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Does sediment resuspension by storms affect the fate of polychlorobiphenyls (PCBs) in the benthic food chain? Interactions between changes in POM characteristics, adsorption and absorption by the mussel *Mytilus galloprovincialis*

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Abstract

Environmental parameters and gross sedimentation rates (GSR) were monitored at a fixed site located in the Bay of Banyuls-sur-Mer (NW Mediterranean), between March 1997 and April 1998, together with the main biochemical characteristics of both sedimenting and sedimented particulate organic matter (POM). Three storms which occurred during this time period resulted in natural sediment resuspension. This is indicated by the corresponding increase in GSR and a decrease in the enzymatically hydrolysable amino acids/totally hydrolysable amino acids ratio (EHAA/THAA), within the sedimenting POM. Only the strongest storm resulted in (1) a transitory increase in fine-grained particles, (2) concomitant increases in organic carbon, carbohydrates, lipids and THAA, and (3) a decrease in the EHAA/THAA ratio in surficial sediments. For most of the assayed parameters, the values recorded after the December 1997 storm corresponded to extremes for the whole period under study. This emphasises the role of storms in controlling the characteristics of sedimented and sedimenting POM.

Ten sediment types, with contrasting biochemical characteristics, were selected for experiments; these were based on the results of the monitoring survey and were used during adsorption and absorption experiments involving ^{14}C tetrachlorobiphenyl (TCB). Adsorption rates differed significantly between the sediment types, but did not correlate with any of the assayed biochemical parameters. Absorption efficiency by the mussel *Mytilus galloprovincialis* also differed between the sediment types; it correlated positively with all the assayed biochemical parameters, except lipids. Comparison between the magnitudes of the increase in GSR, together with the decrease in absorption efficiency during

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resuspension events, suggests that resuspension tends to enhance the transfer of organic pollutants in the benthic food chain.

Keywords: Sediment resuspension; POM; PCB; Food chain; Adsorption; Absorption; *Mytilus galloprovincialis*

1. Introduction

The frequent occurrence of disturbances is one of the main factors structuring coastal ecosystems (McCall, 1977). Depending upon their nature, perturbations usually result in either a direct (organic pollution) or an indirect (i.e., due to defaunation) increase in organic matter availability (Pearson and Rosenberg, 1978). Storms are clearly one of the major causes of natural disturbance in coastal ecosystems. Such events often result in important resuspension processes, which may directly affect benthic fauna (Thistle et al., 1995) and the functioning of coastal ecosystems, through changes in nutrient, light and organic matter availability. Field surveys assessing the effects of storm-induced resuspension on the functioning of coastal ecosystems have mostly been carried out in estuaries (Shideler, 1984; Gabrielson and Luketelich, 1985; Demers et al., 1987; Weir and MacManus, 1987; Simon, 1989) and in coastal lagoons (Arfi et al., 1993, 1994; Bouvy et al., 1994; Arfi and Bouvy, 1995). Such studies have shown (1) the direct effect of resuspension, on both the quantity and the size spectra of suspended particles, and (2) the indirect effect (through nutrient regeneration and light conditions) on both phytoplanktonic (Arfi et al., 1993) and bacterial communities (Wainright, 1987; Ritzrau and Graf, 1992).

A recent survey carried out in the NW Mediterranean (Grémare et al., 2003a) has confirmed the transfer of important amounts of nutrients, from the sediment to the water column, and significant increases in bacterial biomass and production after a major winter storm. This study also highlighted the effect of this storm on the quantity and quality of the POM associated with settling and sedimented particles. These results confirmed those obtained previously by Grémare et al. (1997, 1998) and Medernach et al. (2001) using another sampling strategy. More specifically, it was shown that resuspension resulted in (1) an increase of refractory organic matter associated with particles collected in sediment traps and (2) the transitory deposition of fine-grained particles at the surface of the sediment, resulting in an increase of refractory organic matter. The time duration needed for the return to pre-storm conditions tended to increase with depth in the water column; it was maximal (i.e., about 2 weeks) in surficial sediment. As with most of the field studies dealing with the effects of resuspension, this survey was a short-term one (i.e., of about 1 month) and based on high-frequency sampling. Information regarding the importance of resuspension events, relative to those of other environmental fluctuations, is thus still largely lacking .

Beside particles and refractory POM, the sediment also constitutes a repository for many anthropogenic chemicals, including organic contaminants such as polychlorinated biphenyls (Gunnarsson et al., 1999). Non-polar organic pollutants have a strong sorption affinity for organic matter (Karickhoff et al., 1979). Sediment resuspension may then both increase the quantity of contaminants in the water column and affect the availability of suspended POM, to primary consumers (Grémare et al., 1997). In so far as hydrophobic contaminants are bioavailable, feeding becomes an important route of contamination for benthic invertebrates (Landrum and Faust, 1994; Gilek et al., 1997). However the bioavailability of contaminants depends upon the nutritional quality of the sedimentary organic matter (Landrum and Faust, 1994; Gunnarsson et al., 1999). Thus, the effects of sediment resuspension can interact in controlling the fate of contaminants in the benthic food chain, as suggested already for the interaction between long-term eutrophication and an increase of contaminants (Gunnarsson et al., 2000).

Within this context, our first aim was to compare the magnitude of the effect of winter storms on the biochemical characteristics of both sedimenting and sedimented POM with those of other fluctuations occurring during a year cycle. This objective was achieved through a 1-year-long monitoring carried out at a fixed site located within the Bay of Banyuls-sur-Mer (NW Mediterranean). This sampling strategy also allowed for the comparisons of the effects of three storms, differing in their intensity. Our second aim was to assess potential interactions between changes in POM biochemical characteristics associated with storms with the fate of contaminants. This objective was achieved through two sets (i.e., adsorption and absorption) of experiments, involving POM collected at different periods of the year and corresponding to different resuspension regimes. These two sets of experiments were carried out using ^{14}C tetrachloro biphenyl. Absorption experiments were carried out with the suspension-feeding bivalve *Mytilus galloprovincialis*.

2. Materials and methods

2.1. Environmental parameters

Maximum wind speed and direction were registered daily, between March 1, 1997 and April 20, 1998 together with rainfall at the Cape Béar METEO-FRANCE station (Fig. 1). Wave height and direction were derived from the VAGMED model (METEO-FRANCE). Water samples were collected weekly (42° 29′ 302N and 03° 08′ 700E) in the Bay of Banyuls-sur-Mer (NW Mediterranean) to assess chlorophyll *a* (chl *a*) concentrations. Samples (0.2 L) were collected at 3 and 24 m depth; they were filtered immediately on Whatman GF/F filters and the chl *a* was assessed on triplicates, after Lorenzen (1966).

2.2. Monitoring of the characteristics of sedimenting and sedimented POM

Two sediment traps were deployed, between March 1997 and April 1998, at the same site. The traps consisted of polyethylene pipes, prolonged by a cone and a collector. The inner diameter of the pipe was 40 cm, with a total height of 190 cm; this provided an aspect ratio of 4.75. The two traps were mounted on the same frame, with the centres of their mouths being less than 1 m apart (Charles et al., 1995). The mouths of the traps were located 3 m above the seabed located at 26 m. The contents of the traps were collected weekly by SCUBA diving. They were taken to the laboratory where they were pooled, centrifuged (2952*g*, 15 min), rinsed briefly with distilled water, frozen, freeze-dried and weighed. Gross sedimentation rates (GSR) were defined as the total amount of material sampled in a sediment trap, with a known cross-sectional area and over a known length of time. Biochemical assays were then run on the material which passed through a 200 μm mesh. This fraction was stored at −20 °C, prior to analysis. Each week, 3 cores of sediment were collected by SCUBA divers. Back at the laboratory, these cores were sliced and biochemical assays were run on the first centimetre of sediment, which was processed as described for the sediment trap material.

Sediment trap and surface sediment granulometry were assessed using a Malvern® Mastersizer 2000 laser microgranulometer. The *d*(0.5) (i.e., the diameter corresponding to the median of particle volumes, assuming that all particles are spherical) was used as an index of the particle size. All the biochemical assays were run on triplicates. Organic carbon and nitrogen contents were measured, after acidification (1 N HCl), using a CHN Perkin Elmer® 2400 analyser. Carbohydrates and lipids were assayed after the methods of Dubois et al. (1956) and Barnes and Blackstock (1973), respectively.

In order to assess total hydrolysable amino acids (THAA), 15 mg dry weight (DW) of sediment was submitted to a strong acid hydrolysis (500 μL of 6 N HCl, 100 °C, 24 h, under vacuum). 0.4 mL subsamples of the hydrolysates were neutralised with 0.4 mL of 6 N NaOH and buffered with 0.8 mL of H_3BO_3 (0.4 M, pH 8). Fluorescent derivatives were obtained by adding 6 μL of an orthophthaldialdehyde solution (125 mg in 2.5 mL of methanol and 0.125 mL of mercaptoethanol) and 400 μL of H_3BO_3 to 100 μL of those samples. THAA quantification was based directly on

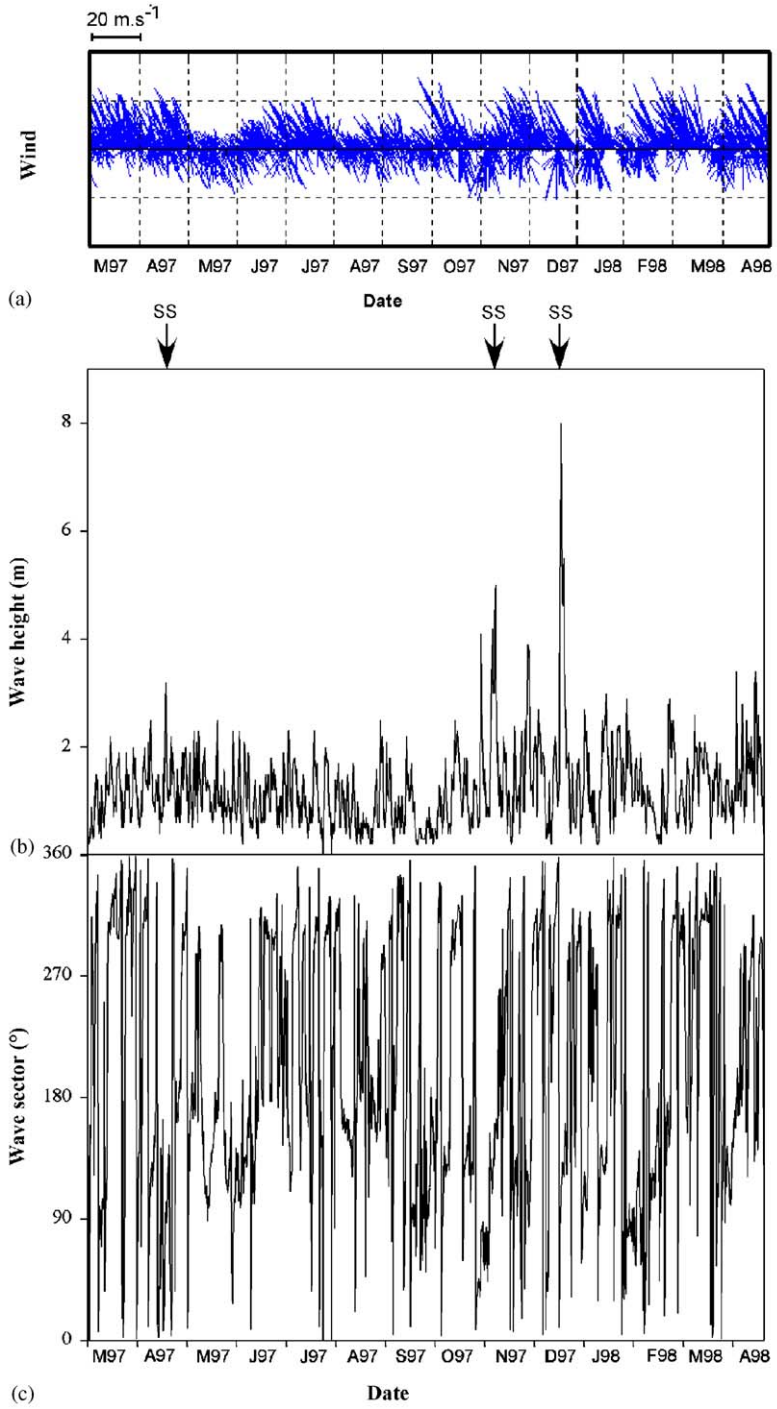

Fig. 1. Temporal changes in the wind regime (a), wave height (b) and wave direction (c) during the period under study. The main southerly storms (SS) are indicated by vertical arrows.

fluorescence measurements and was achieved through comparison with a standard containing 19 amino acids. The excitation wavelength was 335 nm and the emission wavelength was 450 nm.

Enzymatically hydrolysable amino acids (EHAA) were extracted, following the biomimetic approach proposed by Mayer et al. (1995). 100 mg DW of sediment was poisoned with 1 mL of a solution containing 2 inhibitors of bacterial active transport systems (0.1 M sodium arsenate and 0.1 mM pentachlorophenol, within a pH 8 sodium phosphate buffer). This mixture was left to incubate for 1 h at room temperature. 100 μL of proteinase K solution (1 mg mL^{-1}) was then added and the samples were incubated for 6 h at 37 °C; they were centrifuged then to discard remaining particulate material. 75 μL of pure trichloracetic acid (TCA) was added to 750 μL of supernatant, to precipitate macromolecules; these are considered to be non-suitable for absorption. 750 μL of the supernatant was then hydrolysed and processed as described for THAA. In addition, a blank (accounting for possible degradation of the enzyme) was carried out. EHAA were quantified using the same procedure as for THAA. EHAA/THAA ratios were expressed in percent.

The significance of temporal changes in the characteristics of the surficial sediments was assessed using Kruskal–Wallis non-parametric ANOVAS (using the 3 cores as replicates). Since the contents of the 2 traps were pooled, we did not test the significance of temporal changes in the characteristics of sediment trap collected material (see Grémare et al. (1997) and Medernach et al. (2001) for the discussion of a similar problem).

2.3. [U-^{14}C]TCB adsorption experiment

Since the amount of material collected changed with the collection dates, we had to combine the samples from several dates in order to provide enough material to run the adsorption and absorption experiments. This approach has led us to define 9 sediment trap samples, corresponding either to strong resuspension events or to periods preceding or following such events (Fig. 2). Samples I, III, IV, IX and X corresponded to sediment trap materials collected during periods with low GSR (Figs. 3–6). Samples II, V and VII corresponded to sediment trap materials collected during the resuspension events associated with April 1997, November 1997 and December 1997 storms, respectively. Sample VI corresponded to sediment trap material collected during a resuspension event of lower intensity, which occurred during early December 1997. Sample VIII corresponded to surficial sediment collected below the sediment traps on January 6 1998. This overall approach resulted in the selection of 10 sediment types.

Adsorption experiments consisted of the mixing of wet sediment, with [U-^{14}C]-3,3′,4,4′-tetrachlorobiphenyl (TCB, IUPAC 77) (Amersham Pharmacia Biotech®, specific activity: 3.85 GBq $mmol^{-1}$) coated on the inside wall of glass test tubes (Ditsworth et al., 1990). The inside walls of the test tubes were first coated with 0.97 μg of [U-^{14}C]TCB, dissolved in acetone which was evaporated in a fumehood. 500 mg dry weight of moistened sediment was added to each tube. Test tubes were then capped and rolled for 24 h, before being kept for 6 weeks at 4 °C to reach equilibration between solid and liquid phases (Murdoch et al., 1997). Sediment slurries were then centrifuged (1700*g*, 10 min, 15 °C), rinsed twice with filtered (GF/F Whatman) seawater and freeze-dried. [U-^{14}C]TCB was then extracted in 5 mL of acetone:hexane (v:v) (ambient temperature, 12 h). This step was repeated 4 times and the radioactivity of the corresponding acetone/hexane fractions was measured using liquid scintillation counting (see below). Five replicates were carried out for each sediment type.

2.4. [U-^{14}C]TCB absorption experiment

The 9 sediment trap samples were sieved and absorption experiments were run on the size fraction below 40 μm. In addition to the assessment of the biochemical characteristics listed above, the organic contents of the 10 sediment types were assessed, by measuring their weight loss after combustion (450 °C, 5 h) on triplicates. For each sediment type, 500 mg of wet sediment was labelled with 18.4 μg of [U-^{14}C]TCB, as described above. Absorption efficiencies of [U-^{14}C]TCB, by *M. galloprovincialis*, were assessed using 'pulse–chase' feeding design experiments carried out on

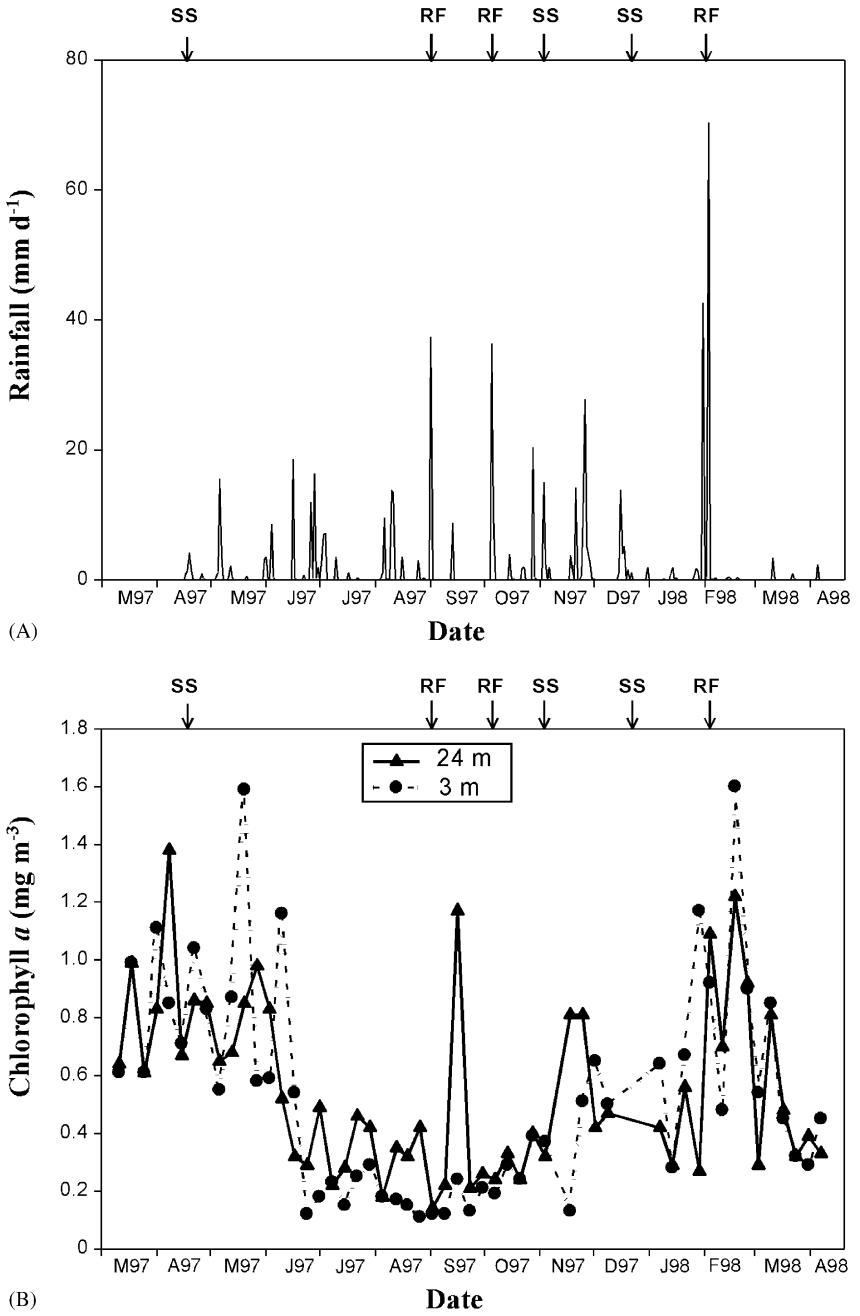

Fig. 2. Temporal changes in the daily rainfall (a) and chlorophyll *a* concentrations at two depths in the water column (b) during the period under study. The main meteorological events (SS: southerly storm; RF: heavy rainfall) are indicated by vertical arrows.

mussels (30.8 ± 1.6 mm in shell length) collected during spring 2003, in the immediate vicinity of the laboratory. Three mussels were introduced in an experimental 'pulse' chamber containing 1.5 L (GF/F) of filtered sea water, where they were provided with three 7.5 mg DW pulses of labelled

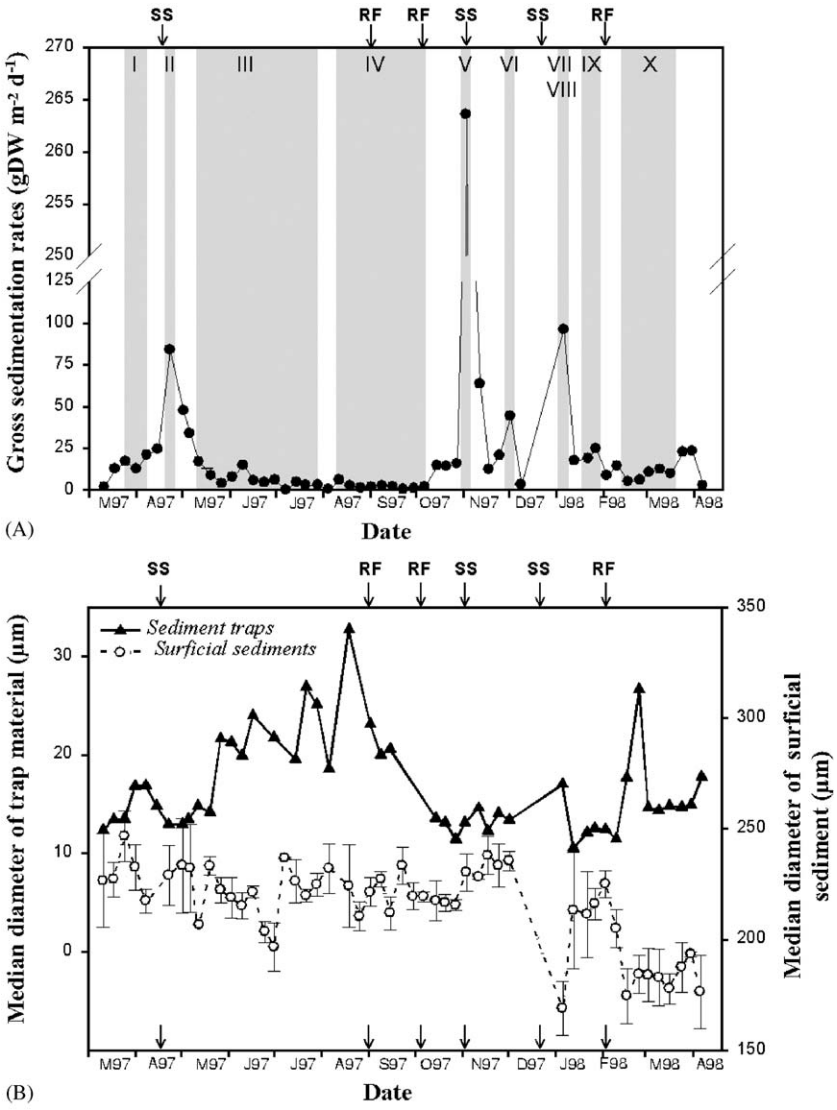

Fig. 3. Temporal changes in the gross sedimentation rates (a) and in the median diameters of sediment trap material and surficial sediment (b) during the period under study. The main meteorological events (SS: southerly storm; RF: heavy rainfall) are indicated by vertical arrows. Vertical bars are standard deviations. Shaded areas and roman figures refer to the 10 sediment types, as used during the adsorption and absorption experiments (see text).

sediments over 1 h. Mussels were moved then to clean 'chase' chambers, containing 1.5 L of filtered (GF/F) seawater, and then were fed regularly (7.5 mg DW every 30 or 40 min) with the same but non-labelled sediment type for the next 24 h. The partitioning of the radioactivity, within the 'chase' chambers, was assessed 1, 3, 6, 12, 18 and 24 h after the initial transfer of the mussels. The entire volume of seawater was filtered on a GF/F filter and the mussels were moved to another 'chase' chamber. Radioactivity in the filtrate was assessed from two subsamples of 10 mL each. Radioactivity

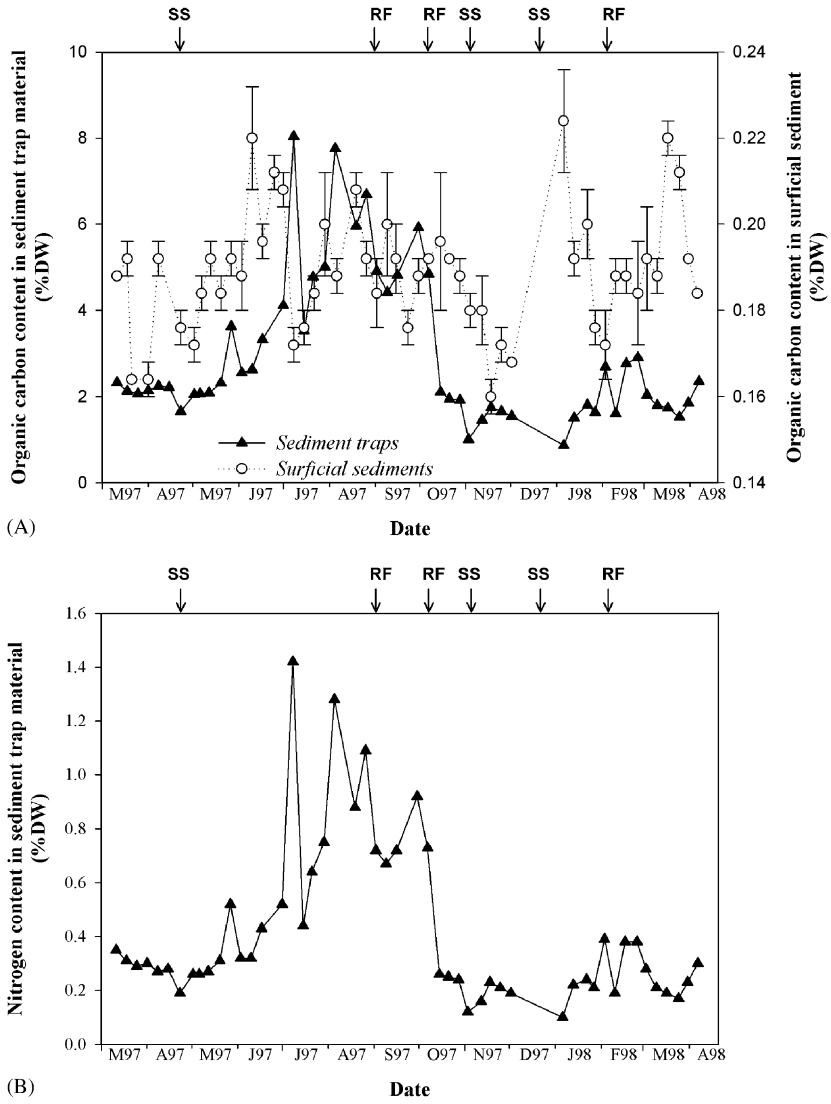

Fig. 4. Temporal changes in the organic carbon of sediment trap material and of surficial sediment (a), and in the nitrogen content of sediment trap material (b). The main meteorological events (SS: southerly storm; RF: heavy rainfall) are indicated by vertical arrows. Vertical bars are standard deviations.

in the biodeposits was determined from the GF/F filter. At the end of the 24 h depuration period, the mussels were dissected. Soft tissues were hydrolysed at 60 °C, in a known volume of NaOH, 1 N, and the amount of radioactivity in the mussel flesh at the end of the 'chase' period was measured from two 1 mL subsamples of the hydrolysates. Absorption efficiency (AE) was then computed according to the following formula:

$$AE\ (\%) = ([ING] - [BIODEP]) * 100/[ING],$$

where [ING] is the radioactivity ingested during the 'pulse' period and [BIODEP] is the cumulated amount of radioactive biodeposits produced by the end of the 'chase' period.

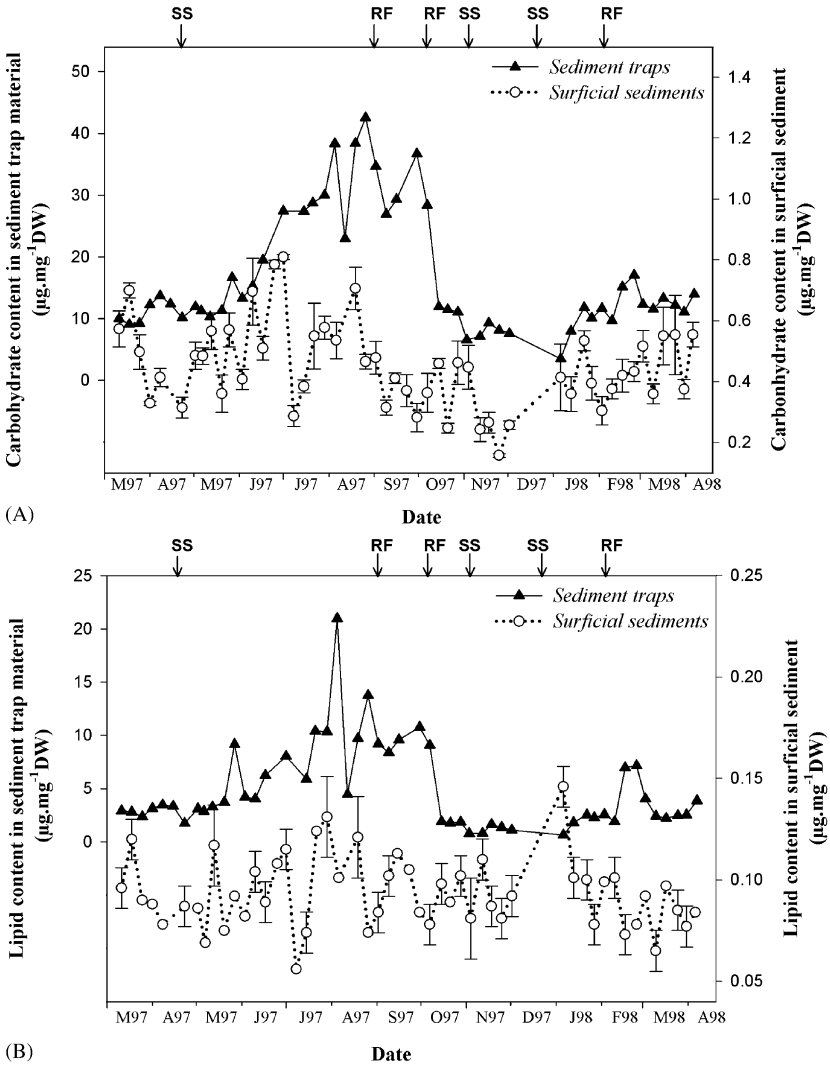

Fig. 5. Temporal changes in the carbohydrate (a) and lipid (b) contents of sediment trap material and the surficial sediment. The main meteorological events (SS: southerly storm; RF: heavy rainfall) are indicated by vertical arrows. Vertical bars are standard deviations.

Experiments were run at $15 \pm 0.5\,^{\circ}C$. A strong air bubbling system aerated both the 'pulse' and the 'chase' experimental chambers, maintaining the sediment particles in suspension. There were five replicates undertaken for each sediment type.

All the radioactivity measurements were carried out using a Beckman® LS 5000 CE scintillation counter. All the samples were corrected for quenching, chemiluminescence and background counting.

3. Results

3.1. Environmental parameters

Northerly winds were dominant during the year under study (Fig. 1a). Throughout the year, northerly winds were alternating with shorter periods of southerly winds. Likewise, analysis of the wave data indicate that southerly winds were associated with storms during wintertime

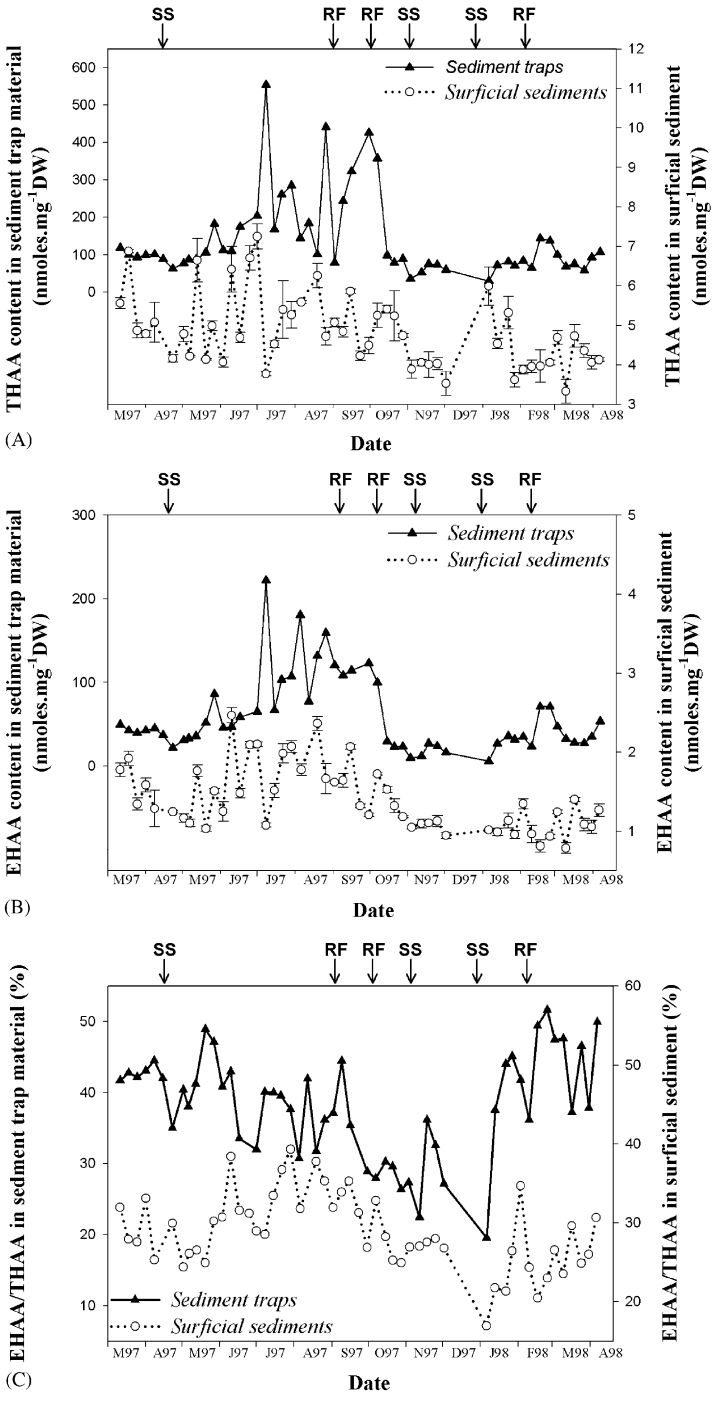

Fig. 6. Temporal changes in the THAA (a), EHAA (b) and EHAA/THAA ratios (c) of the sediment trap material and the surficial sediment. The main meteorological events (SS: southerly storm; RF: heavy rainfall) are indicated by vertical arrows. Vertical bars are standard deviations.

(Figs. 1b and c). Three storms of this kind took place in 1997. The first one occurred between April 16 and 19, the second one between November 2 and 6, and the third one between December 16 and 19. The last storm was by far the strongest, with wave heights of up to 8 m.

The heaviest rainfall (Fig. 2a) occurred on September 1, 1997 ($37.4\,mm\,d^{-1}$), October 5, 1997 ($36.4\,mm\,d^{-1}$) and February 3, 1998 ($70.2\,mm\,d^{-1}$). The December 1997 storm was associated also with heavy rains (23 mm during a 3-day period).

Temporal changes in chl *a* concentrations in the water column (Fig. 2b) show clearly a seasonal cycle, with higher concentrations during winter and spring and lower concentrations during summer and autumn. Some of the highest concentrations, at both depths, were recorded on February 22, 1998, this corresponding, probably, to the late-winter diatom bloom described by Jacques (1970).

3.2. Monitoring of the characteristics of sedimenting and sedimented POM

The GSR ranged between 0.3 (July 8, 1997) and $263.5\,gDW\,m^{-2}\,d^{-1}$ (November 3, 1997, Fig. 3a). GSR were extremely low between mid-March and mid-October 1997. During the remainder of the period under study, the GSR were somewhat variable and sometimes very high. There were 3 marked peaks in the GSR (i.e., on April 23, 1997, November 3, 1997 and January 6, 1998), which were associated closely with storms. Heavy rainfall had almost no effect on the GSR.

Particles collected in the sediment traps were always fine-grained (the median diameter ranged between 11 μm on January 13, 1998, and 33 μm on August 19, 1997, Fig. 3b). The median diameter tended to be higher during summer than during the rest of the year, except during the end of February 1998 (median diameter of 27 μm on February 25); this is a period characterised by high chl *a* concentrations in the water column (see above). There appears to be no clear effect of storms and heavy rainfalls on the size of the particles collected in the sediment traps. Surficial sediment grain size was around 200 μm and did not show any seasonal trend. However, temporal changes in the median diameter of the surficial sediment were significant (Kruskal–Wallis ANOVA, $p<0.0001$). The most important change was an abrupt reduction, from 235 to 169 μm, between December 2, 1997 and January 6, 1998; this was linked probably to the December 1997 storm. This change was transitory and the $d(0.5)$ returned to 213 μm on January 13, 1998.

The organic carbon and nitrogen contents of the sediment trap materials were between 0.9 and 8% DW (on January 6, 1998 and July 7, 1997), and 0.1 and 1.4% DW (on January 6, 1998 and July 7, 1997), respectively (Figs. 4a and b). These two parameters showed the same seasonal pattern, with higher values during spring and summer and lower values during autumn and winter. There was no evident effect of storms on the organic carbon and nitrogen concentrations of the sediment trap material. In both cases, temporal changes were dominated clearly by the abrupt reduction in concentration associated with the transition between summer and autumn. This reduction followed immediately the October rainfalls. Temporal changes in the organic carbon content of the surficial sediments were significant (Kruskal–Wallis ANOVA, $p<0.001$); this parameter was between 0.15 and 0.31% DW (on November 18, 1997 and January 6, 1998, respectively) (Fig. 4a). The value tended also to be higher during spring and summer than during autumn and winter, although the trend is less clear than for the sediment trap material. Both the April and the November storms did not affect significantly the organic carbon content of the surficial sediment. Conversely, the December storm resulted in an increase in organic carbon concentration, from 0.17 (December 2, 1997) to 0.31% DW (January 6, 1998). The latter value was the highest of the whole of the period under study. In relation to the surficial sediment granulometry, this increase was transitory and the organic carbon concentrations dropped to 0.18 on February 10, 1998.

Carbohydrate concentrations of the sediment trap materials were between 3.5 (January 6, 1998) and $42.6\,\mu g\,mgDW^{-1}$ (August 26, 1997) (Fig. 5a). Concentrations tended to be higher during spring and summer and lower during autumn and winter.

The most important change in the carbohydrate concentrations occurred in October 1997, corresponding to the transition between these two periods; as observed for organic carbon and nitrogen, it followed immediately the October 1997 rainfalls. All three storms resulted in a small decrease in the carbohydrate concentrations in the sediment trap materials. At the end of February 1998, there was a secondary peak in the carbohydrate concentrations. Temporal changes in the carbohydrate concentrations of the surficial sediments were significant (Kruskal–Wallis ANOVA, $p<0.0001$). The concentrations were between 0.16 (November 25, 1997) and 0.81 μg mgDW^{-1} (July 1, 1997). There was no clear effect of storms or rainfalls on the carbohydrate concentrations in surficial sediments.

Lipid concentrations in the sediment trap materials were between 0.7 (January 6, 1998) and 21.0 μg mgDW^{-1} (August 5, 1997) (Fig. 5b). This parameter showed the same seasonal pattern as carbohydrates, with higher values during spring and summer and lower values during autumn and winter, together with a sharp transition between these two periods. There was also a secondary peak in the lipid concentrations, during late February 1998. There was no clear effect of storms on the lipid concentrations of the sediment trap materials. Temporal changes in the lipid concentrations of the surficial sediments were significant (Kruskal–Wallis ANOVA, $p<0.0001$). Concentrations were between 0.05 (July 8, 1997) and 0.15 μg mgDW^{-1} (January 6, 1998). The April and November 1997 storms had no clear effect on the lipid concentrations. Conversely, the December 1997 storm resulted in an increase in the lipid concentration, from 0.09 (December 2, 1997) to 0.15 μg mgDW^{-1} (January 6, 1998). As for the surficial sediment granulometry and carbohydrate concentrations, this increase was transitory and the lipid concentrations fell to 0.10 μg mgDW^{-1}, on January 13, 1998.

THAA concentrations in the sediment trap materials were between 29.7 (January 6, 1998) and 587.9 nmol mgDW^{-1} (August 5, 1997) (Fig. 6a). Seasonal changes were characterised by (1) higher values during spring and summer and lower values during autumn and winter, (2) a sharp transition between these two periods, and (3) the occurrence of a secondary peak in February 1998. The April and November storms did not have any clear effect on the THAA concentrations. The December 1997 storm resulted only in a slight reduction in the THAA concentrations. However, it should be pointed out that the concentration measured just after this storm was the lowest of the whole period under study. Temporal changes in the THAA concentrations, within the surficial sediments, were significant (Kruskal–Wallis ANOVA, $p<0.0001$); these were between 3.3 (March 10, 1998) and 7.3 nmol mgDW^{-1} (July 1, 1997). The most important temporal change was associated with the December 1997 storm; it consisted of an increase from 3.5 (December 2, 1997) to 6.0 nmol mgDW^{-1} (January 6, 1998). The concentrations returned to 3.6 nmol mgDW^{-1} on January 27, 1998.

The EHAA concentrations in the sediment trap materials were between 5.8 (January 6, 1998) and 221.9 nmol mgDW^{-1} (July 8, 1997) (Fig. 6b). Seasonal changes were very similar to those described for THAA. The three storms did not have any clear effect on the EHAA concentrations of sediment trap materials. However, once again, the lowest concentrations of the whole period under study were recorded immediately following the December 1997 storm. Temporal changes in the EHAA concentrations of the surficial sediments were significant (Kruskal–Wallis ANOVA, $p<0.0001$); these were between 0.8 (March 10, 1998) and 2.5 nmol mgDW^{-1} (June 10, 1997). As opposed to what was observed for THAA, the December 1997 storm did not result in any significant increase of EHAA concentrations in surficial sediments.

EHAA/THAA ratios in the sediment trap materials were between 19.5 (January 6, 1998) and 51.6% (February 25, 1998) (Fig. 6c). All three storms resulted in a decrease of EHAA/THAA ratios; the most important one was associated with December 1997, which resulted in the lowest values of the whole period under study. The highest EHAA/THAA ratios were recorded during the end of February 1998, concomitant with the secondary peaks of carbohydrates, lipids, THAA and EHAA in the sediment trap materials (see

above). Temporal changes in the EHAA/THAA ratios of the surficial sediments were significant (Kruskal–Wallis ANOVA, $p<0.0001$). EHAA/THAA ratios were between 16.9 (January 6, 1998) and 39.3% (July 29, 1997). The most important temporal change was associated with the December 1997 storm, resulting in a decrease from 26.8 (December 2, 1997) to 16.9% (January 6, 1998). Once again, this reduction was only transitory and the EHAA/THAA ratios were back to 26.4% on January 27, 1998. Both the April and November storms did not have any evident effect on the EHAA/THAA ratios of the surficial sediments.

3.3. [U-^{14}C]TCB adsorption experiment

The main characteristics of the tested sediment types are presented in Table 1. The sediments covered a broad range of nutritive value, with organic carbon contents and EHAA concentrations differing by a factor 14 and 98, respectively. The results of the adsorption experiments showed that the labelling rates were between 1284 and 1406 dpm mgDW^{-1}. Labelling rates differed significantly between sediment types (Kruskal–Wallis one-way ANOVA, $p=0.001$), but did not correlate significantly with any of the biochemical characteristics of these sediment types (Table 2).

3.4. [U-^{14}C]TCB absorption experiment

Control experiments, run in the absence of mussels, showed that [U-^{14}C]TCB remained bound to particles large enough to be trapped on a GF/F filter; this means that the uptake of label by *M. galloprovincialis* resulted only from the ingestion of sediment. When pooling the results of all the sediment types, mussels removed 43.9±4.8% (mean±SD) of the radioactivity provided in the 'pulse' chamber. During the depuration period, radioactivity transfers corresponded exclusively to the production of biodeposits by the mussels. The monitoring of the production of biodeposits showed that about 95% of the undigested

Table 2
^{14}C-TCB adsorption experiment: correlation coefficients and probability levels of simple linear regression models, linking the main biochemical characteristics of the tested sediment types and the corresponding labelling rates of ^{14}C-TCB

	Correlation coefficient (r)	Probability level (p)
Organic matter	0.408	0.241
Organic carbon	0.384	0.272
Nitrogen	0.404	0.246
Lipids	0.282	0.429
Carbohydrates	0.353	0.316
THAA	0.379	0.278
EHAA	0.408	0.241

Table 1
Main biochemical characteristics, particle median diameter and absorption efficiency of ^{14}C-TCB by *Mytilus galloprovincialis* of the 10 sediment types used during the adsorption and absorption experiments

	OM (%DW)	OC (%DW)	N (%DW)	Lipids (μg mgDW^{-1})	Carbohydrates (μg mgDW^{-1})	THAA (nmol mgDW^{-1})	EHAA (nmol mgDW^{-1})	d(0.5) (μm)	Abs (%)
I	8.3	2.2	0.3	3.0	11.8	98.3	42.7	16	3.6
II	7.6	1.7	0.2	1.8	10.1	62.9	22.0	13	2.8
III	10.1	2.9	0.4	5.1	16.5	135.5	55.5	21	2.4
IV	13.6	4.3	0.6	4.3	28.2	266.8	98.1	24	7.9
V	5.6	1.0	0.1	0.8	6.6	35.1	9.8	13	4.6
VI	6.1	1.5	0.2	1.1	7.6	59.9	16.2	14	5.7
VII	3.8	0.9	0.1	0.6	3.5	29.7	5.8	18	1.6
VIII	1.8	0.3	0.1	0.1	0.4	4.5	1.0	188	0.6
IX	7.0	1.7	0.2	2.2	10.3	77.9	32.5	11	4.4
X	7.5	2.1	0.3	3.7	13.1	92.8	43.2	18	6.8

OM: organic matter; OC: organic carbon; N: nitrogen; THAA: totally hydrolysable amino acids; EHAA: enzymatically hydrolysable amino acids; d(0.5): median diameter; Abs: absorption efficiency by *Mytilus galloprovincialis*.

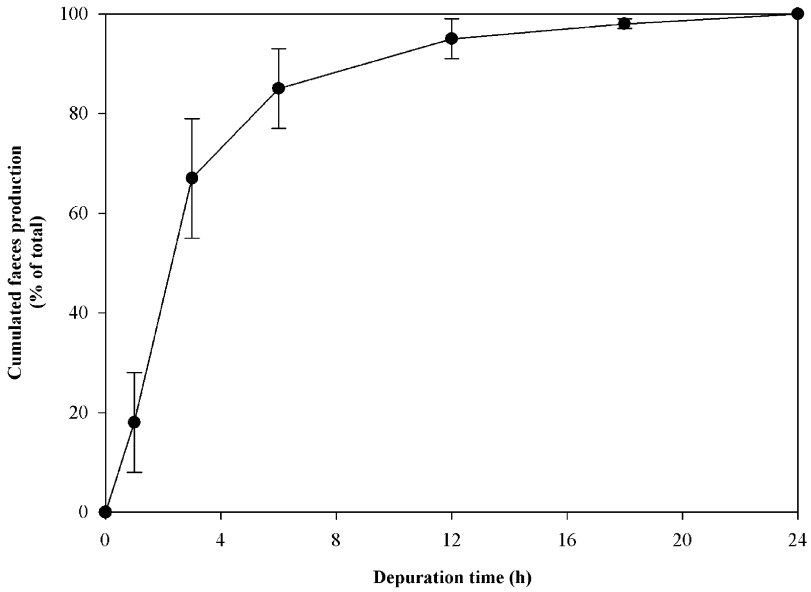

Fig. 7. ^{14}C-TCB absorption experiment: cumulated production of radioactive faeces by *Mytilus galloprovincialis*. Values correspond to means ($N = 47$). Vertical bars are standard deviations.

radioactivity was expelled within the first 12 h of the depuration period (Fig. 7). The absorption efficiency of the [U-^{14}C]TCB, by *M. galloprovincialis*, was affected significantly by sediment type (Kruskal–Wallis one-way ANOVA, $p < 0.001$) (Table 1). The lowest absorption efficiency (0.6%) was recorded for the surficial sediment collected on January 6, 1998 (Sediment Type VIII). The highest absorption efficiency (7.9%) was recorded for the sediment trap material collected during the end of the summer 1997 (Sediment Type IV). The absorption efficiency of [U-^{14}C]TCB correlated positively with the organic content of the tested sediment types (Fig. 8). Absorption efficiency also correlated positively with all the other biochemical parameters studied, except lipids (Table 3).

4. Discussion

4.1. Impact of storms on the characteristics of the sedimenting POM

Field studies assessing the effects of storm on the functioning of coastal ecosystems have been mostly restricted to estuaries and coastal lagoons. Besides the increase in suspended POM due to resuspension, these studies have been dealing with specific processes, such as primary and bacterial productivity (Wainright, 1987; Ritzrau and Graf, 1992; Arfi et al., 1993). Grémare et al. (2003a) first assessed the effects of storms on the biochemical characteristics of sedimenting and sedimented POM, based on high-frequency sampling, before and after a storm. Our own sampling strategy is complementary, since it is involved in a much longer observation period and a lower sampling frequency.

There were three storms of different intensity during the period under study. All three storms resulted in an increase in GSR, which is consistent with the increase in suspended POM induced classically by storms (Bock and Miller, 1995); this is in response either to direct wave action (Puig et al., 2003) or changes in the bottom currents (Tengberg et al., 2003). The analysis of our wave data reveals the occurrence of lower values during April 1997 and the November 1997 storm, together with higher ones during the December 1997 storm. Differences between the intensity of

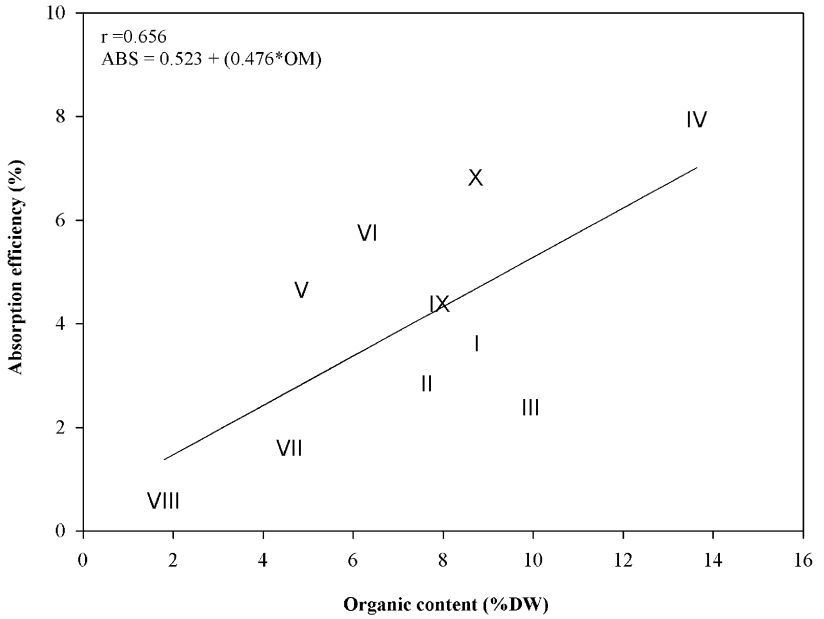

Fig. 8. ^{14}C-TCB absorption experiment: relationship linking the organic content of the 10 tested sediment types and the corresponding mean absorption efficiency of ^{14}C-TCB by *Mytilus galloprovincialis*. Roman figures refer to the tested sediment types.

Table 3
^{14}C-TCB absorption experiment: correlation coefficients and probability levels of the simple linear regression models linking the main biochemical characteristics of the tested sediment types and the corresponding absorption efficiencies of ^{14}C-TCB by *Mytilus galloprovincialis*

	Correlation coefficient (r)	Probability level (p)
Organic matter	0.656	0.043*
Organic carbon	0.639	0.047*
Nitrogen	0.642	0.046*
Lipids	0.440	0.202
Carbohydrates	0.685	0.028*
THAA	0.647	0.043*
EHAA	0.633	0.049*

*$p<0.05$.

these three storms are represented by the integrated area delimited by the GSR curve (Fig. 3). The inversion in the GSR peak height, between the November and the December 1997 storms, results from a sampling artefact. Due to technical problems, the first samples after the December 1997 storm were collected on January 6, 1998 (i.e., about 3 weeks after the peak of the storm). As such, it should be emphasised that the effect of this storm has probably been underestimated.

All three storms resulted in similar effects on the characteristics of the sedimenting POM. These effects consisted of a diminution in the concentrations of all the assayed biochemical parameters, relative to the dry weight. This result is in agreement with the observations carried out in the Bay of Banyuls-sur-Mer during the November 1999 storm (Grémare et al., 2003a). It supports the negative relationship linking GSR and various biochemical descriptors of sedimenting POM collected in the Bay of Banyuls-sur-Mer; this has been interpreted as being indicative of the important resuspension processes in controlling the biochemical characteristics of sedimenting POM (Grémare et al., 1997, 1998; Rossi et al., 2003). Such a result is in good agreement with the decrease in the organic content of suspended POM during storms, as recorded by Bock and Miller (1995) near Cape Henlopen (Delaware, USA).

The decrease in the concentrations of biochemical parameters in the sediment trap material, following the 1999 storm, was more pronounced for lipids than for carbohydrates (Grémare et al., 2003a). Since lipids appear to be representative of a more labile fraction of POM than carbohydrates (Grémare et al., 2003b), these authors concluded that the decrease in concentrations of the sediment trap material resulted in an increase in the amount of suspended particles (i.e., inorganic material) and from the presence of refractory organic matter, associated with resuspended sediment grains. During the present study, the occurrence of refractory organic matter after the three storms is supported by the analysis of temporal changes in the EHAA/THAA ratios of sediment trap material. This ratio accounts for the proportion of the amino acid pool that can indeed be used by benthic deposit-feeders (Mayer et al., 1995; Medernach et al., 2001; Grémare et al., 2002). All three storms resulted in a decrease in EHAA/THAA ratio for the sediment trap material. Thus, it appears that storms not only increase particle loads, but also result in the mixing, with more refractory organic matter originating from resuspension. Based upon a series of in situ experiments, as opposed to what has been predicted previously (Sloth et al., 1996; Wainright and Hopkinson, 1997), Tengberg et al. (2003) reported a negative effect of resuspension on oxygen fluxes. The hypothesis, as put forward by these authors, related to the stirring up of the refractory POM degraded initially by bacteria within the sediment; this resulted in a poorer efficiency of the mineralisation processes. Our results confirm such a dilution and can be used to parameterise changes in the lability of suspended POM in mathematical models, assessing the effects of resuspension on oxygen fluxes (Wainright and Hopkinson, 1997).

4.2. *Impact of storms on the characteristics of sedimented POM*

In contrast to sedimenting POM, the effect of storms on sedimented POM has been largely neglected (Bock and Miller, 1995). During the present study, there were clear differences identified in the temporal changes caused by the three storms. The April 1997 and the November 1997 storms did not have any marked effect on these characteristics. Conversely, the December 1997 storm resulted in a decrease in particle size, an increase in carbohydrates, lipids and THAA concentrations, no change in EHAA concentrations and, consequently, a decrease in EHAA/THAA ratios. These results are in good general agreement with those of Grémare et al. (2003a), who reported already similar changes following the November 1999 storm. Conversely, Bock and Miller (1995) argued that storms may increase temporarily the food quality of the surficial sediments, due to the transitory deposition of small and light organic-rich particles during the transition between stormy and calm conditions. This statement was based on temporal changes in the total proteins, within the surficial sediments. During the present study, we did observe the transitory deposition of small particles after the December 1997 storm and an associated increase in THAA, but no significant changes in EHAA. Consequently, a decrease in the EHAA/THAA ratios was indicative of a reduction in food quality. Thus, it is concluded that changes in sedimented POM characteristics resulted from (1) the general relationship between particle size and organic content (Mayer, 1994) and (2) the association of mostly refractory organic matter with those transitory settled small particles. Besides the nature of the assayed biochemical parameters, part of the discrepancy between our results and those of Bock and Miller (1995) may result from the fact that in the Bay of Banyuls-sur-Mer, southerly storms are often associated with heavy rains, which may also induce important terrestrial runoffs (Grémare et al., 1997). This association may also account for differences in the duration of the relaxation periods (i.e., 1 day according to Bock and Miller (1995) compared with several weeks according to Grémare et al. (2003a, b) and the present study).

4.3. *Importance of major storms relative to those of other environmental events*

Both for sedimenting and sedimented POM, most of the biochemical concentrations and the

EHAA/THAA ratios recorded, following the December 1997 storm, were the lowest of the whole period under study; this, once again, emphasises the importance of a major winter storm in controlling the characteristics of both sedimenting and sedimented POM. This observation was especially clear for the sedimented POM, where the December 1997 storm constituted clearly the most important event of the whole of the period under study, since both heavy rainfall and the late winter phytoplanktonic bloom did not result apparently in any significant effect. The situation was less clear for sedimenting POM. Our results show that the effects of the December 1997 storm are larger than those of the sedimentation of the late winter phytoplanktonic bloom. However, the most important changes in sedimenting POM were associated with the transition between the spring–summer and the autumn–winter period, and thus linked to the October 1997 rainfalls. Here again, it should be pointed out that the effects of the December 1997 storm may have been underestimated due to sampling problems. Consequently, further studies need to provide an improved assessment of the importance of major winter storms on sedimenting POM, relative to those of other meteorological events.

4.4. Interactions of resuspension with the adsorption of ^{14}C PCB

Absorption efficiency measurements rely upon short-term experiments, involving radiolabelled contaminants. As such, the tested food sources must be labelled high enough to detect radioactivity in the different end-products of digestion. During the present study, this was achieved by mixing wet sediment and ^{14}C-TCB coated on a glass wall (Ditsworth et al., 1990; Boese et al., 1995; Murdoch et al., 1997). Using this method, there were significant differences between the adsorption rates on the tested sediment types but no significant correlation with their organic contents; this is consistent with data presented in the literature. Using the same contamination method, Murdoch et al. (1997) showed that sediment contamination levels could be predicted correctly from the amounts of PCB used for sediment spiking; they also identified the lack of any particle-size effect on sediment spiking. In summary, the mixing of wet sediment and dry PCB appears to result in contamination levels that are largely independent of the intrinsic characteristics of the sediment particles. This result is somewhat unsatisfactory, because the partitioning of the contaminant is related usually with the organic content and the particle size of the tested substrates.

There are two other main methods used to spike sediment with hydrophobic organic pollutants. The first approach consists in eluting the contaminant coated on glass beads into the recycling water (Veith and Comstock, 1975); this is probably the best method in mimicking contamination mechanisms taking place in nature. However, it is both time-consuming and expensive. Thus, it has been used only rarely and results apparently in rather low contamination levels (Gunnarsson and Rosenberg, 1996). To our knowledge, this method has never been used for comparing the adsorption of contaminants on various sources of POM. During preliminary experiments, we used a somewhat similar approach to run adsorption experiments on the two most contrasting sediment trap materials. We evaporated 14C PCB on Siporex® glass, then extracted in distilled water in a Soxhlet. We used this contaminated water to run the adsorption experiments. The results obtained were rather inconclusive. Labelling rates correlated apparently positively with sediment organic contents, but differences between labelling rates were of only a few dpm and the contamination levels were low.

The second method consisted of mixing wet sediment with the contaminant, diluted in a small volume of acetone (Landrum and Faust, 1994; Gunnarsson et al., 1996; Bruner et al., 1994). As with the first approach, this method results in contamination levels high enough to run toxicity tests and bioaccumulation experiments; it has been used also for assimilation assessment (Björk and Gilek, 1999; Bruner et al., 1994). This approach presents the main drawback of potentially affecting the biochemical characteristics of contaminated organic matter. To our knowledge, this method has never been used to assess adsorption rates of contaminants on various sources of POM,

although Gunnarsson et al. (1999) has used it to infer labelling of a sediment enriched with various sources of organic matter.

In conclusion, the lack of an effect of sediment resuspension on adsorption rates of TCB, as recorded during the present study, is in agreement with data in the published literature, obtained using the same contamination method. However, it should be emphasised that the lack of a correlation between adsorption rates and the biochemical characteristics of the tested POMs may largely derive from the contamination method which was used. At present, it remains unclear whether or not other existing protocols can be used reliably to run comparative adsorption experiments on natural sources of POM. Consequently, further developments of contamination protocols are clearly needed in order to better assess the adsorption process; this is essential for the characterisation of the fate of PCBs.

4.5. Interactions of resuspension with absorption of ^{14}C PCB

During our absorption experiments, the TCB always remained tightly sorbed to the tested sediments, the radioactive pulse lasted only 1 h, and mussels were feeding actively on sediment in suspension, defecated normally undigested food. All these observations have permitted us to be confident in our assessment of the absorption efficiencies, which were between 0.6% and 7.9%. This observation confirms that mussels can take up significant amounts of contaminants through nutrition processes. Absorption efficiencies are in the lower range (i.e., 1–30%) of those reported for zebra mussels fed with various sources of POM contaminated with two PCB congeners (Bruner et al., 1994). The data available on the absorption efficiency of sediment-associated POM, by suspension feeding bivalves, are still scarce. Nevertheless, Charles et al. (1995) reported absorption efficiencies of between 6.4% and 21.0% for *Abra ovata* fed with POM collected over a year cycle in sediment traps moored in the Bay of Banyuls-sur-Mer. However, caution should be taken in comparing these results with those of the present study, especially because the two surveys deal with bivalves with different feeding modes. Nonetheless, it should be pointed out that differences between the absorption efficiencies of bulk sedimentary POM and ^{14}C TCB should not be surprising, since contaminants must first desorb from particles before being absorbed (Brunner et al., 1994).

The mechanisms responsible for differences in the absorption efficiencies of PCBs, bound to different sources of POM (Brunner et al., 1994), are still unclear. Up until now, these differences have been documented mostly based upon the comparison between phytoplanktonic cells and sediments, i.e., two highly contrasting POM sources. To our knowledge, the present study is the first to be undertaken which compares absorption efficiencies of PCB bound to sediments, of the same kind collected at different periods of the year and corresponding to different resuspension regimes. Our results show that ^{14}C TCB absorption efficiencies differ significantly between tested sediment types. Moreover, these differences appear to be related with the organic contents of the sediment types. Indeed, the two highest values were recorded during August 1997 (Sediment Type IV, corresponding quasi-exclusively to material originating from the water column) and during February 1998 (Sediment Type X, corresponding to material resulting from the sedimentation of the late winter–nearly spring phytoplanktonic bloom). Conversely, the two lowest values corresponded to surficial sediment and sediment trap material collected after the December 1997 storm, which was the strongest during the whole period under study; it resulted in the lowest values of EHAA/THAA ratios, both in the sediment trap material and in the surficial sediment (see above). This pattern, of temporal changes in the TCB absorption efficiency, is similar to that reported by Charles et al. (1995) for the absorption efficiency of organic carbon. Moreover, during the present study, there were significant correlations between most of the biochemical descriptors of the 10 tested sediment types and corresponding TCB absorption efficiencies (even if it should be pointed out that the significance of those correlations resulted partly from the results obtained with the tested surficial sediment). Overall, our results tend to support the concept that resuspended pollutants

are absorbed at a lower efficiency by benthic organisms than those bound on POM originating directly from the water column.

4.6. Conclusions

Storms tend to increase the concentration of suspended POM in the water column through sediment resuspension. Such events also tend to reduce the quality of the suspended POM, which results in a lower absorption efficiency of the adsorbed PCB, by the suspension-feeding bivalve *M. galloprovincialis*. Our results suggest that sediment resuspension affects the transfer of PCB, in the benthic food chain, in two opposing ways: (i) it contributes to increase the PCB concentration in the water column, due to a transfer from the sediment; but (ii) it reduces PCB bioavailability, due to the low nutritional value of resuspended POM. Comparison can be made of the magnitude during resuspension events of (a) the increase in the gross sedimentation rates (ratio of 1:28) and (b) the decrease in absorption efficiency (ratio of 1:5). These patterns suggest strongly that resuspension tends to enhance the transfer of organic pollutants in the benthic food chain.

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