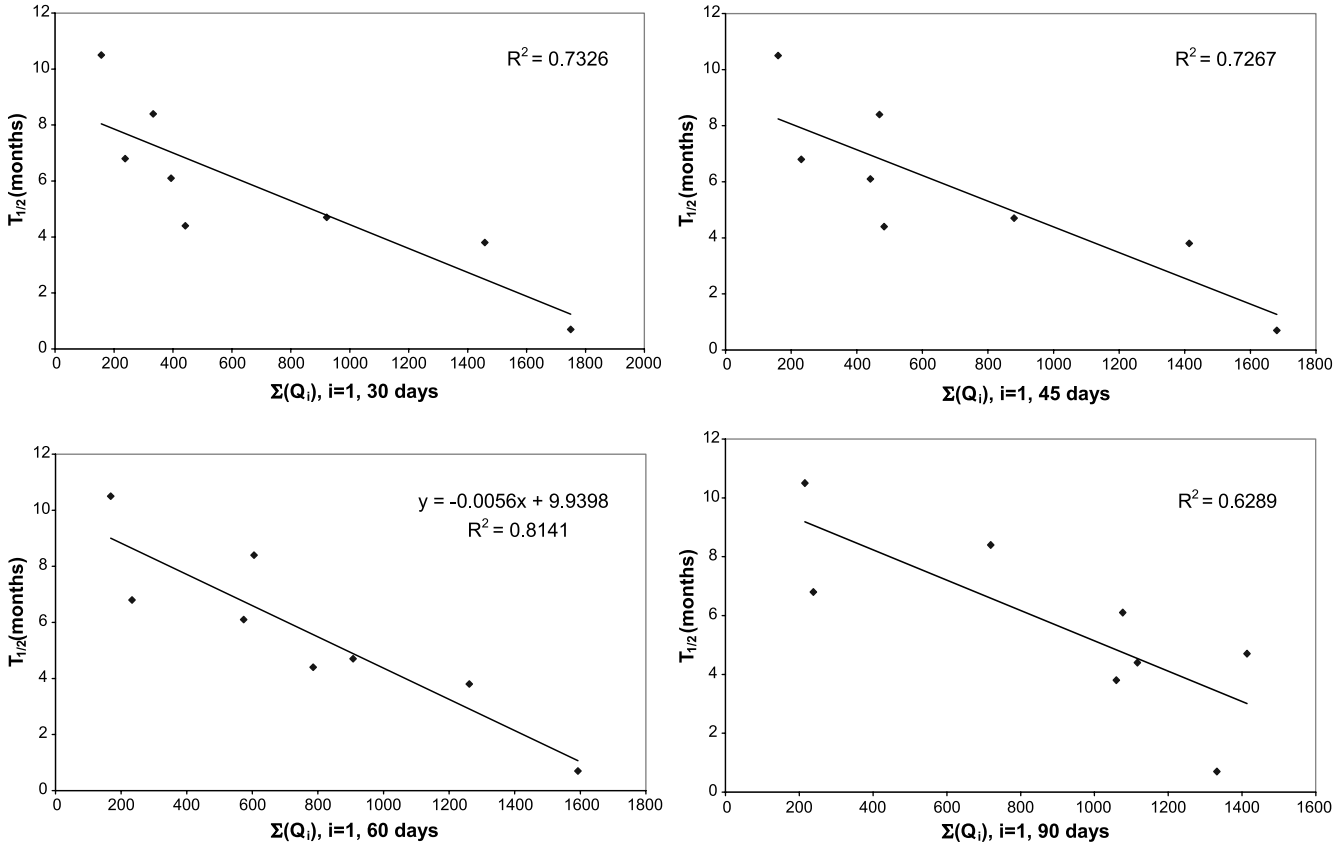

Fig. 4. Correlation between $T_{1/2}$ and the term $(\sum_{i=1}^{i=n_i} Q_i)$ (sum of flow rates during n_i days preceding the sampling date) ($n_i = 30$, 45, 60 or 90 days).

The best relationship which was determined between $T_{1/2}$ and flow rates in the Loire river is the following one:

$$T_{1/2} = 9.9 - 5.6 \times 10^{-3} \sum_{i=1}^{i=60} Q_i. \quad (7)$$

Such a relationship could be used for further applications aiming at studying the dynamics of particles and associated contaminants in the Loire estuary.

6. Applications

6.1. Kinetic evolution of the mass of particles within the turbidity maximum

Eq. (7), allowing the calculation of standard half life of particles in the turbidity maximum, may be used to simulate the kinetic evolution of the mass of particles within the turbidity maximum, according to the following relationship:

$$\frac{dM}{dt} = -\frac{\ln(2)}{T_{1/2}} M + Q_{riv} SPM_{riv} + M_{in\ situ} \quad (8)$$

where

M is the mass of particles within the turbidity maximum (g),

Q_{riv} the flow rate of the Loire river just upstream of the estuary (Montjean station) ($m^3 j^{-1}$),

SPM_{riv} the suspended particulate matter in the Loire river just upstream of the estuary (Montjean station) ($g m^{-3}$)

$M_{in\ situ}$ is the mass of particles produced in situ in the estuary (g).

The formation of new particles at low–high salinity contact by flocculation has been studied by several authors (Aloïsi et al., 1982; Martin et al., 1994; Negrel, 1997; Sholkovitz, 1976, 1978). In particular, Negrel (1997) has investigated the chemical composition of bed sediments in the Loire estuary and their evolution between landward and seaward. He concluded that about 50% of the sediments transported seaward is of terrestrial origin and that 50% are sediments produced in situ. Consequently, in the present study, two scenarios are considered: (i) there is no in situ production of particles in the estuary; and (ii) it is supposed that

the mass of particles produced in situ in the estuary ($M_{\text{in situ}}$) is equal to the inputs from the Loire river watershed.

A relationship $\text{SPM}_{\text{riv}} = f(Q_{\text{riv}})$ was previously calibrated using daily turbidity measurements in the Loire river over several years (Luck, 2001). The correlation coefficient between SPM (in winter) and flow rate obtained by the relationship proposed by Luck (2001) is 0.74. In particular, such a relationship properly simulates increase of SPM concentration in the Loire river during flood events. The relationship proposed by Luck (2001) was used to simulate the SPM in the Loire river just upstream of the estuary (SPM_{riv}) (Fig. 5). Kinetic evolution of $T_{1/2}$ was calculated using Eq. (7) (Fig. 5). The kinetic evolution of the mass of particles within the turbidity maximum, simulated by Eq. (8) and input data represented in Fig. 5, are shown in Fig. 6. Calculations show the followings:

- Except for flood periods, the mass of SPM present in the turbidity maximum is in the range 3×10^5–5×10^5 t when no in situ production of particles is considered and in the range 5×10^5–9×10^5 t when in situ production of particles is considered. These values are in good accordance with evaluations published elsewhere. According to Migniot (1993), the turbidity maximum zone represents a total mass of particles of about 5×10^5 t.
- A major part of particles are expelled to the sea during flood events (flow rate higher than 3000 $\text{m}^3\,\text{s}^{-1}$). For intermediate flow rates, calculated mass of particles expelled to the sea (or removed by sedimentation in estuarine depositional areas) may be compared to values published elsewhere. For example, the mass of particles expelled to the sea was evaluated during three in situ sampling campaigns performed in periods characterised by flow rates situated between 1000 and 2000 $\text{m}^3\,\text{s}^{-1}$ (Projet de centrale du Carnet. Etudes hydrosédimentaires de l'estuaire externe de la Loire. Mesures en nature, 1984). Values thus estimated were in the range 12,000–43,000 t day^{-1}. Calculated values for periods characterised by similar flow rates (for example, winter 1995–1996 and 1998–1999) are in the range 3000–13,000 t day^{-1} when no in situ production of particles is considered and in the range 6000–26,000 t day^{-1} when in situ production of particles is considered. Annual exportation loads by the Loire estuary were also estimated in several studies: Figueres et al. (1985) and Negrel (1997) estimated that the mean annual exportation load is 10^6 and 4.3×10^6 t year^{-1}, respectively. Calculations performed by the model presented here gave similar estimations (2.2 and 1.3 t year^{-1}) when in situ production of particles is considered or not, respectively. Such results appeared to be in good accordance with previous studies.

Consequently, such an application shows that standard half life estimated by ^{7}Be budgets may be an alternative method for studying the dynamics of particles within an macrotidal estuary.

6.2. ^{60}Co concentrations in the turbidity maximum

^{60}Co was measured in all the SPM samples collected in the estuarine Loire turbidity maximum (see list of samples in Table 1). Furthermore, total daily inputs of ^{60}Co to the Loire estuary were calculated by Luck (2001), taking into account daily releases from nuclear power plants situated along the Loire watershed and hydrodynamics of the Loire river. Results presented by Luck (2001) are given in Fig. 7. Such input data were used to estimate the particulate ^{60}Co activity in the turbidity maximum, according to the following relationship:

$$\frac{dA_p^{\text{Co-60}}}{dt} = -\frac{\ln(2)}{T_{1/2}} A_p^{\text{Co-60}} - \lambda A_p^{\text{Co-60}} + \text{Flux}_{\text{riv}}^{\text{Co-60}} \left(1 - \frac{1}{1 + K_d\,\text{SPM}}\right) \frac{1}{M},$$

where

$A_p^{\text{Co-60}}$	is the particulate ^{60}Co activity in the turbidity maximum (Bq kg^{-1}),
λ	the ^{60}Co radioactive half life (s^{-1}),
$\text{Flux}_{\text{riv}}^{\text{Co-60}}$	the total ^{60}Co inputs from the Loire watershed to the Loire estuary (Bq s^{-1}),
M	the mass of particles within the turbidity maximum (g),
SPM	the suspended particulate matter in the turbidity maximum (g m^{-3})
K_d	is the ^{60}Co distribution coefficient in the turbidity maximum ($\text{m}^{-3}\,\text{g}^{-1}$). K_d values specifically determined for the Loire estuary by Ciffroy et al. (2003) were used in the present model.

Results of the model are presented in Fig. 7 and compared to measurements performed in samples collected in the turbidity maximum. It was observed that calculated values are close to measured values. Consequently, such an application shows that the method proposed in this study could be used for estimating contaminant concentrations in the estuarine turbidity maximum.

7. Conclusions

The present work has shown that ^{7}Be could be an efficient time sensitive tracer of particles in a macrotidal estuary. Indeed, it was experimentally observed that ^{7}Be activities in SPM collected in the turbidity maximum are much lower than those measured in SPM

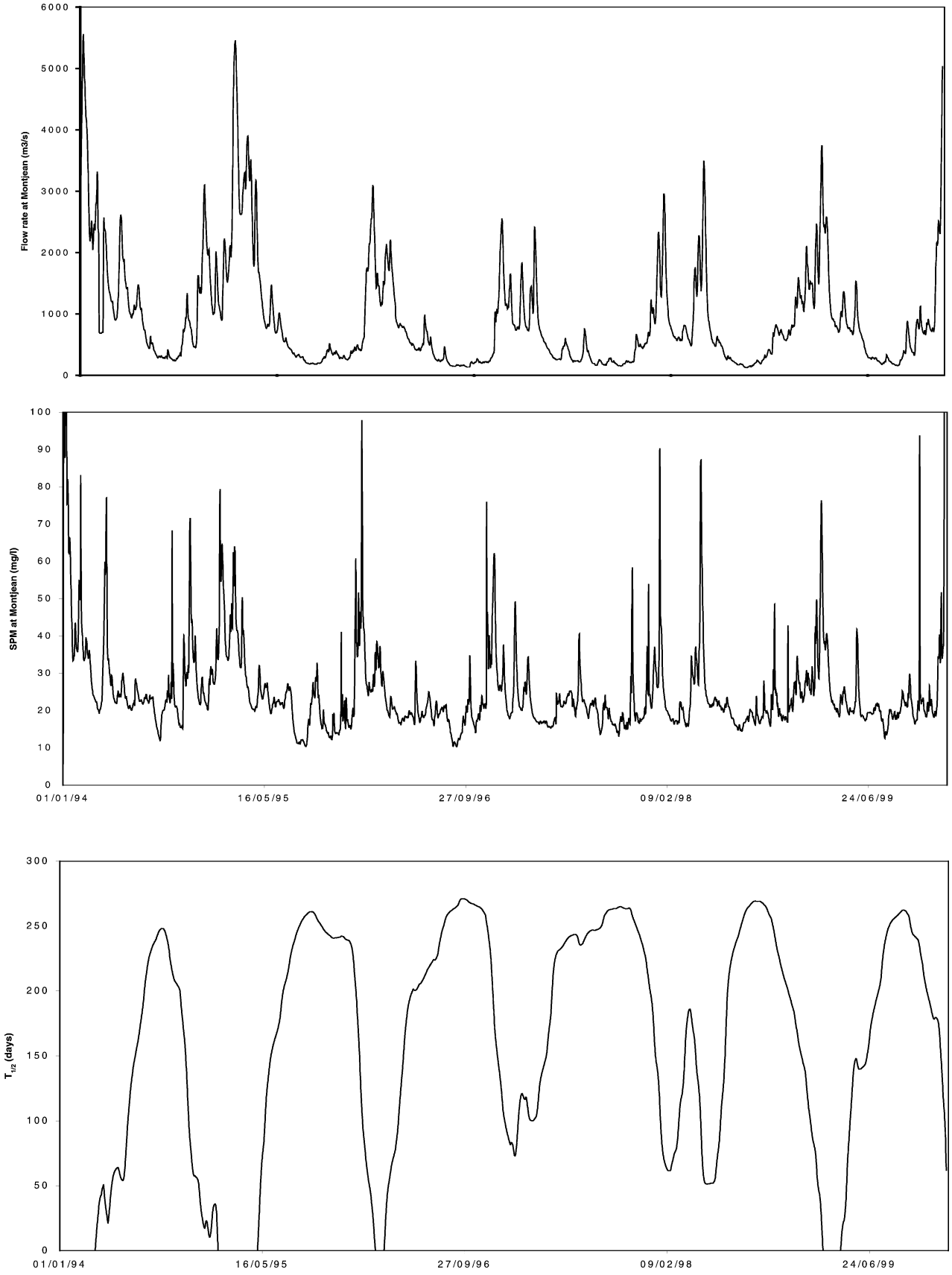

Fig. 5. Flow rate, SPM in the Loire river, and standard half life of particles ($T_{1/2}$) in the turbidity maximum.

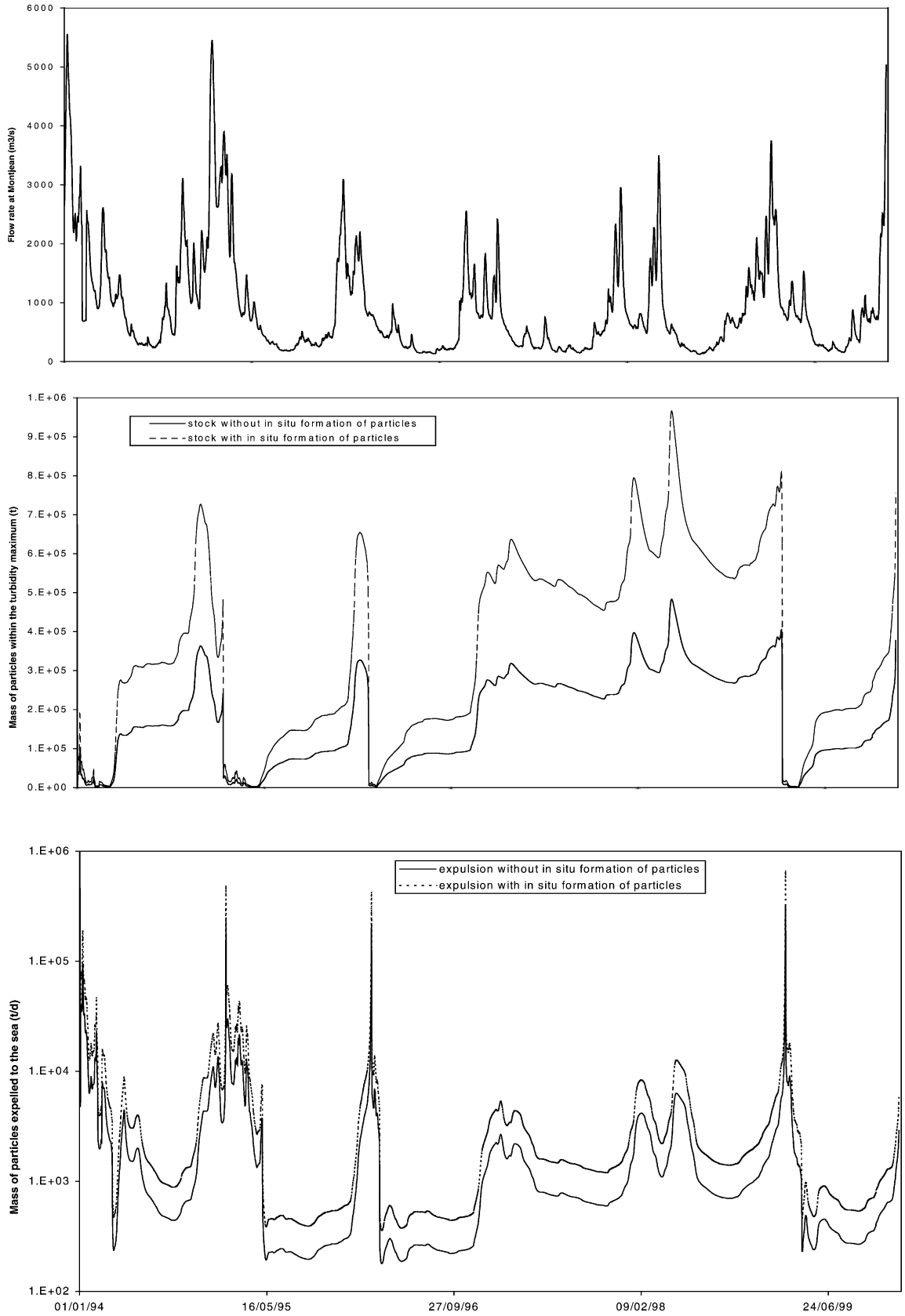

Fig. 6. Flow rate in the Loire river, mass of particles within the turbidity maximum, and mass of particles daily expelled to the sea.

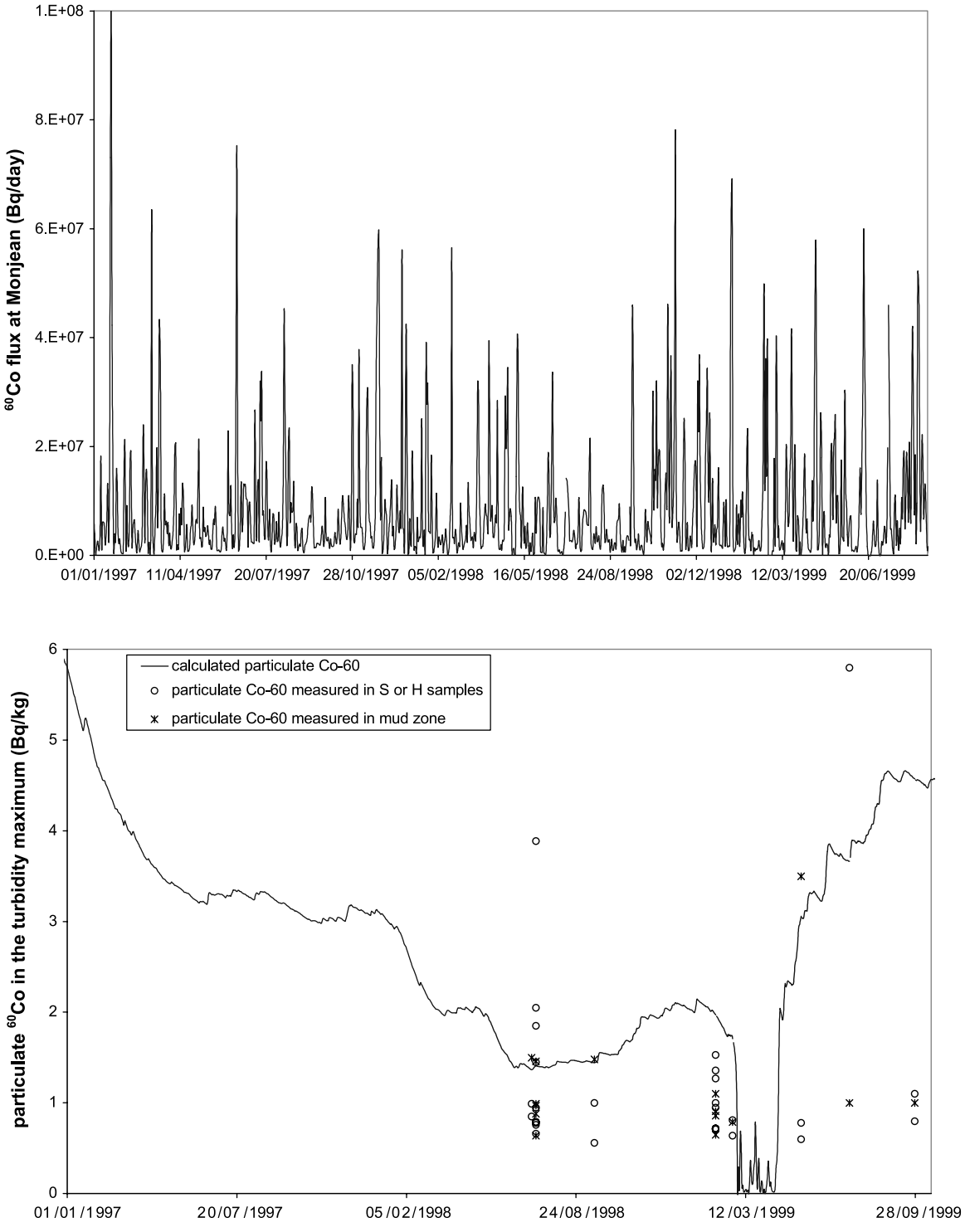

Fig. 7. Inputs of ^{60}Co to the Loire estuary (Bq day^{-1}), and particulate ^{60}Co activity in the turbidity maximum (Bq kg^{-1}) are shown.

collected upstream of the estuary. Such an experimental observation could be explained by different factors: (1) the transit time of particles through the estuary may be long enough to lead to an observable ^{7}Be radioactive decay; (2) old unlabelled particles may be supplied to the turbidity maximum from the seaward delta front; (3) desorption of ^{7}Be from particles along the salinity gradient could be a loss process in the mass balance; and (4) a significant part of dissolved ^{7}Be may be removed from the estuary on the ebb tide. In this study, the first factor was considered (transit time of particles) as predominant for explaining the behaviour of ^{7}Be in the

Loire estuary and the other ones could be neglected. Even if some experimental observations showed that such a choice is reasonable, it is obvious that such assumptions should be better investigated in future works.

Considering such assumptions, experimental measurements of ^{7}Be could be quantitatively interpreted by using a mathematical model. This model took into account the following processes, which can influence ^{7}Be activity in the estuarine turbidity maximum: inputs from river particles and inputs from atmospheric deposition, radioactive decay, sorption onto particles. Such a model allowed the calculation of standard half life of particles in the estuary. Results showed that the half life of particles in the turbidity maximum depends on the season: for summer conditions, it is generally situated in the range 6–10 months, while for winter or spring conditions, it is significantly lower (in the range 4–5 months). For the experiment performed during a flood, the standard half life of particles present at the water surface is very low (about 0.7 month). Furthermore, it was observed a rather good linear correlation between the standard half life of particles $T_{1/2}$ and the sum of flow rates in the Loire river during 60 days before each sampling date. The kinetic evolution of the mass of particles within the turbidity maximum could be estimated by this method and appeared to be consistent with previous studies. Moreover, the method proposed in this study could be properly used for estimating ^{60}Co concentrations in the estuarine turbidity maximum. Consequently, this study showed that the use of ^{7}Be is a promising tool for investigating the dynamics of particles in a macrotidal estuary. However, future works should improve this approach by investigating exhaustively all the processes which could influence the activity of ^{7}Be in water and particles.

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