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Nutrient dynamics at the sediment-water interface in a Mediterranean lagoon (Thau, France): influence of biodeposition by shellfish farming activities

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Abstract

The Thau Lagoon, a French Mediterranean shallow lagoon, is a site where extensive shellfish farming occurs. The aim of the present work is to evaluate the role of this activity on nutrient exchange at the sediment-water interface in relation to organic matter (OM) sedimentation and degradation. Two stations inside (C5) and outside (C4) of the shellfish farming areas were sampled at 3 seasons. Porewater chemistry surveys and calculated diffusive fluxes were used to evaluate the trophic status of the Thau lagoon. Quantitative (Particulate Organic Carbon) as well as qualitative OM (Hydrogen Index, Carbohydrates) analyses were performed on sediments to assess OM characteristics. Results emphasized that surficial sediments at C5 are always more enriched in OM. Porewater nutrient concentrations are 10 to 20 times higher at C5 than at C4. In June 2003, the porewater profiles exhibit a sharp gradient at the bottom waters, indicating a hypereutrophic status, leading to an anoxic crisis.

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1. Introduction

The French Mediterranean coast is bordered by several shallow lagoons formed during the later stages of the Holocene transgression. These lagoons, located in a densely populated transition zone between the continent and the sea, are very fragile ecosystems, and have been heavily impacted by human activity. These lagoons, and especially Thau, suffer from eutrophication as a result of (i) excessive nutrients inputs from the catchment, (ii) very low water renewal (water residence time of about 3 months) and (iii) specific climate conditions (high temperature, limited rainfall periods, low tidal currents, Bacher et al., 1996).

Thau, the biggest of the lagoons, is the site of shellfish farming. The shellfish production zones cover 1/5 of the lagoon area and its annual production represents about 15 000 t. This production is made according to an original technique of binding oysters (or mussels) on ropes, themselves fixed on metallic structures (tables). Then, the continual immersion of oysters-mussels ropes involves an abundant epibiose development. The presence of these heterotrophic organisms (epibiose) in the water column modifies the transfer and the transformation of the organic matter within the lagoon ecosystem (Mazouni et al., 1996; 1998). For example, sedimentation fluxes have been estimated from 100 to 400 mg C.m\(^{-2}\).h\(^{-1}\) in shellfish areas. These fluxes, 2 to 4 times higher than in the areas without shellfish, can significantly increase the sedimentation rate (Grenz et al. 1992). In addition, the combination of high Organic Matter (OM) productivity, high summer temperature (25°C water surface temperature) and low wind speed induces the rapid depletion of dissolved oxygen and subsequent anoxia (Harzallah & Chapelle, 2002). Anoxic conditions develop in summer both at the sediment-water interface and at the bottom of the water column. These anoxic episodes, locally named “malaigues”, induce a high turbidity and H\(_2\)S smell during the summer months with a large shellfish mortality.

Previous studies have established that the sediment compartment is a sink for particulate phosphate (Boström et al., 1982, 1988; Caraco et al., 1990). This compartment could also become a source through the release of dissolved species under well defined pH and Redox conditions (Boers, 1991; Song & Müller, 1999). Numerous parameters influence the exchange of nutrients at the sediment-water interface and accentuate the influence of multi-environmental parameters such as bacterial activity (Gächter & Meyer, 1993), iron-hydroxide chemistry or oxygen conditions (Maine et al., 1992; De Montigny et al. 1993). Moreover, OM buried in sediments may impact the nutrient release by forming refractory organo-metallic complexes with iron and phosphorus (Paludan and Jensen, 1995; De Groot and Golterman, 1993; Hirata, 1985). Even if these complexes are subjected
to microbiological mineralisation as a carbon and an energy source, their low availability may delay
the nutrient release. If this low bioavailable OM quantity exceeds the bacterial mineralisation
capacity of the sediment, these complexes will accumulate in the sediment compartment as a
‘residual organic phosphate’ fraction (De Groot and Golterman, 1993) that will only be remobilised
with difficulty. The determination of bulk sedimentary OM content alone is therefore not sufficient
to characterize nutrient release at the sediment-water interface: measurements of OM quality and
degradation rate are needed to fully assess the extent of degradation of (and potential nutrient
release from) sediment organic components.

The aim of the present work is to discuss the role of shellfish farming on the nutrient
exchange dynamic at the sediment-water interface in two contrasting sites of the Thau Lagoon:
inside and outside of the shellfish farming areas. A seasonal porewater survey was investigated at
these two sites in Winter 2001-02, Spring 2002 and Summer 2003. The objective of this study is to
focus on OM characterization, as well quantitative (Particulate Organic Carbon) and qualitative
(Hydrogen Index, Carbohydrates) measures of organic matter content. In addition, the degradation
rate of organic matter is examined, to assess the potential influence of shellfish biodeposits on the
exchange of nutrients at the sediment-water interface.

2. Material and methods

2.1 Site

The ‘Thau’ Lagoon, a Mediterranean shallow coastal lagoon is located in the South of
France (43°20’N to 43°28’N and 3°32’E to 3°42’E). Its total surface is about 75 km² with a length
of 19.5 km and a width of 4.5 km. Its depth varies from 4 to 10 meters. At either end, the lagoon is
connected to the sea with a water residence time of about 5 months (Abadie et al., 2004).

Two different sampling stations were chosen in contrasting areas of the shallow lagoon: the
first site (station C4) was located in the middle of the lagoon (43°2.018’N, 3°36.703’E); average
depth: 8 m.) and the second site (station C5) inside of the oyster bank zone A, adjacent to
Bouzigues (figure 1; 43°25.994’N, 3°39.657’E; average depth: 8.5 m.).

Fieldtrips were carried out, at the two sampling stations, at three different times of year,
namely in (i) Winter 2001-02 (05/12/01 to 09/12/01), (ii) Spring 2002 (15/04/02 to 21/04/02), (iii)
Summer 2003 (19/05/03 to 23/05/03).
2.2 Sediment characteristics

Sediment from the two sampling sites consists of shell-rich, grey silty clay, with no obvious sedimentary structures. The granulometric distribution performed with a Malvern Laser particle sizer (Schmidt et al. accepted, 2005) indicates that sediment consists mainly of fine silt. The mean grain size varies from 10 to 20 µm (Schmidt et al. accepted, 2005). The mixed layer, at the C4 site, is restricted to the upper 3 cm. In contrast, the C5 site has a mixed layer of 10 cm depth (Schmidt et al. accepted, 2005). Composition of sediments, at both sites, has given the following composition: CaO=18.4%, MgO=1.97%, K2O=1.71%, Na2O=3.72%, Fe2O3=2.02%, Al2O3=8.54%, SiO2=26.7% (Péna et Picot, 1991; Elbatz-Poulischet, 2006, Personal Comm.)

At the C4 site, the porosity is constant with depth with a value around 0.85. In contrast, the porosity of C5 site decreases from 0.90 at the sediment surface to 0.75 at 35 cm depth (Metzger et al. accepted, 2005). At the sediment surface to 40 cm depth, pH values vary from 8 to 8.4 at the C4 site and 7.4 to 8 at the C5 site (Metzger et al. accepted, 2005). The redox potential (Eh) in the sediment surface, is seasonally constant at both sites, about 200 mV whereas the redox measured in the first 10 cm of sediment showed significant seasonal variation, Eh = 125 mV in winter and Eh = -50 mV in summer.

2.3 Sampling techniques

The pore water sampling was performed using diffusion samplers (Hesslein, 1976), with an inert polysulfone membrane of 0.2 µm porosity (Millipore, Durapore). The diffusion sampler design employed consists of a 51.5 x 22 x 3 cm Plexiglas sheet in which fixed volume (5.6 cm³) chambers are spaced at 1 cm. Each diffusion sampler has two series of 40 chambers. In order to avoid oxygen contamination in the chambers, the diffusion samplers were bubbled with nitrogen in deionised water bath before their insertion. At the C5 and C4 sites at each sampling period, two diffusion samplers were inserted in the sediments, by scuba-divers, in an area of 2 m², leaving 6 or 7 chambers above the sediment surface and 33 or 34 below. The equilibration time required for the diffusion sampler is 3 weeks according to Bally et al. (2005). The diffusion samplers have been inserted, at both sites, in winter (09/12/01), Spring 2002 (02/03/02) and Summer 2003 (23/05/03) and they have been removed after 3 weeks equilibration, except in summer (26/06/03) after one month equilibration. Pore water samples were removed using plastic syringes, in a glove box filled with nitrogen to prevent the co-precipitation of phosphate with iron-hydroxide. The samples were acidified with 1N HCl and stored in conditioned 5 ml haemolysis tubes which were frozen until
analysed for Soluble Reactive Phosphate (SRP), ammonium (NH$_4^+$) and Dissolved Organic Carbon (DOC) determination.

Sediment cores (diameter = 12 cm and length = 25 cm) were sampled by scuba-divers with Plexiglas® cores, at the C4 and C5 stations, in April 2002. Coring operations were carried out very carefully in order to prevent any disturbance of the top sediment where the water content is very high (96 % at the sediment surface; 86 % at 20 cm depth). Immediately after the sampling, sediment cores were cut into slices of 0.5 cm (for the top 2 cm) and into slices of 1 cm until 19-20 cm for C4 and until 21-22 cm for C5. The sediment sections obtained were stored in plastic bags filled with nitrogen and immediately frozen.

2.4 Sediment analysis

Rock-Eval pyrolysis (Espitalié et al., 1985) was used for organic matter quantitative and qualitative characterisation. Two main parameters may be provided by this technique: Total Organic Carbon (“POC”) and the Hydrogen Index (HI) which depends on the hydrogen OM content and the corresponding C/H (Espitalié et al., 1985). The POC and the HI are expressed in % Organic Carbon weight and mg of Hydrocarbons per g POC (or mg HC .g$^{-1}$ POC), respectively.

Rock-Eval® pyrolysis has been performed with an RE6 device of Vinci TechnologiesÒ. The analyses were carried out on 100 mg of crushed samples with a temperature of 200° C (20 min) up to 600° C at 25° C min$^{-1}$ under a N$_2$ flow, followed by oxidation at 600° C for 7 min under an oxygen flow. All the analyses were performed in duplicate. Reproducibility determined after 68 analysis with the IFP 55000 reference material was +/- 2 % for POC content and +/- 6 % for HI (Disnar et al. 2003).

Carbohydrate analyses were performed on the top (0-2 cm) and the base (13-22 cm in C5 core and 10-20 cm in C4 core) in order to characterize the availability of carbohydrate at the sediment-water interface in contrast with deeper sediment. The procedure used for carbohydrate analysis derives from previous works (Bethge et al., 1966; Oades et al., 1970; Modzeleski & Laurie, 1971; Cowie & Hedges, 1984a). Briefly after hydrolysis (100°C; 3h.) with 0.5 M H$_2$SO$_4$ and cooling, an internal standard (6-deoxy-D-glucose; Wicks et al., 1991) is added to the hydrolysate. The anomeric carbohydrate mixture is equilibrated with lithium perchlorate (0.2 %) in pyridine (Bethge et al., 1966) and silylated with N,O-Bis(trimethylsilyl)trifluoroacetamide. Finally, the silylated carbohydrates were analysed on a Perkin-Elmer™ Auto System XL gas chromatograph. Peaks were identified through retention times and quantified using a standard mixture of eight neutral monosaccharides, namely, arabinose, rhamnose, ribose, fucose, xylose,
mannose, galactose and glucose. Quantification was based on one of the major and better-resolved anomeric peaks given by each studied compound (Bethge et al., 1966). Total carbohydrates contents were calculated as the sum of the compounds identified and quantified. Analytical errors varied between 2.6 and 13% depending on the type and the abundance of the compound considered (Ogier et al., 2001).

2.5 Dissolved component analysis

Phosphate (Soluble Reactive Phosphate, SRP) and ammonium (NH$_4^+$) were measured using a colorimetric method (Stainton et al., 1977) with a Bran+Luebbe™ auto-analyser Continuous Flow Analysis, according to the methods of Treguer & Le Corre (1975). The detection limits are 0.45 µM and 0.55 µM for SRP and ammonium, respectively. DOC was analysed with a Shimatzu 5050™ TOC analyser with a detection limit of 0.83 µM. Measurement precisions are 5% RSD determined from repeated measurements (5 times) of the same sample and standard samples (Bally et al., 2004).

2.6 Method for calculating Nutrient diffusive fluxes

The calculated diffusive fluxes were calculated using the Fick’s first law adapted for sediments (I):

$$ J_s = \phi \times D_s \times \frac{dC}{dx} \quad (I) $$

$J_s$: Calculated diffusive flux (mmol.m$^{-2}$.d$^{-1}$)

$\phi$: Sediment porosity (dimensionless)

$D_s$: Diffusion coefficient of the species in sediment (m$^2$.d$^{-1}$)

$\frac{dC}{dx}$: Profile gradient (mmol.m$^{-4}$)

The diffusion coefficient in water ($D^0_w(X)$) was corrected from the Stokces-Einstein formula (II & III) given in Li & Gregory (1976):

$$ D^0_w(NH_4^+) = 19.8 + 0.4(T-25) \quad (II) $$

$$ D^0_w(PO_4^{3-}) = 7.36 + 0.16(T-25) \quad (III) $$

with
**3. Results**

3.1 *Bulk organic matter characterization (POC and HI)*

At C5, POC content decreases sharply from 4.4% at the top of the core to 3.3% at 1.75 cm depth. This decreasing trend continues smoothly down to 8 cm depth to reach a value of 1.7%. Below this depth, POC content remains rather constant around 2% down to the base of the core (Figure 2a, Table 2a). The HI values, remain constant with values around 370 mg HC g\(^{-1}\) POC in the upper 5 cm of the core. Below this depth, HI values first decrease downward to reach a value of 323 mg HC g\(^{-1}\) POC at 8 cm depth (Figure 2a), then fluctuate between 217 and 335 mg HC g\(^{-1}\) POC (Table 2a).

At C4, POC contents increase slightly from 3% at the top of the core to 3.2% at 2 cm depth. Between 2 cm to 8 cm POC decreases with depth to reach a value of ca. 2.6% at 8 cm depth and below this depth values remain constant down to 11 cm depth. Below 11 cm, POC contents vary between 2.8 to 4.2% (Figure 2b, Table 2b). Hydrogen Index (HI) values decrease downcore from 354 mg HC g\(^{-1}\) g POC at the sediment water interface to 303 mg HC g\(^{-1}\) g POC at the base of the core (20 cm depth) (Table 2b).

3.2 *Carbohydrate analysis*

At C5, the total neutral carbohydrate concentrations decrease sharply with depth with a value of 3.6 mg g\(^{-1}\) (dry weight) near the sediment surface, 3.2 mg g\(^{-1}\) at 2 cm depth and only 1.3 mg g\(^{-1}\) at 22 cm depth (Table 2a). These compound concentrations indicate that carbohydrates contribute only to 7 to 12% of POC. In the surficial sediments (0-2 cm) individual neutral carbohydrates are dominated by rhamnose (24%) and fucose (21%), followed by glucose (15%),...
galactose (12 %), xylose (11 %), arabinose (9 %) and mannose (8 %) (Figure 3a; Table 2a). Ribose which was often below the detection limit is thus not discussed further. At the base of the core (13-22 cm), neutral carbohydrates are dominated by fucose (24 %) and rhamnose (18 %), followed by glucose (14 %), arabinose (14 %), xylose (12 %), mannose (9 %) and galactose (9 %) (Figure 3a).

At C4, the total neutral carbohydrate concentration presents a rather high top core (0-2 cm) value of 2.5 mg g\(^{-1}\). At 10 cm depth, carbohydrate concentrations are only 1.8 mg g\(^{-1}\). Below this depth, values increase slightly with large variations between 2.1 and 3.2 mg g\(^{-1}\) (Table 2b). These compound concentrations indicate that carbohydrates account for 5.4 % to 10.7 % of the POC. In the surficial sediment (0-2 cm) the weight percentages of individual neutral carbohydrates are dominated by glucose (23 %) and fucose (22 %), followed by xylose (13 %), galactose (13 %), arabinose (11 %), mannose (10 %) and rhamnose (8 %) (Figure 3b, Table 2b). Ribose is present at low concentrations and often below the detection limit. At the base of the core (10-20 cm), the weight percentages of individual neutral carbohydrates in sediments are dominated by glucose (20 %), fucose (19 %) and rhamnose (17 %), followed by xylose (15 %), galactose (14 %), mannose (9 %) and arabinose (8 %) (Figure 3b).

3.3 Nutrient porewater profiles

Soluble Reactive Phosphorus (SRP)
At the C5 sampling station, in the bottom waters (up to 7 cm above the sediment-water interface), SRP concentrations were at the lowest in Winter (January 2001, Figure 4a), and Spring (April, 2002, Figure 4b); they varied between 0.1 and 4 µM. In contrast, in Summer (June 2003, Fig. 2c), at the same site, SRP concentrations reached very high levels in the bottom waters: 20 µM at 10 cm over the water-sediment interface, up to 60 µM 5 cm below and finally 10 times higher values (600 µM) at the sediment interface. Always in summer, SRP concentrations decreased slowly below the sediment surface to reach 100 µM at 5 cm depth.

At the C4 sampling station, SRP concentrations reached about 1 µM in the bottom waters. However, these concentrations showed no change with depth. Moreover, no SRP concentration gradient was observed in the sediment at this site throughout the year.

Ammonium concentrations (NH\(_4^+\))
At the C5 sampling station, NH\(_4^+\) fluctuated between 15 to 30 µM in Winter (Figure 5a) and Spring (Figure 5b) in the bottom waters. In contrast, in Summer (June 2003, Figure 5c) NH\(_4^+\) increased sharply, to values of 500 µM, i.e. about 10 times more than during the previous sampling periods.
NH$_4^+$ profiles are quite similar to those obtained for the other dissolved species (i.e. SRP and DOC). A gradient was apparent between –5 and –15 cm, except for the Summer period (June 2003), where the concentration gradient was located in the bottom waters. The concentrations reached values of about 8000 µM at the sediment-water interface. Thus a concentration gradient is present in the bottom waters, but not in the sediment under the interface.

The porewater chemistry at the C4 sampling station is much more uniform with depth. At this site concentrations levels were low (< 50 µM) and there were no concentration gradients, the profiles being vertical (Figure 5d, 5e).

**Dissolved Organic Carbon (DOC)**

At the C5 site the bottom waters (5 cm above the sediment-water interface) showed DOC contents that increased sharply in Summer (June 2003, Figure 6c) to reach values up to 10 000 µM. Then, DOC concentrations in the bottom waters were about 20 times higher than in Spring and Winter. In addition, a concentration gradient is observed in the bottom waters but not in the sediment as usually expected. During Winter and Spring at C5 (Figure 6a, 6b) DOC concentrations increased continuously with depth in the sediments, to delineate a gradient down to 15 cm. Below, DOC concentrations remained constant, with values around 2000 µM. The variations of DOC concentrations with depth were better marked in Spring than in Winter.

In Summer (June 2003, Figure 6c), a sharp DOC concentration gradient was observed in the bottom waters: DOC concentration raised up to 20 000 µM of Carbon. Then, there is not marked concentration change with depth in the sediment.

At the C4 sampling station, DOC profiles showed the same shape than those already described for SRP i.e. without any change all over the 30 cm of water and sediment investigated. DOC concentrations reached about 500 µM in the bottom water and in the sediment in Winter and Spring (Figure 6d, 6e).

### 3.4 Diffusive nutrient fluxes

Fluxes have been calculated from concentration gradients observed for the nutrient profiles (Figures 4, 5 and 6). During Winter and Spring, these gradients were located between 5 and 15 cm below the sediment-water interface. Then, the fluxes are directed from 15 cm to 5 cm, that is from the more to the less concentrated layer. At C5 station, calculated flux values fluctuated from 0.62 to 1.0 mmol.m$^{-2}$.d$^{-1}$ NH$_4^+$ and from 0.3 to 0.14 mmol.m$^{-2}$.d$^{-1}$ SRP for Winter and Spring, respectively (Table 3). In contrast, at C4, the calculated fluxes values fluctuated from 0.2 to 0.05 mmol.m$^{-2}$.d$^{-1}$
NH$_4^+$ and from 0.03 to 0.02 mmol.m$^{-2}$.d$^{-1}$ SRP, for Winter and Spring, respectively (Table 3). Whatever the season, a spatial variation is evident, with fluxes about 10 times higher at C5 than at C4.

In Summer (June 2003) at C5 site, concentration gradients were not located below the sediment water interface (- 5 cm depth), but in the bottom waters (Figures 4c and 5c). High NH$_4^+$ and SRP flux values have been derived from these concentration gradients in the bottom waters (10.7 mmol.m$^{-2}$.d$^{-1}$ and 0.96 mmol.m$^{-2}$.d$^{-1}$ or respectively for NH$_4^+$ and SRP) (Table 3).

4. Discussion -

4.1 Nutrient profiles, a tool to assess ecosystem trophic levels

As generally observed in Mediterranean shallow coastal lagoons, the phosphate inputs originated from urban and agricultural effluents. In the Thau lagoon, these inputs have decreased by a half from 1971 to 1996 as a result of an improvement in wastewater treatment on the Thau catchment-coastal lagoon system (La Jeunesse & Elliot 2004). Over 30 years monitoring of water-column chemistry highlighted the decrease of phosphate concentrations. In the Seventies, the average phosphate concentration was 6 µM, increasing to 10 µM during anoxic summer periods. In contrast, the average phosphate concentration, actually measured in the water column, fluctuated from 0.04 to 1.2 µM. The decrease of 90% of annual mean phosphate concentration in the water column from 1971 to 1994 (Souchu et al. 1998) can be explained not only by the reduction of domestic effluents but also by changes in the land-use, through a dramatic decrease in area planted with vineyards (La Jeunesse et al. 2002). Phytoplanktonic biomass is present in higher concentrations in the lagoon than in sea water, with an average chlorophyll a of 2 µg/l with maxima of 5 µg/l in summer periods (Casellas et al. 1990). This primary production depicts a high trophic level in the lagoon, especially in the shellfish (Casellas et al. 1990, Picot et al. 1990; Chapelle et al. 2000). During summer, OM degradation and nutrient release at the sediment-water interface lead to a high phytoplanktonic activity, particularly in shellfish areas (Casellas et al. 1990; Plus et al. 2001, Souchu et al. 2001).

The knowledge of porewater chemistry is a prerequisite for assessing nutrient dynamic at the sediment-water interface. The presence of significant amounts of OM buried in the sediments is probably the main factor influencing nutrient fluxes at the sediment-water. The shape of nutrient profiles reflects the presence of OM layers undergoing mineralisation and thus acting as a source of dissolved nutrients. In coastal shallow lagoons, OM inputs originate mainly from land, in the form of plant residues, or in situ, in the form of phytoplankton production and shellfish biodeposits.
Indeed, the nutrient profiles led us to follow OM mineralization over depth. At the C5 station the sediments are anoxic. The depth of oxygen penetration is, at a maximum, limited to 5 mm in winter and 0.5 mm in summer (Dedieu et al. accepted 2005). This anoxic sedimentary environment supports anaerobic OM degradation, with oxidation reactions successively driven by the reduction of Mn and Fe oxides, then of sulphates and finally by methane fermentation processes (Song & Müller, 1999). All these processes are known to release monomeric low molecular weight compounds such as organic acids which may acidify the porewaters (Burdige & Gardner, 1998). The measurements of porewater redox-sensitive species at the C5 station (Metzger et al. accepted 2005) confirm the establishment of such anaerobic OM degradation. Another consequence of this process is the greater acidity of the sediment at C5 station than at C4 station over the upper 40 cm (Metzger et al. accepted, 2005). Moreover, these observations are consistent with the presence of more labile OM at the C5 station than at C4.

Discussion on the trophic level of aquatic ecosystems is often approached using porewater gradients. In oligotrophic ecosystems, the nutrient profiles are vertical, i.e. the concentrations remain at the same level throughout the water column and the sediment. In contrast, in eutrophic ecosystems nutrient profiles exhibit sharp gradients near the sediment-water interface. At C4 station, especially in winter, nutrient profiles exhibit a vertical shape, reflecting low biological activity loading to low input of biodegradable OM. In contrast, at the C5 station, nutrient profiles exhibited high gradients. The nutrient concentration increased down to 10 cm below the sediment-water interface (Figs 2c, 3c, 4c). Above, the nutrient profiles are vertical, with the same concentration levels in the water column as in the sediment. One hypothesis for this vertical profile is the homogenisation of the sediment, explained by the 10 cm upper mixed layer in relation to rather high biological activity at the C5 station (Schmidt et al. accepted 2005).

Previous works on porewater nutrient chemistry, in the Thau lagoon (Mesnage, 1994, Mesnage & Picot, 1995, Metzger et al. accepted, 2005), have demonstrated that nutrient concentrations (SRP and ammonium) were three times higher inside the oyster banks than outside. Moreover, a sharp concentration gradient that appears below the sediment-water interface (-2 cm) demonstrates nutrient accumulation in porewaters (Mesnage, 1994). The impact of oysters on phosphate accumulation in surface sediments under the influence of their pseudo-faeces was also demonstrated: sediments inside of shellfish bank zone (zone B, Figure 1) have greater concentrations of particulate phosphorus than those outside of the shellfish farming zone (Chapelle et al. 1994).
In contrast with these previous works (Mesnage & Picot, 1995; Metzger et al. Accepted 2005), the most relevant fact in the present study, is the specific shape of nutrient porewater profiles measured in summer 2003. The concentration gradient is not limited to the first 10 cm below the water-sediment interface, instead, it extends above the sediment-water interface, at the base of the water column. This kind of profile (Figures 4a, 5a and 6a) is very close to the theoretical one, described by Enell & Löfgren (1988), for shallow eutrophic lagoons exposed to high organic matter inputs (fish farming, phytoplankton or macro-algae sedimentation following the spring bloom). Thus, geochemical porewater results at the C5, indicate a great seasonal variation evidenced by important increase of nutrient concentrations from Winter to Summer and provide a useful tool to evaluate the aquatic ecosystem trophic condition, the difference between Summer and Winter nutrient concentrations being more pronounced in eutrophic ecosystems.

4.2 Buried organic matter: influence on nutrient release

The specific shape of nutrient porewater profiles (Summer 2003) is also supported by the data obtained on organic matter buried in the sediments. Indeed, POC values (3 to 4 % in the surficial sediments), are in the range of those found in carbon-rich aquatic ecosystems especially in other impacted lagoons (Crawford et al., 2003). Such high values suggest a probable contribution of shellfish faeces to the OM buried in sediments. According to the literature, Mediterranean surficial sediments present POC contents lower than 1 % : 0.7 to 0.9 % in Gulf of Lions (Accornero et al., 2003), 0.3 to 0.82 % in Cretan sea (Gogou & Stephanov, 2004). Other shallow lagoons in the world also present POC values in the same 0.1 - 2.8 % range (Rigollet et al., 2004; Paez-Osuna et al., 1998). When surficial lagoon sediments present higher POC values, it is always in a particular context e.g. at sewage outfall (Mudge et al., 1998) or within shellfish farming zones (Crawford et al., 2003).

The combination of a higher sedimentation rate (Grenz et al. 1992) and higher POC values in the sediment (Figure 4a) ensure a much higher OM flux at C5 than at C4. While the significance of POC content is straightforward, the HI index is related to the hydrogen content of organic matter and is both dependent on its biological origin (marine and/or terrestrial) and its degradation state (Espitalié et al., 1985). As generally assumed, a HI value higher than 600 mg HC g⁻¹ POC in immature sediments, represents a lacustrine material enriched in hydrocarbonaceous moieties. An organic material with a HI ranging between 300 and 600 mg HC g⁻¹ POC often originates from algae (e.g. phytoplankton). Finally, a material with a HI lower than 300 mg HC g⁻¹ POC is poor in hydrocarbonaceous moieties and usually represents an organic material derived from higher plants.
(Espitalié et al., 1985; Tissot & Welte, 1984). Organic material of the two sampling stations present HI values comprised between 300 and 350 mg HC g⁻¹ POC suggesting a dominant contribution from autochthonous phytoplanktonic production. The slight difference in HI values between C5 and C4 top sediment (0-2 cm, Figure 2) could be explained by an OM content richer in hydrocarbonaceous moieties and thus supposedly more able to sustain microbial activity at C5 than at C4 sampling station.

At the C5 sampling station, the sharp POC decrease may be the result of bacterial OM mineralisation, which is enhanced by the bioturbation in the surface sediment. Indeed, a mixed layer of 10 cm in thickness has been reported at the top of the core (see section 4.1). These processes (assimilation or mineralisation) easily explain why a decrease in HI values (Figure 5) is observed down to 10 cm. Moreover, OM degradation is known to favour a preferential attack of hydrogen-rich compounds (Anderson and Johns, 1986). In addition, DOC is considered as an OM degradation product (Chang and Berner, 1999). Thus, high DOC fluxes measured in Summer at C5 may attest a high OM degradation rate. All these processes may explain this sharp HI decrease and the resulting high nutrient availability.

In contrast, at the C4 site, the surface mixed layer (0-3 cm), exhibits constant values of POC and HI. This confirms the refractory property of OM and/or the balance between OM degradation and OM buried in sediments. Below 3 cm depth, a concomitant and progressive decrease in POC and HI values may result from a slight consummation of OM by microbial community through diagenesis. To conclude, the C5 site, in contrast with the C4 site, revealed both stronger bacterial activities enhanced by the availability of nutrients and thicker mixing layer implying a significant oxidation of OM. The consequence is an increase of DOC fluxes resulting from the OM degradation.

OM quantification and characterisation by POC and HI measurements have been supplemented by neutral carbohydrate analysis. Carbohydrates are usually present in lower quantities in phytoplankton (and bacteria) than in land plants where they are dominant especially as structural components, namely cellulose, hemicelluloses and pectin. However, in all cases they present the advantage of being easily degraded which makes them good tracers of microbial activity. The rather low carbohydrate content of the studied sediments indicates that these compounds have most probably been already actively recycled in the water column.
The major possibilities of distinction of potential OM sources through neutral carbohydrate analysis are summarized in Table 4. The high levels of rhamnose (followed by fucose) in the upper C5 sediment layers were attributed to heterotrophic microorganisms that developed in bottom waters, mostly at the expense of the primary production (mainly diatoms and cyanoabacteria). The decrease of rhamnose in the sediment was attributed to an easier recycling of the microbial material than that of the primary production, fucose included. Similar features to those recorded at C5 have recently been observed in an eutrophic lake (Ogier et al., 2001). Indeed, the predominance of non-ubiquitous compounds such as fucose and rhamnose, strongly suggest an autochthonous organic production from phytoplankton and/or bacterial material (e.g. Moers et al., 1990).

Therefore, quantification of OM, through carbohydrate, depicts a higher OM accumulation as well more biodegradable at the C5 station than in C4. Thus, the rapid turn-over of autochthonous OM (phytoplankton, oyster-mussel faeces, micro-organisms...) should impact the dissolved exchange processes at the sediment water interface of the Thau lagoon.

### 4.3 Diffusive nutrient fluxes, relationship with sedimentary OM load

Whatever the season, calculated diffusive flux values were always higher at C5 than at C4. In summer (June 2003) at C5, diffusive nutrient fluxes were 10 times higher than during other seasons. This could be primarily explained by the quantity and the quality of the OM buried in the sediments. Other flux measurements, in summer, in an area also subject to biosediment deposition (Oyster bank zone: B, Figure 1) did not give such high flux values (Mesnag 1994). Furthermore, concentrations gradients existed in sediments but not in the bottom waters, except in summer (Metzger et al. accepted, 2005). The later discrepancy could probably be explained by the fact that our porewater survey (retrieval of dialyser sampler) occurred one month later after that of Metzger et al. (accepted, 2005).

Combined with the prevailing high temperatures, low wind and limited water circulations, the nutrient gradient in the bottom waters can be taken as an indicator of the dystrophic status of the ecosystem preceding the onset of an anoxia episode (“malaigues”). In addition, oyster farming zone A (Fig. 1) is less exposed to the winds. The water exchange between the lagoon and the Mediterranean Sea is limited by two narrow mouths and drained by numerous, small temporary rivers. These hydrological conditions create a weak renewal of water in the Thau lagoon (Bacher et al., 1996), which is insufficient to remove the organic matter (faeces, pseudo-faeces) produced by oysters. The impact of oyster farming installations on the sedimentation rate and current speed has been demonstrated and evaluated. Previous studies have shown that biodeposits increased the sedimentation rate and that current speed can be reduced by around 60 % (Grenz et al. 1992).
Recent modelling of anoxia crisis in the Thau Lagoon, has defined the main controlling factors: wind speed and the presence of oyster tables (Chapelle et al., 2001). Moreover, the anaerobic degradation of organic oyster biodeposits is enhanced by the semi-enclosed area (lower current efficiency) and the increase of the water column temperatures during Spring and Summer months. Trophic status as well as nutrient gradients in the bottom waters depict the particular situation preceding an anoxia crisis. As mentioned above, such a crisis effectively occurred with a high intensity, in August 2003, in the Thau Lagoon.

5. Conclusion

Higher deposition rate and higher OM concentration of both POC and DOC, result in greater OM fluxes at the C5 station in the shellfish production zone, than at the C4 site in open waters. This geochemical discrepancy between both these two sites is true whatever the season and could be, at least primarily, simply explained by the contribution of oyster faeces to sediment load.

The Hydrogen Index that gives a rough estimate of quality of the OM depicts a material richer in hydrocarbon and more biodegradable at C5 station than at C4. This is also in agreement with the porewater chemistry which revealed nutrient fluxes to be always 10 times higher at C5 than at C4. Moreover, qualitative analysis of OM through neutral carbohydrates confirms the labile character of OM in the surficial sediment at this site (C5). Indeed, fucose and glucose, the main carbohydrates measured in the surficial sediments suggest in situ phytoplanktonic production to be the main OM source.

Porewater profiles revealed the trophic status at C5 station. At this site, nutrient profiles showed a deep seasonal variation between winter and summer. Sharp gradients that have been demonstrated below the sediment-water interface also extended above, in the bottom waters, in summer. The presence of such nutrient gradients at the bottom of the water column, in June 2003, is an indication of an impending anoxia crisis, which occurred in August 2003 immediately after our field survey.

All our results (particulate and dissolved organic species) indicate a biogeochemical gradient between the C5 and C4 stations. Indeed, nutrient exchange dynamics are higher at C5 station, demonstrating the impact of oyster farming in the buried labile OM. With its rapid turn-over, in situ OM production controls nutrient exchange at the sediment-water interface.

6. Acknowledgements
This study was supported by a national French research program PNEC-ART1 “Microbent”. We acknowledge Ifremer-Sète for providing the field-laboratory and a research boat. We would also like to thank: B. Bombled, J-J. Bourrand, D. Jézéquel, G. Sarasin, and the “COM” team for their assistance and specially for diving during the cruises, Dr. John Taylor for his very helpful comments and corrections of the English translation and D. Kéravis for Rock-Eval pyrolyses.

References


Table 1

Temperature and porosity used in the flux calculation

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<th>June</th>
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<td>C4</td>
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Table 2
Particulate Organic Carbon (POC), Hydrogen Index (HI) and Total neutral carbohydrate composition of sediments at the (a) C5 sampling station inside oyster farming area and (b) C4 sampling station outside oyster farming area, n.m : not measured

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<th>TCH2O (mg/100 mg POC)</th>
<th>POC (%)</th>
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Table 3

Calculated diffusive fluxes (Js) of Ammonium (Js(NH$_4^+$)) and Phosphate (Js(PO$_4^{3-}$)) at C5 sampling station inside oyster farming area and C4 sampling station outside oyster farming area; nm : not measured

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<td>$J_s$($NH_4^+$) mmol.m$^{-2}$.d$^{-1}$</td>
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<td>$J_s$($PO_4^{3-}$) mmol.m$^{-2}$.d$^{-1}$</td>
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Table 4
Literature sources for potential sources of Organic Matter (OM), through Neutral Carbohydrate association

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<td>Terrestrial plant tissues</td>
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Figures Caption -

Figure 1 : Location of the study site : the « Thau » Lagoon (France).

Figure 2 : Particulate Organic Carbon (POC %) and Hydrogen Index (HI) in mg Hydrogen Carbon g⁻¹ Particulate Organic Carbon (mg HC g⁻¹ POC), (a) at C5 station inside oyster farming area and (b) at C4 station outside oyster farming area.

Figure 3 : Average weight percentages of individual neutral carbohydrates, (a) in C5 station inside oyster farming area at the surficial sediment (0-2 cm) and at the end of the core (13-22 cm); and (b) in C4 station outside oyster farming area at the surficial sediment (0-2 cm) and at the end of the core (10-20 cm).

Figure 4 : Porewater profiles of Soluble Reactive Phosphate (SRP) versus depth at the C5 station inside oyster farming area (a) in winter, (b) in spring, (c) in summer; and at the C4 station outside oyster farming area (d) in winter, (e) in spring. Two diffusion samplers have been deployed at each season and sampling station giving two profiles except in winter at C5 station inside oyster farming area (a).

Figure 5 : Porewater profiles of ammonium (NH₄⁺) versus depth at the C5 station inside oyster farming area (a) in winter, (b) in spring, (c) in summer; and at the C4 station outside oyster farming area (d) in winter, (e) in spring. Two diffusion samplers have been deployed, giving two profiles, at each season and sampling station except in winter and spring at C5 station inside oyster farming area (a, b), in winter at C4 station outside oyster farming area (d).

Figure 6 : Porewater profiles of Dissolved Organic Carbon (DOC) versus depth at the C5 station inside oyster farming area (a) in winter, (b) in spring, (c) in summer; and at the C4 station outside oyster farming area (d) in winter, (e) in spring. Two diffusion samplers have been deployed at each season and sampling station.
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Figure 5: Porewater profiles of ammonium (NH$_4$+) versus depth at the C5 station inside oyster farming area (a) in winter, (b) in spring, (c) in summer; and at the C4 station outside oyster farming area (d) in winter, (e) in spring. Two diffusion samplers have been deployed, giving two profiles, at each season and sampling station except in winter and spring at C5 station inside oyster farming area (a, b), in winter at C4 station outside oyster farming area (d).
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