

Particulate organic carbon in the estuarine turbidity maxima of the Gironde, Loire and Seine estuaries: origin and lability

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Abstract A study of the particulate organic carbon (POC) in the estuarine turbidity maxima (ETMs) of the three major French macrotidal estuaries shows that the average contents are 1.5, 3.3 and 3.1% (expressed in % of dry suspended sediment) in the Gironde, Loire and Seine Estuaries, respectively. There is no seasonal variation of POC contents in the Gironde Estuary, whereas, they often increase in the Loire and the Seine Estuaries in spring and summer. The lability of the

estuarine particulate organic matter was estimated by two analyses: 1/labile organic matter was measured as the organic carbon loss during incubation tests over one month; 2/ the hydrolysable organic fraction was determined after 6N HCl digestion. The organic fractions of the ETMs are mainly refractory. Any increase in the amount of POC as compared to the background levels (cited above) is always correlated to an increase of organic matter lability. The yearly average fluvial contributions by various particulate organic pools (soil and litter organic matter; organic matter of phytoplanktonic and human origin) that enter the three estuaries were quantified. In the Garonne River, soil and litter are the major (90%) POC sources. In the Loire system, due to the eutrophication of the river water, phytoplankton contributes up to 50% of the total POC load. In the Seine river, soil and litter contribute 70% of the total POC input; POC of human origin is also significant (10%), due to the impact of the City of Paris (10 million inhabitants). The lability of the different types of organic matter ranks as follows: phytoplankton ~litter > human-origin organic matter > > soil. By combining the POC budgets and the lability of each type of organic fraction, it was possible to explain why the POC of the three ETMs is different and characterizes its refractory *vs.* labile nature.

Guest editors: R. Lafite, J. Garnier & V. De Jonge
Consequences of estuarine management on hydrodynamics and ecological functioning

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Keywords Estuaries · Particulate organic matter · Source · Lability

Introduction

Annually, world-rivers transport large quantities of carbon to coastal seas (0.9 GtC of which 40% is organic and 60% inorganic; Meybeck, 1993). Estuaries are major land-ocean interfaces for this material before it reaches the coastal zone and the ocean. In estuaries, riverine particulate organic carbon (POC) is subjected to sedimentation, retention and net mineralization, as reported for several estuaries world-wide (Ittekkot & Llane, 1991; Zhang et al., 1998; Abril et al., 1999).

In European macrotidal estuaries, mineralization is particularly strong: due to tidal influence, residence times for both water and suspended matter are long, which allows an estuarine turbidity maximum (ETM) to develop. In this area, photosynthesis is strongly limited by light availability (Cole et al., 1992; Irigoien & Castel, 1997; Garnier et al., 2001) and bacterial activity is very high (Heip et al., 1995): heterotrophy by far dominates autotrophy (Billen et al., 1995; Gattuso et al., 1998; Goosen et al., 1999; Garnier et al., 2001, 2004).

The purpose of this paper, which focuses on the particulate organic fraction of the ETMs of the three major macrotidal French estuaries (the Gironde, the Loire and the Seine Estuaries), is to answer the following questions:

- What are the levels of POC in these ETMs? What are the drivers of the seasonal variations?
- What proportion of the POC is labile (i.e. biodegradable or bioavailable)?
- Can the quantity, origin and/or lability of the fluvial organic matter reaching the estuaries explain the contents and fate of the POC in the ETMs?

Materials and methods

Study area

The map and the studied sections of the estuaries are shown in Fig. 1 and their main hydro-sedi-

mentological characteristics are summarized in Table 1.

These data have already been described in details by many authors (see for the Gironde Estuary: Allen (1972), Allen et al. (1977), Castaing (1981), Jouanneau & Latouche (1981); for the Loire Estuary: Galenne (1974), Le Douarec (1978), Manickam (1982), Migniot (1991); for the Seine Estuary: Avoine (1981), Brenon (1997) and Guézenc et al. (1999)).

Briefly, the Gironde is the largest European estuary in terms of surface area and water discharge and one of the less human-impacted in Europe (Abril et al., 2002). Due to a strong marine influence, the residence times of water and total suspended sediment (TSS) are long, leading to a well-developed ETM, with high TSS concentrations (from several hundred mg l⁻¹ to several g l⁻¹), which lead to a total mass of 4–5 million tons (Table 1).

The Loire Estuary is characterized by a smaller surface area (despite a larger catchment) and greater human pressure than the Gironde Estuary (Table 1). Moreover, the Loire is known as one of the most eutrophic rivers in Europe (Meybeck et al., 1988; Lemaire et al., 2002b; Moatar & Meybeck, 2005). As the marine influence is weaker than in the Gironde Estuary, the residence times of water and TSS are also shorter. Moreover, the rapid oxygen depletion of the water in the summer is particularly marked in the Loire Estuary (Thouvenin et al., 1994), although it occurs in all three studied estuaries; resuspension of anoxic “fluid mud” at spring tides, i.e. benthic layers with a high TSS concentration (one to several hundred g l⁻¹), appears to be the main reason for the hypoxia (Abril et al., 2003, 2004)), as reported for several other macrotidal estuaries (Parker et al., 1994; Uncles et al., 1998).

The Seine Estuary is the smallest of the three estuaries (Table 1). Owing to the presence of big cities (including Paris), the Seine Estuary is subjected to the greatest human impact (Servais & Garnier, 1993; Idlafkih, 1998) and to fluvial inputs from the eutrophic Seine River (Garnier et al., 1995, 1998). Due to a moderate marine influence, the residence times of the water are of the same order as in the Loire Estuary. Those of the TSS in the Seine Estuary are much shorter

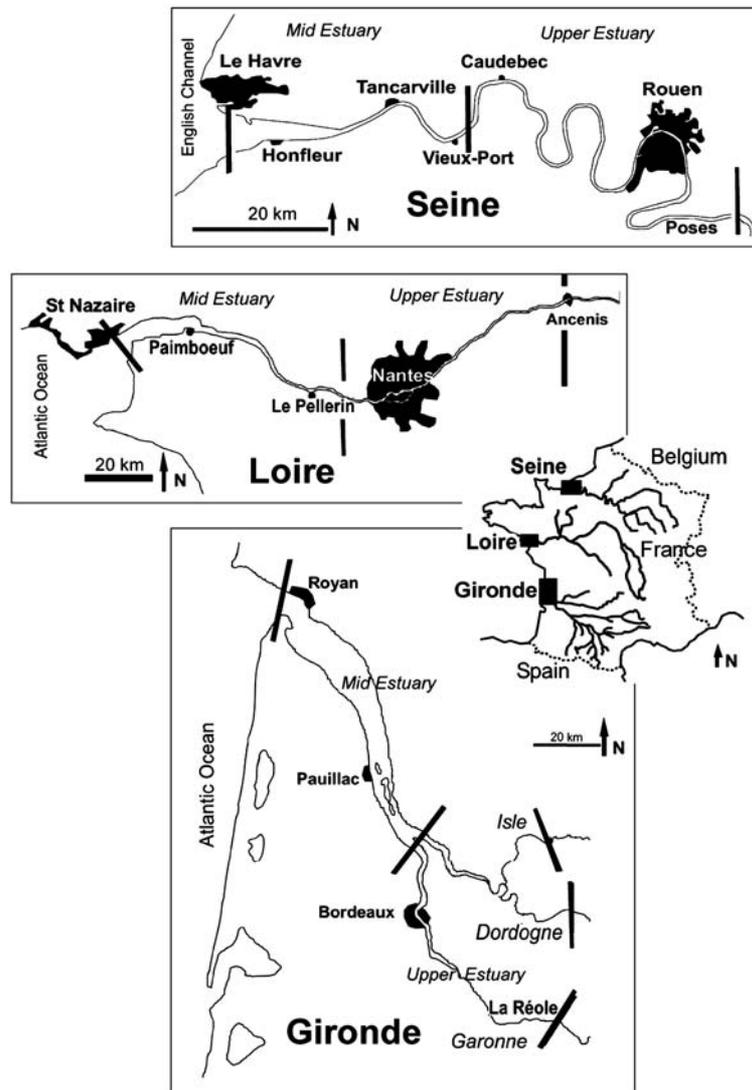


Fig. 1 Map of the Gironde, Loire and Seine estuaries. From upstream to downstream, black lines underline the limits of the tidal influence and of the salt intrusion

(1–3 months, Table 1), and fluvial TSS inputs are moderate, which creates a relatively small ETM in the estuary. Finally, a typical feature characterizing the Seine Estuary is a long high tide slack (up to 4 h), which leads to a temporary sedimentation of TSS and a net decrease of TSS concentrations in the surface waters.

Sampling and database

The data used in this paper were obtained from a number of different programs (Table 2). They

cover the four seasons over the last two decades in the three estuaries (ETM areas) and at their reference river stations (inlet of the estuaries at the upper limit of the tidal influence) with the exception of the daily TSS data from the Loire River dating from the fifties and sixties.

In the ETMs, the water was differently sampled was differently in the three estuaries. A 7-years monthly sampling survey was carried out in the Gironde Estuary. Three stations were monitored most of the time, but occasionally only two in the winter. In the Loire and the

Table 1 Main characteristics of the Gironde, Loire and Seine Estuaries

		Gironde	Loire	Seine
Surface area	River basin (km ²)	71000 (11)	115000 (11)	79000 (11)
	Estuary * (km ²)	442 (2)	102 (2)	50 (2)
Population density	River basin (inhab. km ⁻²)	61 (11)	71 (11)	203 (11)
	Estuary (inhab. km ⁻²)	129 (11)	166 (11)	168 (11)
River discharge	Instantaneous (m ³ s ⁻¹)	1000 (1)	830 (1)	500 (1)
	Specific (L s ⁻¹ km ⁻²)	14.1 (1)	7.2 (1)	6.3 (1)
Inhabitants/discharge ratio	(inhab./m ³ s ⁻¹)	4300 (1)	9800 (1)	32000 (1)
Tidal prism	Spring tide (m ³)	2 × 10 ⁹ (3)	300 × 10 ⁶ (4)	380 × 10 ⁶ (5)
	Neap tide (m ³)	1.1 × 10 ⁹ (3)	140 × 10 ⁶ (4)	200 × 10 ⁶ (5)
Tidal prism / fluvial in-flow ratio		35 (1)	6 (1)	13 (1)
Tidal range	(m)	4 (2)	4 (2)	5 (2)
Residence time	Water (months)	0.5 to 3 (3)	≤0.5 (12)	≤0.5 (12)
	Particles (months)	12 to 24 (3)	4 to 10 (6)	1 to 3 (7)
Estuarine turbidity maxima	(tons)	4 to 5 × 10 ⁶ (8)	1.5 × 10 ⁶ (9)	0.4 × 10 ⁶ (10)

*Only the inner estuary surface; (1) this work; (2) Abril (1999); (3) Jouanneau & Latouche (1981); (4) Laboratoire de Géologie Marine et Appliquée (1978); (5) Brenon (1997); (6) Ciffroy et al. (2003); (7) Bonté et al. (2000); (8) Allen (1972); (9) AELB Report (2000); (10) Dupont et al. (1996); (11) Ignasse et al. (2002); (12) Migniot (1991)

Seine Estuaries, respectively 13 and 6 cruises, were made in the ETMs throughout the four seasons, including a variable number of stations. In this study, only samples with a TSS concentration higher than 200 mg l⁻¹ were considered as ETM samples. Water samples were taken with a Niskin bottle at a depth of 1 m. Aliquots were filtered through pre-combusted (500°C, 4 h) and pre-weighed glass-fiber filters (Whatman GF/F) in order to measure the TSS and POC contents. For some selected samples, two other aliquots were used for incubation tests and 6N HCl digestions.

Regarding river data, the DIREN (French Department for the Environment) provided daily water discharges, covering at least 10 years for the three estuaries. Daily or weekly TSS measurements were obtained in the Garonne-Dordogne Rivers over 8 years (Table 2), by filtration of water samples (Coynel et al., 2004). Because no detailed survey currently exists for the Loire river inputs, Berthois' data set (e.g., Berthois 1963, 1972) was used and a discharge versus TSS relationship (Migniot, 1991) was established. Daily TSS concentrations were thus calculated for recent water discharges (Table 2). In the Seine River, daily TSS data were provided by the SNS (Service de Navigation de la Seine) over 4 years (Table 2) by direct field OBS (Optical

Backscattering Sensor) measurements after calibration with data obtained by filtration.

In the Garonne and Dordogne rivers, the POC contents were measured weekly, and even daily during the high-water periods (Veyssey et al., 1996, 1999). In the Loire River, POC data were obtained from 13 cruises and from twice-monthly sampling that started in January 2004 (unpublished data, not cited in Table 2). The POC data were interpolated by using POC% versus TSS curves established for the Loire River (Salot et al., 1984; Meybeck et al., 1988; Relexans et al., 1988). In addition, owing to the large contribution by phytoplankton in the Loire River, monthly chlorophyll *a* data, over 12 years (AELB, Agence de l'Eau Loire-Bretagne, a French water authority agency), were used; chlorophyll *a* concentrations were multiplied by 35 to obtain the algal POC contents. In the Seine River, POC data were regularly obtained from seasonal cruises and additional twice-monthly sampling with the help of the SNS. Some data from the PIREN SEINE programme (Servais et al., 1998; Idlafkih, 1998) were used as well.

The labile and hydrolysable fraction of the different fluvial organic matter pools (soil, litter and phytoplanktonic organic fraction) were studied on some selected samples from the Garonne and Loire Rivers at freshwater, non-tidal, stations (Lin, 1988; Lin & Etcheber, 1994):

Table 2 List of the programmes dealing with the data used in this study

Estuarine data	Gironde Estuary			Loire Estuary			Seine Estuary			
	TSS & POC%	Programme	Sampling	Date	Programme	Sampling	Date	Programme	Sampling	Date
		- SOMLIT	S.S.	1997–2003	- GRECO-ICO	10 S.C.	1982–1985	- SAV2	6 S.C.	2002–2004
					- BIOGEST	1 S.C.	1998			
					- FLORE	2 S.C.	2003			
Fluvial data	Q	- DIREN	S.S.	1959–2000	- DIREN	S.S.	1990–2003	- DIREN	S.S.	1994–2003
	TSS	- DBT	S.S.	1989–1992	- Berthois data	S.S.	1953–1967	- SNS	S.S.	1994, 2001, 2002 & 2003
		- GIS-ECOBAG	S.S.	1994–1997						
		- DBT	S.S.	1989–1992	- AELB *	S.S.	1987–1999	- PIREN SEINE	S.S.	1993–1996
		- GIS-ECOBAG	S.S.	1994–1997	- GRECO-ICO	10 S.C.	1982–1985	- SAV ₂	S.C.	2002–2004
	POC %				- BIOGEST	1 S.C.	1998			
					- FLORE	2 S.C.	2003			

S.S.: Seasonal Survey; S.C.: Seasonal Cruises; * chlorophyll *a* data (Agence de l'Eau Loire-Bretagne); Q: discharge values; TSS: Total Suspended Sediment; POC%: Particulate Organic Carbon content; SNS: Service de Navigation de la Seine

- Two late-winter (February) samples from the Garonne River, where soil OM was predominant (Veyssey et al., 1999);
- One early-autumn (October) sample from the Garonne River, rich in riparian litter organic matter (Veyssey et al., 1999);
- Three fluvial samples from the Garonne and Loire Rivers and a marine sample from the lower Loire estuary, in spring and summer, high in phytoplankton content (Relexans & Etcheber, 1982).

In addition, three samples of treated water from a sewage treatment plant “Lyonnaise des Eaux” located in Bordeaux, were studied for lability.

Determination of organic carbon contents and their labile fraction

After filtration, Whatman GF/F filters were dried in an oven at 50°C and weighed for determination of TSS. The organic carbon content was determined on the dry filters by combustion in an LECO CS 125 analyzer (Abril et al., 2002). Samples were acidified in crucibles with 2N HCl to remove carbonates, then dried at 60°C to remove inorganic C and most of the remaining acid and water. The analyses were performed by direct combustion in an induction furnace and the CO₂ formed was determined quantitatively by infrared absorption.

An hydrolysis of samples by 6N HCl (110°C, 16 h) was performed in a Pyrex screw-cap tube with a Teflon liner (Buscail et al., 1990, 1995; Etcheber et al., 1999). Hydrolysable organic carbon (HOC), was estimated as the difference between the POC and the residual organic carbon (ROC) after 6N hydrolysis, expressed as a percentage of POC (% HOC).

The lability of POC entering the estuaries or associated with the ETMs was also estimated by a second approach, which consisted in classical in vitro incubations (Servais et al., 1987, 1995; Lin & Etcheber, 1994; Lemaire et al., 2002b). Each water sample was incubated for 30 days, at constant temperature (20°C) and gently stirred to maintain oxygenation. The POC contents were

measured in aliquots sampled throughout the duration of the incubations. The labile organic carbon (LOC) was considered as the loss of POC between the first and the last day of the incubation experiment and expressed as a percentage of the total POC (% LOC).

Fluvial POC inputs and origin

For each river entering the three estuaries, an annual average, solid discharge-weighted POC content in riverine suspension (noted POC%* and expressed as a percentage of TSS) was calculated, according to:

$$\text{POC}\%^* = \frac{\sum_{i=1}^{365} (Q_i \times [\text{TSS}]_i \times [\text{POC}\%]_i)}{\sum_{i=1}^{365} (Q_i \times [\text{TSS}]_i)}$$

The calculation was performed using the daily discharge, the daily TSS concentrations (field data in the Gironde and Seine and interpolated data in the Loire), and the POC% versus TSS relationships for each river. The origins of the different particulate organic matter pools were estimated by the same procedure as described by Veyssy et al. (1999) and Coynel et al. (2004, 2005) in the Garonne and Pyrenean Rivers. After a compilation and analysis of POC and TSS data from numerous rivers over a full hydrological year, the relative contribution from four different POC inputs were differentiated. The autochthonous phytoplanktonic contribution was dominant during low-water stages and low TSS, consistent with a chlorophyll *a*/POC ratio close to 35 (Relexans & Etcheber, 1982; Dessery et al., 1984; Garnier et al., 1998). The allochthonous (terrestrial) POC dominated during high and medium water discharges and TSS concentrations (Lin, 1988). Based on the progressive decrease in POC content during the successive floods, generally observed from October to February in these rivers, terrestrial POC can be separated into two fractions (Veyssy et al., 1999): the “organo-mineral” or POC, associated with the soil mineral matrix corresponds to a threshold POC content during late winter; the POC content due to riparian litter, which is quantified as the

additional POC content above the late-winter threshold and is high from October to January (Veyssy et al., 1999). Finally, in the Seine River, where domestic (sewage) loads are significant, the human-origin POC input was estimated as the difference in concentrations upstream and downstream of the city of Paris at high and low water (Idlafkih, 1998).

Results

POC contents in the TSS from the ETMs of the Gironde, Loire and Seine estuaries

For the ETM samples, characterized by TSS concentrations above 200 mg l⁻¹, the mean annual and seasonal organic carbon contents are shown in Fig. 2. The minima, maxima, lower quartiles and upper quartiles of annual and seasonal POC contents illustrate the inter-seasonal and intra-seasonal variability. The Kruskal–Wallis test (non-parametric one-way ANOVA test) was used to compare the relative variability of these averages. The null hypothesis H₀ signifies that all population means are equal, at least one mean being different under the alternative hypothesis H₁. A *p*-value of 0.05 was considered as statistically significant, below this value the null hypothesis was rejected.

In the Gironde Estuary, in any season, the POC contents did not vary significantly around 1.5% (the standard deviation is 0.116). The same constant value in the Gironde ETM has been reported in earlier studies with much fewer data (Latouche et al., 1982; Fontugne & Jouanneau, 1987; Laane et al., 1987), which reveals a long-term stability. Moreover, the intra-seasonal variability was extremely low, lower and upper quartiles always ranged between 1.4 and 1.6%. With *p* = 0.1359, the Kruskal–Wallis test confirms this stability by accepting H₀.

In the Loire Estuary, the annual mean POC content was 3.3%, close to values cited in the literature (Salot et al., 1984; Relexans & Etcheber, 1986; Meybeck et al., 1988; Relexans et al., 1988; Abril et al., 2002). With a standard deviation of 0.3, the variability associated with this

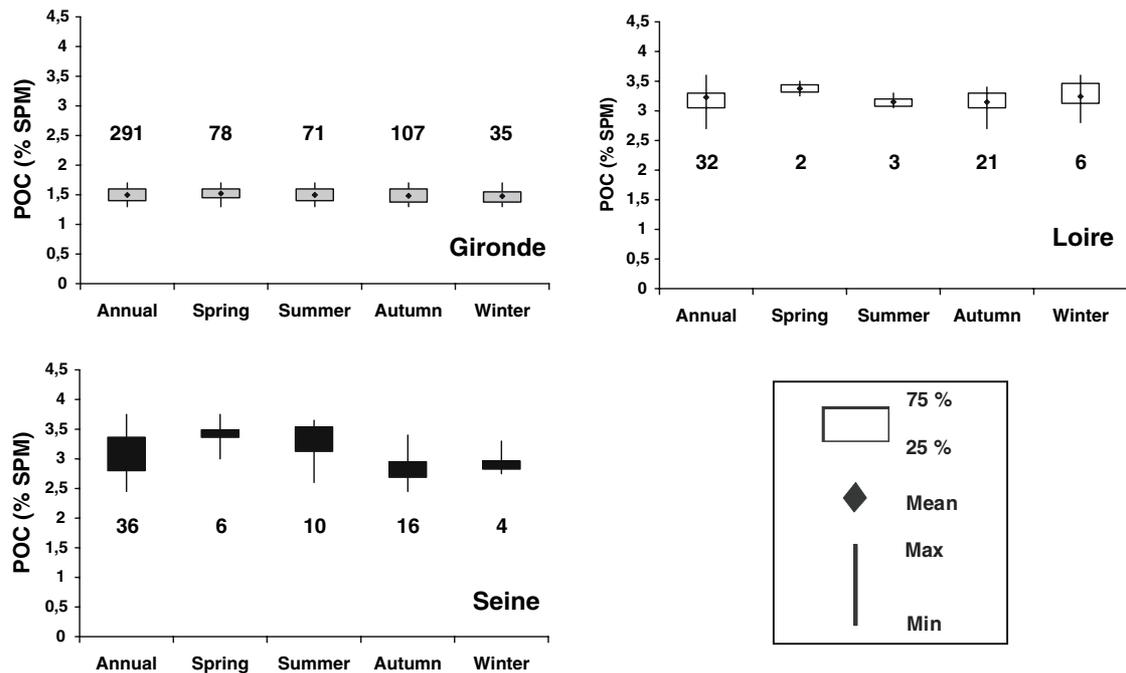


Fig. 2 Annual and seasonal POC contents (expressed in % of TSS) in the ETM of the Gironde, Loire and Seine estuaries. The number of the samples analyzed are

indicated on the graphs. Statistical parameters are shown in a legend box as relative variabilities (%), means, minima and maxima and lower and upper quartiles

annual mean was more pronounced than in the Gironde Estuary. However, the differences between seasons in the Loire ETM were not significant ($p = 0.54$); the number of samples taken in spring and summer was too small to adequately describe the seasonal variability. Some high values (up to 4.5–5%) were observed in June 1982 and September 1998. However they were restricted to the upstream limit of the ETM around the City of Nantes, an area strongly influenced by upstream phytoplanktonic bloom accumulation and by local factories. Therefore, they have purposely not been taken into account for the estimate of summer mean.

In the Seine Estuary, the annual mean POC content was 3.1%. With a standard deviation of 0.4, the variability of this annual mean was similar to that found for the Loire Estuary. However, the Kruskal–Wallis test ($p = 0.0042$) rejected H_0 . The seasonal variations were significant, the highest values occurring in spring and summer. Furthermore, in the Seine Estuary, as soon as surface TSS

concentrations decreased at tidal slacks (settling of suspended particles), organic carbon contents increased; at intermediate TSS concentrations ($100\text{--}200\text{ mg l}^{-1}$) the POC was always significantly higher than the annual mean of 3.1%.

The mean annual POC contents in the Loire and Seine estuaries were also compared; the annual means were not statistically different ($p = 0.19$). The POC contents in the Seine and Loire ETMs were in the same range as in several other European estuaries (2–4%, Uncles et al., 2000; Abril et al., 2002), whereas the POC content in the Gironde Estuary was significantly below this range.

Lability of organic matter in the ETMs of the Gironde, Loire and Seine

In the Gironde ETM, among the eight samples treated by 6N HCl digestion, seven showed HOC percentages (HOC/POC) lower than 5.7% (Table 3). Only one ETM sample had a HOC

Table 3 Results of organic carbon content and lability obtained from 6N HCl digestions and of the incubation tests

Sample	Estuary	Month	Acid treatment		Incubation					
			POC content (% of TSS)	ROC (HCl 6N) (% of TSS)	HOC (% of POC)	T ₀ (% of TSS)	T ₀₊₃₀ (% of TSS)	LOC (% of POC)		
ETM	Gironde	February	1.56	1.54	1.3	1.54	1.53	0.6		
		February	1.62	1.57	3.1	1.58	1.54	2.5		
		May	1.72	1.48	13.9	1.62	1.52	6.2		
		June	1.64	1.58	3.7	1.63	1.57	3.7		
		July	1.53	1.51	1.3	1.46	1.42	2.7		
		July	1.58	1.49	5.7	1.51	1.53	0.0		
		October	1.52	1.53	0.0	1.49	1.47	1.3		
		November	1.49	1.47	1.4	1.57	1.52	3.2		
	Loire	February	4.15	3.6	13.3	4.05	3.65	9.9		
		May	3.35	3.25	3.0	3.25	3.20	1.5		
		September	3.2	3.1	3.1	3.35	3.20	4.5		
		September	3.05	2.9	4.9	2.90	2.80	3.4		
		February	4.55	4.1	9.9	4.85	4.60	5.2		
	Seine	February	3.3	3.15	4.5	3.20	3.25	0.0		
		August	2.62	2.49	5.0	–	–	–		
		September	3.35	3	10.4	–	–	–		
		October	2.79	2.93	0	2.69	2.63	2.2		
February		2.83	2.81	0.7	2.83	2.78	1.8			
March		3.46	3.03	12.4	3.48	3.04	12.6			
May		7.25	3.00	58.6	7.25	3.80	47.6			
May		7.35	3.10	57.8	7.35	–	–			
Fluvial	Gironde	Soil	February	5.2	3.7	28.8	5.10	3.95	22.5	
		February	2.32	1.72	25.9	2.43	1.94	20.2		
		Litter	October	23.65	7.55	68.1	23.65	12.60	46.7	
		Phytopl.	May	12.15	2.8	77.0	12.05	4.90	59.3	
	Loire	June	14.85	2.6	82.5	14.60	5.80	60.2		
		Soil	–	–	–	–	–	–		
		Litter	–	–	–	–	–	–		
		Phytopl.	September	13.15	3.35	74.5	13.70	5.30	61.3	
		Anthrop.	Gironde	March	13.45	9.15	32.0	13.10	10.04	23.4
				March	14.1	10.05	28.7	14.20	10.85	23.6
Marine	Loire	February	31.5	13.55	57	33.80	18.30	45.9		
		September	11.1	8.9	73.9	10.80	4.00	63.0		

ROC: Residual Organic Carbon; HOC: Hydrolysable Organic Carbon (6N HCl digestion); LOC: Labile Organic Carbon (30-day incubations); phyto: phytoplankton; anthrop: anthropogenic; T₀: initial POC value; T₀₊₃₀: POC value after 30 days of incubation. POC, ROC, T₀ and T₀₊₃₀ values are expressed in % of TSS; HOC and LOC values are expressed in % of POC

fraction higher than 10% (13.9% in May). The results of the incubation tests confirmed the low lability of the POC: values were lower than 3.7% except in May with a LOC percentage (LOC/POC) of 6.2%.

In the Loire ETM, HOC percentages remained lower than 9.9%, except in one sample in February (13.3%). Results of incubation tests were again consistent with results of 6N HCl digestion: the LOC percentages were lower than 5.2%, except in the same sample in February (9.9%).

In the Seine ETM, a seasonal trend of HOC and LOC was observed with the lowest values in autumn and winter (HOC < 0.7% and LOC < 2.2% in October and February), intermediate values in early spring (HOC = 12.4% and LOC = 12.6% in March) and highest values in late spring and early summer (HOC = 58.6% and LOC = 47.6% in May) (Table 3).

It is worth noting that any increase in the labile fraction is always associated with an increase in the POC content. The most striking example is the Seine ETM sample in May: the labile fraction

was about half of the POC and the associated POC was 7.2% (Table 3). The last value is more than twice the annual mean POC content in the Seine ETM (3.1%; Fig. 2). This suggests that there are two different POC pools in the ETMs: (i) a “background” POC, mainly refractory that dominates all year round in the Gironde and Loire ETMs where the POC remains constant and in autumn and winter in the Seine ETM, when the POC values are the lowest; the labile organic fraction in all these samples was lower than 5% (Table 3); (ii) an additional POC, highly labile that becomes significant in the Seine ETM in spring and summer; this is also consistent with other measurements in the Seine ETM in spring and summer, when LOC percentages averaged 20% (Servais & Garnier, 2006).

Lability of organic matter in the riverine material entering the estuaries

The acid treatment and the incubation tests also gave consistent results for the different types of tested riverine organic material (Table 3, Fig. 3)

- HOC ~ 30% and LOC ~ 20% in the Garonne river (Station La Réole) during a late summer flood, when soil organic matter dominated;

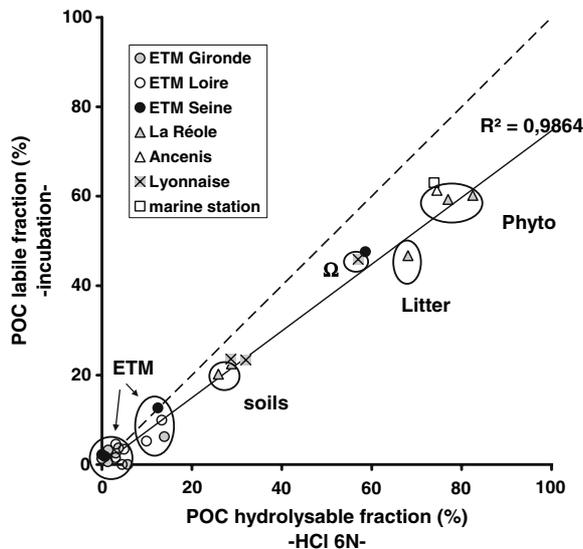


Fig. 3 Linear correlation between percentages of labile organic fraction estimated by incubation tests and of hydrolysable organic fraction estimated by 6N HCl digestion

- HOC ~ 70% and LOC ~ 45% in the Garonne river during the early autumn floods when litter organic matter dominated;
- HOC ~ 85% and LOC ~ 60% in the Garonne (La Réole) and Loire (Ancenis) rivers, when phytoplanktonic organic matter dominated (low TSS, high Chl a);
- HOC ~ 60% and LOC ~ 45% for wastewater organic matter in February, and HOC ~ 30% and LOC ~ 25% in March.

The labile POC content estimates of these domestic wastewater samples are lower than the ones found by Servais et al. (1999a, b) for many more samples (LOC of 70% determined from 56 samples).

Fluvial organic carbon pools entering the Estuaries of the Gironde, Loire and Seine

Mean discharge-TSS-weighted POC contents (POC*) were higher in the Loire and Seine Rivers than in the Garonne: 8.7, 6.5 and 3.2% respectively (Table 4). The numerous data on the Garonne River ($n > 200$) allow a more precise estimate than is possible for the Loire and Seine (40–50 samples for each).

The different pools of POM entering the Gironde Estuary have already been quantified (Veyssy et al., 1999; see Material & Methods section). A major part of the POC originates from allochthonous organic matter (50% and 40% due to soil and litter, respectively); only 10% are autochthonous (phytoplankton). The threshold POC value, corresponding to 100% of soil organic matter, is estimated at 2% (lowest POC contents observed during winter floods).

With the same approach (Veyssy et al., 1999) applied to the Loire Estuary, on the basis of the POC measurements and chlorophyll *a* contents (allowing an estimate of the algal organic carbon), the percentages of the different organic matter pools were: 35% due to soil, 15% due to litter and 50% due to phytoplanktonic blooms (Table 4). The threshold POC value, as defined above, is estimated at 3.1%.

Table 4 Computed contribution by the different organic pools to the riverine POC* (mean discharge-TSS-weighted POC entering the three estuaries)

	POC* (%TSS)	% Soil	% Litter	% Phytoplankton	% Anthropogenic
Gironde	3.2	50	40	10	<0.1
Loire	8.7	35	15	50	<0.1
Seine	6.5	45	25	20	10

In the Seine River, 45% of the riverine organic matter was due to soil, 25% to litter and 20% to phytoplankton. The remaining 10% was of human origin (Table 4). The latter fraction was estimated by the increase of POC content in suspended matter, by comparing the values measured at three stations (Idlafkih, 1998), upstream from Paris, downstream from Paris and at the Poses dam station (the limit between the lower Seine River and upstream estuary, 200 km from Paris). The impact of the wastewater effluents was demonstrated by the increase in POC content observed during high water events when the phytoplanktonic fraction was small. The threshold POC value is 2.9%.

Discussion

Limitations of the labile organic carbon determination

The partitioning of the organic matter into labile (considered as bioavailable or biodegradable organic matter) and refractory fractions has always been a matter of debate: the definition of degradability itself is arbitrary (depending on time scale, environmental conditions, etc.; Lin, 1988). Even if progress has been made (Hopkinson et al., 1997), all technical approaches remain questionable.

Two rapid and practical techniques, taking into account all of the POC, have been used here. But, the limitations of these techniques must be kept in mind: difficulty of determining the environmental significance of the lability characteristics of the hydrolysable organic fraction; dissimilarity between the enclosed environment of the *in vitro* incubation tests and the *in situ* field conditions.

Nevertheless, when comparing the results of the two approaches, a linear relationship is found between the percentages of the hydrolysable organic fraction and those of the POC mineralized during incubation tests ($R^2 = 0.99$, Fig. 3). Note however, that the percentage of LOC in riverine organic matter, is almost always lower than HOC. This may be due to two facts:

On one hand, incubation test may underestimate the LOC due to the effect of enclosed environmental conditions, changing from field conditions for the entire incubation period, and to the duration of the incubation (30 days) which is sometimes not long enough to reach the asymptotic stage of refractory POC contents. In addition, ETMs are known to accelerate the degradation of fluvial organic matter, as has been demonstrated for pigments (Lemaire et al., 2002a). Here, the incubations of fluvial organic matter were done without taking into account this acceleration effect. On the other hand, HCl treatment may hydrolyze some refractory molecules and overestimate the contribution by labile organic matter. Nevertheless, LOC and HOC data give minimum and maximum values that can be used to predict the fate of the different riverine POC pools in the ETMs.

Refractory POC contents in rivers and in the ETMs

On the basis of yearly fluxes of various organic pools entering the three estuaries (Table 4) and their associated degradability, a theoretical refractory POC can be calculated and compared with the observed POC values in the ETMs. Calculations were done using both HOC and LOC data. In the case of LOC, the calculations can be written as follows:

$$\text{ROC}^* = \text{POC}^* - \text{LOC}^{**}$$

$$\text{With } \text{POC}^* = \text{POC}^*_P + \text{POC}^*_L + \text{POC}^*_S$$

$$\text{And } \text{LOC}^* = \text{LOC}_P + \text{LOC}_L + \text{LOC}_S$$

$$\text{ROC}^* = (\text{POC}^*_P - \text{LOC}_P)$$

$$+ (\text{POC}^*_L - \text{LOC}_L) + (\text{POC}^*_S - \text{LOC}_S)$$

In the case of the Gironde estuary:

$$\text{POC}^*_P = 10\% \times \text{POC}^*$$

$$\text{POC}^*_L = 40\% \times \text{POC}^*$$

$$\text{POC}^*_S = 50\% \times \text{POC}^*$$

And

$$\text{LOC}_P = 60\% \times \text{POC}^*_P$$

$$\text{LOC}_L = 45\% \times \text{POC}^*_L$$

$$\text{LOC}_S = 20\% \times \text{POC}^*_S$$

$$\begin{aligned} \text{ROC}^* = & \left[\left(\frac{\text{POC}^*_P}{\text{POC}^*} \times \text{POC}^* \right) \times \left(1 - \frac{\text{LOC}_P}{\text{POC}^*_P} \right) \right] \\ & + \left[\left(\frac{\text{POC}^*_L}{\text{POC}^*} \times \text{POC}^* \right) \times \left(1 - \frac{\text{LOC}_L}{\text{POC}^*_L} \right) \right] + \\ & + \left[\left(\frac{\text{POC}^*_S}{\text{POC}^*} \times \text{POC}^* \right) \times \left(1 - \frac{\text{LOC}_S}{\text{POC}^*_S} \right) \right] \end{aligned}$$

With ROC^* : theoretical refractory POC entering the Gironde estuary (%TSS).

POC^* : mean discharge-TSS-weighted POC contents in the Garonne River (%TSS).

POC^*_P (POC^*_L ; POC^*_S): phytoplanktonic (litter; soil) fraction of the POC^* (%TSS).

Table 5 Comparison between the POC contents in the rivers and in the ETMs of the Gironde, Loire and Seine estuaries. ROC means refractory organic carbon

	River inputs		ETM
	POC* (%TSS)	ROC (%TSS)	POC (%TSS)
Gironde	3.2	1.6–2.1	1.5
Loire	8.7	3.3–4.9	3.3
Seine	6.5	3.2–4.1	3.1

The maximum ROC value results from the LOC data (incubations) and the minimum value from the HOC data (HCl 6N attack)

LOC^* : mean discharge/TSS-weighted LOC (Labile Organic Carbon) content in the Garonne River (%TSS).

LOC_P (LOC_L ; LOC_S): phytoplanktonic (litter; soil) fraction of the LOC^* (%TSS).

Finally,

$$\text{ROC}^* (\% \text{TSS}) =$$

$$[(0.1 \times 3.2) \times (1 - 0.60)]$$

$$+ [(0.4 \times 3.2) \times (1 - 0.45)]$$

$$+ [(0.5 \times 3.2) \times (1 - 0.20)] = 2.1$$

Table 5 summarizes these calculations using both HOC and LOC data in the three estuaries. Like the POC contents in the three ETMs, the theoretical fluvial refractory organic carbon (ROC^*) is much lower in the Gironde estuary than in the Loire and Seine estuaries. In addition, for the three estuaries, the calculated fluvial ROC^* is very similar to the POC content in the ETM, but closer to it when calculated from the HOC data (HCl 6N attack) than from the LOC data (incubations).

For average hydrological conditions, the low lability of POC in the ETMs (Table 3) supports the assumption that the residence times of TSS are long enough for an almost complete mineralization of the labile organic matter supplied in majority by the rivers. If so, the fluvial ROC^* should exactly match the ETM POC content. It appears (Table 5) that the 6N HCl digestion closely reflects the lability of the fluvial organic matter whereas the incubation approach tends to underestimate it. Several environmental factors that favour POC mineralization in ETMs are not reproduced in the incubations of fluvial material. First, the residence times of TSS (Table 1) are generally longer than the incubation period (1 month). Secondly, high TSS concentrations in ETMs lead to a higher biomass of particle-attached bacteria (Crump et al., 1996) than in the incubations and accelerate the organic matter degradation (Lemaire et al., 2002a). Thirdly, TSS are subjected to sedimentation/resuspension cycles in ETMs, which induce oxic/anoxic oscillations and increase degradation rates (Abril et al., 1999).

The POC content in the ETMs of the three studied estuaries can be explained by the contri-

bution of the four different organic pools supplied by the rivers and their respective refractory fraction as obtained by the HCl 6N attack. Moreover, when degradation has been achieved in the ETMs, the yearly average contribution of the different pools is very different from one estuary to another. We calculated that in the Gironde ETM, 75% of the POC originates from soil, 23% from litter and 2% from phytoplankton. In the Loire ETM, 70% originates from soil, 12% from litter and 18% from phytoplankton. In the Seine ETM, 70% originates from soil, 15% from litter, 6% from phytoplankton and 9% from human effluents.

Despite the caution required when residence times of freshwater and particles are used, here regarded as indicators rather than precise estimates (Uncles et al., 2002), a conceptual framework of the functioning of these estuaries can be proposed: in conditions of low summer water discharge, particle residence times are long enough to assume that the major part of labile organic material will be mineralized in the ETM areas of the three estuaries. For the Seine estuary, this is clearly supported by the decrease in POC content (see Fig. 2) but also by a concomitant decrease of its biodegradable fraction from spring to autumn. By contrast, at high water discharge, residence times of new riverine material entering the estuaries can be very short (cf. the Seine and the Loire Estuaries with residence times of a few hours and from one to a couple of days respectively, Migniot, 1991). In such conditions, material containing higher fractions of labile organic matter (mainly allochthonous) can be delivered directly to the coastal zone. However, this should not be the case for the Gironde Estuary where riverine material can be flushed out in 1 or 2 weeks, if the conditions are extreme: series of floods with a very high final water discharge ($>5000 \text{ m}^3 \text{ s}^{-1}$), and spring tides.

Overall, the probability of POC contents in ETMs being greater than the annual mean POC is higher in the Seine and the Loire Estuaries than in the Gironde Estuary. The pronounced trophic state of the rivers (phytoplankton development in the spring), domestic pollution (large effluent discharge), as well as arrival of fresh riverine organic material (undegraded) during high water

discharges, are factors—together with the residence times in the estuaries—that explain the difference between these three macrotidal estuarine systems.

Conclusion

In the ETMs of the three major French estuaries (Gironde, Loire and Seine), the annual average POC contents (expressed in % of dry suspended sediment) are 1.5, 3.3 and 3.1% respectively. Whereas no seasonal variations of these contents are observed in the Gironde Estuary, high values have been found in the Loire and Seine estuaries, highest in the Seine Estuary. In the three ETMs, the particulate organic matter is essentially refractory, but any POC content increase is due mainly to labile organic matter (phytoplankton blooms, effluent discharge, etc). Complementary experiments on the degradability of the diverse organic-matter pools entering these estuaries reveal that, on average, the labile/refractory organic ratios decrease in the order: phytoplankton \sim litter $>$ human-origin organic matter \gg soil. Finally, the residence times of water and TSS are long enough in the three estuaries to allow the assumption the labile fraction of the organic matter can be mineralized at a seasonal scale. By combining the yearly fluxes of various organic pools entering the estuaries and their associated degradability one can explain the organic contents of the three ETMs, as well as their dominant refractory characteristics. This study shows, according to the river continuum concept (Vannote et al., 1980), the need to analyze the behaviour of the upstream drainage basin to better understand the specificities of these three estuaries. As primary production is strongly light-limited in estuaries, the organic matter content of ETMs together with its biodegradability, are major ecological factors controlling the oxygenation level and estuarine aquatic life.

Acknowledgements This work received financial support from the Seine Aval II programme. The authors are grateful to several colleagues who provided basic data used in the paper: Mr. Nicolas Ampen (DIREN, Nantes) and Ms. Cécile Gauffeny (DDASS, Nantes), who supplied the daily average discharge data of the Loire River for the last

10 years, and some TSS values at the entrance of the Loire Estuary; Mr. André Ficht and Ms. Marielle Olivier (SNS, Rouen), who both provided similar data and water samples from the Seine River and Estuary. Many thanks are due to Axel Romana, director of the programme Seine-Aval, for fruitful discussions and encouragement.

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