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Role of suspended particulate material on growth and metal bioaccumulation in oysters (*Crassostrea gigas*) from a French coastal semi-enclosed production area, Arcachon Bay

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Abstract :

Arcachon Bay is a prominent oyster production area on the coast of Western Europe, and is subject to chemical contamination including by trace metals. Recently, the national “mussel-watch” monitoring network – using local bivalves as semi-quantitative bioindicators of coastal chemical contamination – highlighted a significant increase in copper (Cu) concentrations in oysters from this bay. Here, we conducted a one-year multi-compartment and multi-parameter field study to investigate some aspects of the surrounding environment of oysters that could explain their metal bioaccumulation. Sediment, seawater (through punctual and passive sampling), particles (suspended particulate material of selected sizes, including trophic resources for oysters) and transplanted oysters were regularly collected at two contrasted sites of the bay (i.e. under continental versus more oceanic influence). These matrices were characterised for their total Cu, zinc (Zn), nickel (Ni), cadmium (Cd) and lead (Pb) concentrations. Several physico-chemical and biological parameters (e.g. salinity, particle loads, oysters' growth rate and condition indices, carbon and nitrogen stable isotope compositions, etc.) were also analysed. Overall, sediment, particles and oysters from the outermost site had slightly lower δ13C values, confirming the more oceanic influence in this part of the bay. Among organic particles, although dinoflagellates tended to be more abundant at the outermost site while ciliates were more abundant at the innermost site of the bay, the two sites did not differ in mean total microphytoplankton and diatom densities. However, the variations observed for most of the other parameters studied show that oysters located near the continental shore are exposed to higher loads of particles in general, and to higher metal contents in the dissolved phase and “bulk” seawater (dissolved plus particulate phases) during the year. While fluvial inputs and continental/urban run-offs are suspected sources of anthropogenic metal inputs into the bay, (fine) sediment particle resuspension appears to be a likely major contributor to metal release and hence to the contamination of the bay including oysters. The seagrass beds' decline in the last two decades in the bay could also have decreased the potential of fine particle retention in the sedimentary stratum. Concomitantly, oysters presenting the highest metal concentrations were those with the lower growth rates and condition indices, which may be induced by physical constraints, high amounts of pseudo-faeces produced and/or reduced food and energy acquisition by oysters due to high particle loads in the water column. The findings of this study also imply that using raw metal concentrations in bivalves to monitor marine coastal contamination can lead to misleading interpretations if potentially great spatial variations in bivalve growth rates or condition indices are not considered. Finally, peculiar trends were observed for Cu compared to other metals, highlighting the need of further studies to fully address the Cu contamination in this marine system (e.g. specific sources of contamination for this metal in the bay). This study more broadly raises the issue of potentially man-induced ecological changes (e.g. modification of natural habitats) and their consequences on metal transfer and physiological performance of marine biota.

Highlights

► A multi-compartment and multi-parameter study to unravel oyster metal contamination. ► Higher contamination found in dissolved and particulate seawater phases inside the bay. ► Contamination likely explained by river flows and tidal resuspension of sediments. ► Higher particle loads observed with higher oyster metal concentrations and lower growth. ► Differences in oyster bioaccumulation patterns for copper compared to other metals.

Keywords : North-East Atlantic, temperate marine system, Pacific oyster, bivalve mollusc, inorganic contaminants, trace elements

1. Introduction

Marine coastal ecosystems represent important areas from both human and ecological perspectives. However, they often are the main final sink of chemical contaminants, especially those exported via continental run-off (Guéguen et al., 2011; Lu et al., 2018). Among chemicals, trace metals occur naturally in Earth’s components, including living organisms. Some metals have essential biological functions within a narrow range of optimal concentrations (essential elements, e.g. copper, zinc), while others trace metals have no known biological role (non-essential elements, e.g. lead, mercury). Non-essential elements are recognized for their toxic effects on aquatic organisms even at low environmental concentrations, while essential elements may be either deficient or toxic at low or high concentrations (Mason, 2013). Most of metals have been extracted from deep geochemical reservoirs (e.g. ore and coal deposits) and used for a long time by humans for several purposes, including energy, transport, health, technology, etc. Nowadays, it is estimated that anthropogenic fluxes (e.g. transport, harbours, industrial activities and major coastal cities) surpass their natural counterparts (e.g. volcanism, soil erosion and forest fires) for most trace metals (Sen and Peucker-Ehrenbrink, 2012).

Trace metal contamination and its potential toxicological and ecological impacts in marine systems are constantly evolving and still insufficiently known. Hence, it is imperative to monitor anthropogenic contamination and its associated biological effects for the implementation of effective environmental management strategies (Lu et al., 2018). Along coasts and in littoral marine environments worldwide, the so-called “mussel-watch” biomonitoring programs have been useful to detect and monitor spatio-temporal variations in the concentrations of many environmental chemicals (e.g. trace metals, radionuclides, polycyclic aromatic hydrocarbons (PAHs), polychlorobiphenyls (PCBs), dioxins and furans, hexachlorobenzene, dieldrin, lindane, triazines, polybrominated diphenyl ethers, etc.). In France, the national mussel-watch program called ROCCH (“Réseau d’Observation de la Contamination Chimique”, ex RNO) has used wild local bivalves as semi-quantitative bioindicators of the chemical contamination status and trends along the national coastline since the end of the 1970s (Claisse, 1989). Due to their location close to the coast, aquaculture areas including oyster and mussel production sites are particularly concerned about chemical contamination. Moreover, chemical contamination has been considered one of the potential factors responsible for high mortality events and reproduction impairment in some bivalve species, including the Pacific oyster *Crassostrea gigas* (Samain and McCombie, 2008). This bivalve species is one of the foremost aquaculture resources on a worldwide level and France is the leading European producer (Buestel et al., 2009; Guéguen et al., 2011).

Marine animals including bivalves are exposed to trace metals by different routes. Depending on their bioecological characteristics (e.g mobile or sessile, benthic or pelagic, filter-feeding or predatory organisms, etc.) and on the properties of the contaminant itself (e.g hydrophobicity), organisms can uptake contaminants from particulate and dissolved phases derived from the sediment or the water column, and from their food. The latter pathway (trophic route) remains the main route of exposure and intake of metals by bivalves (Ke and Wang, 2001; Metian et al., 2009a, 2009b; Wang and Fisher, 1996) and the assimilation efficiency of metals by bivalves can vary greatly according to i) the size and nature of the particles and/or phytoplankton species ingested by bivalves, ii) the chemical composition of the particles themselves or iii) the subcellular distribution of metals in these trophic sources (Amiard-Triquet et al., 2006; Metian et al., 2020; Pan and Wang, 2011; Wang and Fisher, 1996). Finally, the assimilation efficiency also depends on the physiology of organisms and on the regulatory mechanisms they have developed to counteract the toxicity of certain non-essential metals, especially in the case of essential metals (Pan and Wang, 2009; Wang and Rainbow, 2010). To assess the processes leading to metal bioaccumulation by a bivalve species in situ, it is therefore crucial to characterise the metal contamination of their surrounding environment (sediments, water column including both dissolved and particulate phases), but also to characterise the nature, the composition and the contamination of the organic particles known to be assimilated by bivalves.

On the French Atlantic coasts, the Arcachon Bay is an important production area of the oyster species *C. gigas* and is also the location of many other economic and recreational activities such as fishing and water sports. In addition, agricultural production (mainly corn) is active on its watershed, and industrial activities are developing in the vicinity of the bay for an ever-growing population. For a long time, this semi-enclosed and anthropized coastal area has been known to be contaminated by organic pollutants such as PAHs, PCBs and various pesticides, and also inorganic contaminants such as tributyltin (TBT) and copper (Cu) among others (Baumard et al., 1998; Claisse and Alzieu, 1993; Devier et al., 2005; Gamain et al., 2018). More specifically, since the early 2000s, the ROCCH network evidenced a strong increase in oyster Cu concentrations, especially at stations located in the innermost part of the bay (Ifremer LERAR, 2012). A previous increase (although weaker) was already observed between 1980 and 1993, probably reflecting the novel intensive use of Cu in antifouling paints, replacing very toxic and hence banned TBT (Claisse and Alzieu, 1993). If the novel increase in Cu concentrations of Arcachon oysters was still the result of boating, this would be either due to an increase in the number of boats in the bay, and/or a change in the composition of anti-fouling paints. However, the number of boats has not shown any significant change since the 1990s, making the first hypothesis uncertain (Ifremer LERAR, 2012). Furthermore, the ROCCH network demonstrated that oysters from stations located in the outermost part of the bay (also located close to parked ships) did not show peculiar trends of Cu increase over time, making the second hypothesis also uncertain. Copper isotopes have recently been used as a forensic tool to infer the anthropogenic origin of bioaccumulated Cu in oysters of the bay, however, the identification of its source(s) has been difficult (Araújo et al., 2021).

From an ecological point of view, the Arcachon Bay is characterized by the presence of large dwarfgrass (*Zostera noltei*) beds and smallest eelgrass (*Z. marina*) beds (Cognat et al., 2018). These species enhance diversity and abundance of aquatic fauna by acting as a bottom stabilizer, dampening hydrodynamic constraints on the sediment, promoting sediment deposition and reducing the rate of resuspension of fine particles, thus promoting water transparency and primary production (Ganthy et al., 2015). However, since the early 2000s, 33% of *Z. noltei* and by 74% of *Z. marina* have disappeared from the inner part of the bay (Auby et al., 2011; Cognat et al., 2018; Plus et al., 2010). Many causes for this decrease have been hypothesised, such as grazing by wintering geese (*Branta bernicla bernicla*) (Auby et al., 2011), heat waves and pollutants through a microcosm experimental study and field sampling (Gamain et al., 2018). Concurrent to this *Zostera* spp. decrease, an increase in the concentration of suspended matter (fine sediment particles) has been observed since the early 2000s (ARCHYD hydrological network; Ifremer LERAR, 2021), especially in the inner part of the bay. These sediments likely act as a sink for many trace elements that have high affinity with organic matter or fine particles, especially in coastal areas (Sharifuzzaman et al., 2016).

In this general context, we conducted a one-year multi-parameter and multi-compartment monitoring field study to challenge, at least in part, some aspects of the environmental context that may explain the increasing Cu concentrations observed for many years in oysters from the inner part of Arcachon Bay. The different biogeochemical compartments of the oysters' environment (i.e. sediments, seawater including particles, and transplanted oysters) were sampled regularly for one year at two contrasted sites of the bay, and were characterised according to their concentrations of Cu and two other essential metals, zinc (Zn) and nickel (Ni), and two non-essential elements, cadmium (Cd) and lead (Pb). For some compartments, two different strategies of collection or analysis were used (e.g. for dissolved metal concentrations, punctual sampling and filtration of seawater *vs.* passive sampling) to compare the observations. Several accompanying parameters were acquired simultaneously with metal concentrations such as physico-chemical parameters in seawater, qualitative and quantitative analysis of particles (including phytoplankton) and carbon (C) and nitrogen (N) stable isotope compositions of particles and oysters. The latter compositions are recognised as valuable tracers to i) characterise the composition and the quality of particulate organic matter (POM; Liénart et al., 2017; Savoye et al., 2012), which constitute the trophic resources available to oysters (Lebreton et al., 2011) and ii) to understand the trophic resources actually assimilated by these primary consumers (Dubois et al., 2014; Kharlamenko et al., 2001).

Our objectives were: i) to assess the temporal and spatial trends of metal contamination in the surrounding environment (seawater, sediment, plankton) and the respective bioavailable fractions (including trophic sources) for oysters at the two contrasted sites of the bay; ii) to study the bioaccumulation of metals in transplanted oysters of known origin during one full year at the two contrasted sites; and iii) to identify, as thoroughly as possible, the potential links between compartments in terms of metal transfers, especially from seawater and/or particles to oysters. We hypothesised that contrasting environmental conditions in the inner *versus* outer parts of the bay likely influence i) metal occurrence and bioavailability, and/or ii) growth rate and iii) metal burdens of oysters. More specifically, we expected that the load and the nature of particles (organic or inorganic, and (taxonomical) composition of organic particles) – which are documented to be more abundant in the inner part of the bay due the seagrass biomass decrease observed for almost two decades – may impact the growth of filter-feeding organisms such as oysters, and/or the metal concentrations observed in oysters and in other biogeochemical compartments over time (e.g. through a metal release by resuspended fine sediment particles).

2. Materials and Methods

2.1. Study sites in Arcachon Bay

Arcachon Bay (44°40’N, 1°10’W) is a semi-enclosed lagoon located in the southeast part of the Bay of Biscay, NE Atlantic. Its surface is about 174 km2 and about 65% of this surface emerges at low tide (Plus et al., 2009). We monitored trace metals in different biogeochemical compartments at two contrasting sites of the bay: i) Comprian, in the inner part of the bay, under the influence of the Leyre river and ii) Grand Banc (GB), in the outer part of the bay, close to the ocean channel and more under oceanic influence (Fig.1). However, there were no facilities for the deployment of passive samplers at the GB site. Passive samplers were therefore deployed at Courbey, which is very close to GB (Fig. 1).

2.2. Sampling design and sample preparation before trace metal analyses

2.2.1 Sediments

Superficial fine sediments were collected at low tide, close to where water was sampled and oysters were deployed at the two sites, Comprian and GB, close to the beginning of the study on 25 April 2017, and at the end of the one-year field study on 22 March 2018. The top 1 cm of sediment was sampled from an approximative 1 m^2 area using a stainless-steel spatula. This composite sample was then split into several subsamples for the different analyses. Two subsamples were frozen at -20°C until further processing before metal analysis and C and N elemental and isotopic compositions. A third subsample was kept at 4°C pending granulometry analysis.

Sediment samples for metal analyses were freeze-dried, homogenized with an agate mortar and pestle and sieved at 2 mm. Approximately 200 mg were weighted in Teflon® bombs and mineralised on a heating block using a multiple-step acid-concentrated procedure with ultrapure HNO_3, HCl and HF as described in detail in Araújo et al. (2019).

2.2.2 Seawater

Seawater samples dedicated to trace metal analyses were collected once per month at the two sites, Comprian and GB, 1 m below the water surface, from May 2017 to March 2018. The collection was carried out at about 2 m from the boat using a wooden pole, to which an acid-cleaned polyethylene bottle was attached. The bottles filled with seawater (~500 mL) were then kept at 4°C and in the dark until filtration within 24 hours in ultra-clean laboratory conditions, under a laminar flow hood. The filtration of a known volume of seawater was then made on 0.45 µm mesh acid-cleaned and pre-weighed polycarbonate filters (Nucleopore®) and at low pressure (<0.5 bar) to prevent any bursting of phytoplanktonic cells. Filtered seawater was finally transferred into acid-cleaned polyethylene bottles and acidified with ultrapure HNO_3 (0.1%) for further dissolved trace metal analyses. Bottles were hermetically sealed, double-bagged and stored in the dark at 4°C pending analytical processing. Polycarbonate filters with particulate material >0.45µm were rinsed with ultrapure water to remove sea salt (very briefly to avoid cell bursting) and kept frozen at -20°C.

Prior to the determination of dissolved metal concentrations, a pre-concentration step using a liquid/liquid extraction was employed on acidified filtrates according to an adapted protocol from Danielsson et al. (1982) and described in detail by Chiffoleau et al. (2002). This protocol first consists of dithiocarbamate chelate formation with ammonium 1-pyrrolidinedithiocarbamate/diethylammonium diethyldithiocarbamate (APDC-DDDC, sodium salts, >97%) in 100 g (~100 mL) of the water phase buffered to pH = 5. This is followed by an extraction into an immiscible organic solution (1,1,2-trichloro-1,2,2-trifluoroethane, >99%) and a back extraction using diluted nitric acid (ultrapure, 1:4 v/v), repeated twice. One mL of the extract is finally diluted to 5 mL with ultra-pure water before analysis.

2.2.3 Diffusive Gradient in Thin films (DGTs)

In addition to seawater sampling, DGT-passive samplers were deployed monthly from May 2017 to March 2018 at the two sites, Comprian and Courbey. This technique is based on the diffusion of metals through a diffusive layer towards a binding phase where they concentrate (Davison and Zhang, 1994; Zhang and Davison, 1995). DGTs thus concentrate metal cations in the form of hydrated ions, mineral complexes and small organic complexes in the water column (i.e. “labile” soluble forms of metals). All of the equipment used for DGT deployments (DGT supports and protection boxes) as well as the equipment for DGT processing and analysis (e.g. elution tubes) were cleaned with 10% ultrapure HNO_3 for several days, rinsed with ultrapure water and stored in double plastic bags. The DGT devices (polyacrylamide diffusive gel/Chelex100 resin, DGT Research©) were immersed in triplicate (i.e. three DGTs fixed on a polypropylene support) for 72h–96h (exact immersion time recorded each time). After recovery, DGTs were rinsed with ultrapure water, placed in boxes dedicated to their storage with their support and kept at 4°C until resin recovery in clean laboratory conditions. DGT opening, resin recovery and immersion in an ultrapure HNO_3 1M solution (for elution) was finally performed under a laminar flow hood, and eluates were kept at 4°C until analysis. Resin blanks were done systematically on each new batch of DGTs.

2.2.4 Particles

Two size fractions of particles (suspended particulate matter, SPM) were considered for trace metal analyses: i) the fraction >0.45 µm, collected on acid-cleaned polycarbonate filters used for seawater filtration (see above) and thus sampled monthly from May 2017 to March 2018 and ii) the fraction 6–100 µm, considered to be the major fraction that can be ingested by oysters (Cognie et al., 2001; Dupuy et al., 2000, 1999) and likely the largest contaminate of oysters through food. The 6–100 µm fraction was sampled two and six weeks before each oyster sampling (after 3, 6 and 12 months of transplantation, see below), thus corresponding to six sampling dates over the one-year study. This fraction was collected directly in the field by successive sampling (with a clean bucket made entirely of plastic) and sieving of tens of litres of seawater on clean nylon sieves of 100 and 6 µm mesh. This allowed concentrating this fraction to get a maximum of material for further analyses, while removing a maximum of seawater. These concentrated samples of SPM 6–100 µm (with few remaining mL of seawater) were finally split into several subsamples for the different analyses. Two subsamples were frozen at -20°C until further processing, with distinct aliquots designated for metal analysis and for C and N elemental and isotopic analyses.

In addition, seawater samples collected using Niskin© bottles were processed through in the field on a clean 100 µm nylon mesh and stored in plastic bottles, to further assess the concentrations of SPM <100 µm and the percentage (%) of organic matter (OM) in particles, and to characterize the phytoplankton composition of particles. This <100 µm fraction is used hereafter as a proxy of the 6–100 µm fraction for these accompanying parameters. Approximately 500 mL of 100 µm-sieved seawater were finally taken and stored in a dark glass bottle for chlorophyll *a* and pheopigment analyses.

In clean laboratory conditions, the polycarbonate filters with particulate material >0.45µm (used for seawater filtration) were dried in an oven for 24h at 60°C, weighted again (to obtain the dry mass on filters and hence concentrations of SPM >0.45 µm), and put in Teflon® bombs. We then followed the same mineralisation procedure as the sediments. Alternatively, the frozen subsamples of concentrated SPM 6–100 µm (with the few remaining mL of seawater) dedicated to trace metal analyses were gently thawed, agitated, and sub-divided into two aliquots of a known volume. The first aliquot (aliquot a) was used to estimate the concentration of concentrated SPM 6–100 µm "without salt" and the second aliquot (aliquot b) was actually used for metal analyses. Aliquot (a) was filtered on 5 µm mesh pre-weighed polycarbonate filters (Nucleopore®). The filters were then briefly rinsed with ultrapure water to remove sea salt, dried in an oven for 24h at 60°C and weighted again. Aliquot (b) was transferred into Teflon® bombs and heated at 90°C on a heating block until the remaining few mL of seawater had evaporated. The dried pellets obtained therefore included a certain mass of "salt-free" SPM 6–100 µm known from aliquot (a), the metals associated with these particles, the salt in the residual seawater, and the metals associated with these few mL of residual seawater (the latter metals being then considered negligible based on the concentrations measured in filtered seawater samples). These dried pellets were finally mineralised for 4h at 90°C with a mixture of ultrapure HNO_3 (3,5 mL) and HCl (5 mL), and the digests were diluted to 50 mL with ultrapure water (and further diluted if necessary, to diminish sea salt concentrations).

2.2.5 Oysters

Approximately 1000 individuals of 18-month-old oysters born and raised since mid-December 2016 on the Arguin Banc site (outside the Arcachon Bay, fully under oceanic influence) were transplanted on 30 March 2017 (beginning of the study – T0), to the two study sites, Comprian and GB, alongside local existing shell farming batches. Then, at each site, 30 individuals were collected for biometric measurements and 15–50 individuals (depending on size) were collected for chemical analyses, 3 months (26 June 2017 – T1), 6 months (25 September 2017 – T2) and one year (22 March 2018 – T3) after transplantation. After collection, transplanted oysters were cleaned of epibiota and depurated for 24 h in a polyethylene aquarium containing decanted water from the respective sampling site to eliminate faeces, pseudo faeces and potential inorganic material in their digestive tracts. For the 30 individuals collected for biometric measurements, total length, total body weight, shell weight and the weight of soft tissues (i.e. flesh weight after drying) were measured individually, and a body condition index was calculated as follows (Lawrence and Scott, 1982): (flesh dry weight / (total wet body weight – shell weight)) * 1000. For the 15–50 individuals for chemical analyses, only the individual total length was measured to ensure similarity with the oysters used for all biometric measurements. The soft tissues of these 15–50 individuals were finally pooled and homogenized with a stainless steel-bladed blender, frozen again (-20°C) and finally freeze-dried pending analyses.

For Ni and Pb determinations, about 200 mg of bivalve samples were placed in Teflon® bombs, mineralised with a mixture of ultrapure HNO_3 acid (3 mL) and ultrapure water (3 mL) using a microwave (ETHOS-UP, Milestone) and the digests were diluted to 50 mL with ultrapure water. For other metals (Cu, Zn and Cd), approximately 100 mg of dried and homogenised samples (three replicates per sample) were digested with HNO_3 acid (3 mL) in polypropylene tubes, heated on a heating block for 3h at 100˚C and 15 mL of ultrapure water were finally added before analyses.

2.3. Trace metal analyses

Concentrations of all metals (Ni, Cu, Zn, Cd and Pb) in the different matrices were determined by Q-ICP-MS (iCAP Qc, KED mode using He gas, Thermo Fisher Scientific). The exceptions were Cu, Zn and Cd concentrations in oysters that were analysed by ICP-OES (700 Series, Agilent Technologies). In sediments, aluminium (Al) concentrations were also determined by Q-ICP-MS for further normalisation of data in this matrix.

The quality assurance of all metal analyses relied on blank controls and on the accuracy and reproducibility of certified reference materials (CRMs), processed jointly with sample batches and measured in each analytical run. Blank values were systematically below the detection limits and CRM values concurred with certified concentrations. Results of the CRM analyses are specified in Table A1, together with the limits of quantification for each metal and each matrix. For DGTs, the concentrations of metals in "labile" soluble forms in seawater (as retained by DGTs) were calculated according to the first law of Fick, taking into account the immersion time and water temperature (Davison and Zhang, 1994).

2.4. Analyses of accompanying physico-biochemical parameters

2.4.1 Sediment granulometry

Sediment was sieved at 2mm and then analysed for granulometry using a Malvern Mastersizer hydro 2000G.

2.4.2 Physico-chemical parameters in seawater

Temperature, salinity and turbidity were measured using a YSI multi-parameter probe (model YSI 6600 V2) positioned 1 m below the water surface, at each site and each time the particle fraction 6–100 µm was sampled (i.e. two and six weeks before each oyster sampling), thus corresponding to six sampling dates over the one-year field study.

2.4.3 Particle concentrations and biological composition of SPM <100 µm

Back to the laboratory, additional seawater samples dedicated to the assessment of particle concentrations (SPM <100 µm) were filtered on pre-weighted Whatman GF/F filters (0.7 µm mesh size) and dried at 70°C to a constant weight. To estimate the inorganic-organic partition of these SPM <100 µm, the loss on ignition was determined by combustion at 500°C for one hour. Concomitantly, the concentrations of SPM >0.45 µm were assessed after freeze-drying the pre-weighted polycarbonate filters used for seawater filtration and trace metal analysis of SPM >0.45 µm (see above).

For quantitative phytoplankton analyses (taxonomical composition), dedicated seawater samples were fixed in the field with Lugol's solution and then counted according to the Utermöhl method (Utermöhl, 1958). Organisms were identified to the lowest possible taxonomical level.

2.4.4 Chlorophyll a and phaeopigments on SPM 5-100 µm

Dedicated subsamples of 100 µm-seaved seawater were filtered again using polycarbonate filters of porosity 5 µm (porosity closest to 6 µm in the absence of 6 µm filters). After filtration, the filters were stored in extraction tubes at -80°C until analysis. Then, filters were extracted using 90% acetone and the fluorescence was measured before and after acidification using a Turner Designs TD-700 fluorimeter for chlorophyll *a* and phaeopigments quantification (Yentsch and Menzel, 1963).

2.4.5 Carbon (C) and nitrogen (N) compositions of sediments, SPM (POM) 6-100 µm and oysters

Dedicated samples of each matrix were freeze-dried and powdered using a pestle and a mortar. Samples were weighed into tin cups for all N elemental and isotopic compositions and for C elemental and isotopic compositions of oysters. For C elemental and isotopic compositions of sediments and SPM, samples were weighed into silver cups and decarbonated using 1.2 N HCl (Kennedy et al., 2005). Cup samples were then analysed for C or N elemental and isotopic compositions using an elemental analyser (Flash 2000, ThermoFisher Scientific) coupled with an isotope ratio mass spectrometer (IRMS, Isoprime, GV Instruments). IRMS daily drift was monitored using home-made standards and, if necessary, data were corrected. Elemental and isotopic compositions were calibrated against homemade standards and CRMs.

2.5. Data treatment and statistical analyses

Total metal concentrations in the “bulk” seawater were calculated summing metal masses from dissolved and particulate phases as follows: dissolved metal concentration in 1 L + metal concentration in particles * concentrations of particles in 1 L. From the same data (dissolved metal concentrations in seawater (filtered samples, <0.45 µm), metal concentrations in particles (SPM >0.45 µm) and concentrations of SPM >0.45 µm), spatio-temporal variations in the percentages of particulate metals could also be determined.

In oysters, to avoid a bias of metal bioaccumulation assessment linked to potential growth differences when only considering metal concentrations, the metal body burden of an average individual (corresponding also to the metal quantity (in µg) in an average individual) was calculated at each site and for each sampling period according to the formula: metal concentration in oysters (considered in µg g^{-1} dry weight (dw) instead of mg kg^{-1} dw) * mean individual flesh (i.e. soft tissues) dry weight (in g). This mean individual flesh dry weight corresponded to those measured on the 30 oysters used for biometric measurements at each site and for each sampling period (see section 2.2.5 above).

Bioaccumulation factors (BAFs) were also calculated for oysters at each site and for each oyster sampling period (T1, T2, T3), according to the formula of Griboff et al. (2018): BAF = C_{oys}/C_{sw}, whereby C_{oys} is the metal concentration in oysters at steady state (dry weight basis) and C_{sw} is theoretically the metal concentration in seawater. Here, for metal concentrations in seawater, the total concentrations in the water column were used (including both dissolved and particulate phases of metals), and all the seawater data acquired before each oyster sampling date were used. These calculated "field-based BAFs" could thus integrate all the possible routes of exposure to metals for the oysters (i.e. dissolved and particulate/trophic routes).

Due to the relatively low number of samples in each data series submitted to statistical analyses, only non-parametric tests were used to assess spatial differences, or correlations between parameters. Thus, Mann-Whitney-Wilcoxon (MWW) tests for paired samples were used to test the differences between the two sites for any data constituted by temporal series, to account for the sampling date and assess if one site or the other always had higher or lower metal concentrations values than the other site over time. Spearman correlation coefficient tests were used to evaluate relationships between metal concentrations in SPM 6–100 µm and SPM (biological) characteristics (chlorophyll *a*, phaeopigments, concentrations of particles, % of OM in particles, δ^{13}C and δ^{15}N values, densities of total microphytoplankton, dinoflagellates, ciliates, and number of phytoplankton taxa). As mentioned above, the characteristics of SPM <100 µm were used when those for 6–100 µm fraction were not available (i.e. concentrations of SPM, % of OM in SPM, taxonomical composition and phytoplankton densities).

3. Results

3.1. Sediments

3.1.1 Granulometry and C and N isotope compositions

Sediment particles exhibited a bimodal distribution with a first mode at 8 µm for both sites and both dates, and other modes at 86 µm (Comprian, April 2017), 212 µm (GB, April 2017) and 177 µm (Comprian and GB, March 2018). The percentage of silt and clays was very variable (69% at Comprian in April 2017, 35% at GB in April 2017, 47% at Comprian in March 2018 and 61% at GB in March 2018).

δ^{13}C and δ^{15}N values were very similar among sites and at both dates, although slightly higher δ^{13}C values were measured in sediments from Comprian (Table 1).

3.1.2 Metal concentrations

When considering raw data, sediment concentrations of all metals were higher at Comprian than at GB in April 2017 (Table 1). However, there was no more differences between sites when considering normalised concentrations to Al 5%. In addition, for all metals, there was no difference between sites in April 2018, whether considering raw or normalised concentrations (Table 1).

3.2. Seawater

3.2.1 Temperature, salinity and turbidity

Seawater temperature had a marked seasonal cycle at both sites (Fig. A1). There was no significant difference between sites, whether considering low or high tide (MWW tests for paired samples, all $p > 0.05$). Salinity was significantly lower at Comprian at both tides (MWW tests for paired samples, $p = 0.031$ in both cases), with particularly low values at low tide in winter 2018 (Fig. A1). This was fully consistent with the daily Leyre river flows available for the study period (Fig. A2), which varied from approximatively 30 to 60 $m^3 s^{-1}$ throughout the winter (January–March) 2018 whereas they were less than 15 $m^3 s^{-1}$ previously (i.e. since the beginning of the study period, end of March/beginning of April 2017). Finally, turbidity was significantly higher at Comprian compared to GB at low tide only (MWW test for paired samples, $p = 0.031$). Moreover, there was no real seasonal cycle for this parameter, except for Comprian in winter 2018 (particularly high value at this site in March 2018; Fig. A1).

3.2.2 Dissolved metal concentrations in filtered samples (<0.45 µm) and in DGTs (passive sampling)

Average dissolved metal concentrations measured in filtered samples over the one-year field study (collected once a month at low tide only) are indicated in Table 2. These concentrations showed seasonal variations for Ni, Cu, Cd and to a lesser extent Zn. Concentrations of Pb were less variable (Fig. 2, left panel). A common downward trend was observed between May (spring) and November 2017 (late autumn) for Ni, Cu and Cd concentrations, while Zn concentrations were more stable during this period. With the exception of Pb, we observed an increase in all dissolved metal concentrations in winter 2018 at both sites (in January and/or March 2018; Fig. 2, left panel). There was a significant difference between sites for all metals except Pb, with consistently higher Ni, Cu, Zn and Cd concentrations measured at Comprian (MWW tests for paired samples, all $p < 0.05$).

Average dissolved metal concentrations measured in DGTs (monthly few days immersions) are also given in Table 2. These concentrations were more variable over time than punctual (filtered) measurements of dissolved concentrations (Fig. 2, right panel). No clear common trend appeared, except slightly higher concentrations measured in October 2017 for Ni, Cu, Cd and Pb (although at Comprian only for Cd), and in November 2017 for Zn. Otherwise, like for filtered samples, an increase in metal concentrations was observed in March 2018 (i.e. late winter 2018; Fig. 2, right panel). There was a significant difference between sites for Cd only, with higher Cd concentrations measured at Comprian compared to GB over time (MWW tests for paired samples, $p = 0.009$ for Cd and $p > 0.05$ for other metals).

3.3. Particles

3.3.1 Concentrations of SPM and inorganic-organic partition

There were no seasonal variations of SPM concentrations over time at both sites, whether considering SPM <100 µm (sieved samples in the field, at both tides) or SPM >0.45 µm (laboratory filtered samples, low tide only), except for Comprian in winter 2018 (particularly high value at this site in March 2018; Fig. 3). There was a significant difference between sites at low tide only, with higher concentrations of SPM <100 µm measured at Comprian (MWW tests for paired samples, $p = 0.031$ at low tide, $p > 0.05$ at high tide), as well as significantly higher concentrations of SPM >0.45 µm measured at Comprian compared to GB (MWW test for paired samples, $p = 0.004$). Among <100 µm particles, the % of OM was globally low, being of 16.9 ± 5.0 % on average (considering both tides and sites).

3.3.2 Chlorophyll a and phaeopigments in SPM (POM) 5–100 µm

There were no seasonal variations for these parameters, except for Comprian at low tide. Particularly high concentrations of chlorophyll *a* and phaeopigments were indeed observed at low tide for this site in summer 2017 (from June to September 2017), and also in winter (March) 2018 (Fig. A3).

3.3.3 C and N isotope compositions of SPM (POM) 6–100 µm

δ^{13}C values of 6-100 µm particles ranged between -22.7‰ and -19.2‰ (-20.7‰ ± 1.0‰), with lower values usually in the outermost water mass (GB at high tide, Fig. A3). No clear temporal pattern was noticeable. δ^{15}N values ranged between 5.0‰ and 7.8‰ (6.0‰ ± 0.7‰), with lower values usually in the innermost water mass (Comprian at low tide, Fig. A3). Like for δ^{13}C values, no clear temporal pattern was noticeable for δ^{15}N values. C:N ratios ranged between 8.1 and 12.8 mol mol^{-1} (10.2 ± 1.2 mol mol^{-1}), with higher values usually in the innermost water mass (Comprian at low tide). C:N ratios were roughly lower and higher in spring 2017 and winter 2018, respectively (Fig. A3).

Values of POC/chlorophyll *a* ratio (not shown) were usually low and characteristic of phytoplankton-dominated POM (<200 µg µg^{-1}), at all dates and in all water masses except at low tide at Comprian in winter 2018 (>300 µg µg^{-1}).

3.3.4 Biological composition of SPM (POM) 6–100 µm

More than 100 species or groups of phytoplankton cells were identified in the samples. Several species or groups were present all year long at both sites (e.g. Cryptophyta, ciliates, dinoflagellates belonging to the order of Gymnodiniales and the diatoms *Asterionellopsis glacialis*, *Nitzchia longissima*, *Chaetoceros* sp., Naviculaceae and *Paralia sulcata*). Despite this high diversity, only a few phytoplanktonic species or groups contributed to a major part of the densities of the total microphytoplankton throughout the year, with four species or groups representing about 70% of the cumulative enumeration: Cryptophyta (33.4%), *Asterionellopsis glacialis* (22.7%), Naviculaceae (7.2%) and ciliates (5.5%). Globally, microphytoplankton densities were maximal during late spring 2017 and early spring 2018, especially due to diatoms' temporal evolution (Figs. 4 and A4). On the other hand, dinoflagellates and ciliates were more abundant during summer 2017 (Figs. 4 and A4). Considering both tides, mean densities of dinoflagellates were significantly higher at GB than at Comprian (MWW tests for paired samples, p = 0.037), while ciliates were more abundant at Comprian (MWW tests for paired samples, p = 0.025, Figs. 4 and A4). There were no significant differences between sites for mean total microphytoplankton and diatom densities, whether considering tides together or separately (MWW tests for paired samples, all p > 0.05).

3.3.5 Metal concentrations in SPM 6–100 µm and SPM >0.45 µm

Average metal concentrations measured in SPM 6–100 µm (collected at both tides two and six weeks before oyster sampling) or in SPM >0.45 µm (collected monthly at low tide only) are summarised in Table 3. There were no major seasonal variations of particulate metal concentrations (Fig. 5), whether considering SPM 6–100 µm (sieved samples on the field) or SPM >0.45 µm (laboratory filtered samples). The noticeable exceptions were: i) in SPM 6–100 µm, higher concentrations measured in spring (May) 2017 for all metals and at both sites, and also in summer (August) 2017 for Cd and Pb at GB (Fig. 5); ii) in SPM >0.45 µm, slightly higher concentrations measured for Ni, Zn and to a lesser extent Cu in June 2017 at GB, and also higher Cu and Cd concentrations in autumn 2017 at GB (between September and December 2017; Fig. 5). There were no major spatial variations when considering SPM 6–100 µm. The only significant difference between sites was for Cu at low tide (with Comprian < GB; MWW tests for paired samples, p = 0.031). Conversely, when considering SPM >0.45 µm, significant differences between sites were observed for Cu and Cd (with Comprian < GB; MWW tests for paired samples, p = 0.002 for both Cu and Cd), and also Pb (Comprian > GB, p = 0.010).

3.3.6 *Correlations between metal concentrations in SPM 6–100 µm and SPM characteristics*

Both tides and sites considered, all metal concentrations in SPM 6–100 µm were significantly and negatively correlated with the concentrations in chlorophyll *a* (Spearman correlation coefficient tests, all $p < 0.05$, Table 4). Concentrations of all metals except Cd were also significantly and negatively correlated with the concentrations in phaeopigments and with the concentrations of particles (i.e. concentrations of SPM <100 µm). Moreover, concentrations of all metals except Cd were significantly but positively correlated with the % of OM in particles, but negatively correlated with the concentration of particles (SPM) <100 µm (Table 4). No significant correlations between metal concentrations in SPM and the other parameters studied were observed (Spearman correlation coefficient tests, all $p \geq 0.05$ with $\delta^{13}C$ and $\delta^{15}N$ values, densities of total microphytoplankton, dinoflagellates, ciliates and number of phytoplankton taxa). The only exception was a slightly significant and positive correlation observed between Cu concentrations and the number of taxa (Spearman correlation coefficient test, $p = 0.040$, Table 4). Overall, Cu was the element for which the significant correlation coefficients with accompanying (biological) parameters were the highest. Finally, metal concentrations in SPM 6–100 µm were all significantly and positively correlated with each other (Table 4).

3.3.7 *Metal partition and total metal concentrations in the water column (dissolved + particulate)*

Percentages of particulate metals varied greatly among metals and over the study period, ranging from 16% to 89% for Ni considering both sites, from 39% to 88% for Cu, from 39% to 93% for Zn, from 16% to 69% for Cd, and from 89% to 100% for Pb. Seasonal variations occurred at both sites and for all metals except Pb (Fig. A5). At Comprian, higher percentages of particulate metals were observed in summer (August–September) 2017, while the opposite was observed at GB (i.e. lower % of particulate metals observed at this period). However, at both sites, an increase of these percentages occurred in late autumn and in winter (January–March) 2018 (Fig. A5). Over the study period, there was a significant difference between sites for Ni, Cd and Pb, with higher percentages of particulate metals observed at Comprian compared to GB (MWW tests for paired samples, $p < 0.05$ for these elements). The difference was less marked for Zn ($p = 0.049$), and no significant difference between sites was observed for Cu ($p = 0.695$).

Considering both the dissolved and particulate fractions of metals, total metal concentrations in the water column could be calculated (Fig. 6). While no seasonal variations occurred at GB, higher total metal concentrations were observed at Comprian in summer (August and September) 2017 and in winter (March) 2018. These corresponded to periods where higher concentrations of SPM >0.45 µm (Fig. 3) and higher % of particulate metals in the water column (Fig. A5) were also observed at this site. A significant difference between sites was observed for all metals over the study period, with higher total concentrations in the water column observed at Comprian than at GB (MWW tests for paired samples, $p < 0.05$ for all elements).

3.4. Oysters

3.4.1 Biometry, growth and condition index

At both sites, oyster length, total wet weight and shell weight increased over the study period (Table 5). Conversely, flesh dry weight showed a rapid increase between March and June 2017, then it decreased between June and September 2017 as a result of massive spawning in early July at both sites. Then, between September 2017 and March 2018, the flesh dry weight of oysters increased much more at GB than at Comprian, with a final Comprian/GB ratio of 0.4 for this parameter (Table 5). Furthermore, while the condition index of GB oysters at T3 (94.8) was similar to those measured at T1 (96.1), the condition index of Comprian oysters was lower by half at T3 (45.7) compared to T1 (87.9). Regardless of the dates, GB oysters had higher values than Comprian oysters for all biometric parameters (Table 5).

3.4.2 C and N isotope compositions

In oysters, C and N isotopic and elemental ratios exhibited low variability over time and between sites ($\delta^{13}C$ = -19.8 ± 0.5‰; $\delta^{15}N$ = 7.3 ± 0.2‰; C:N ratio = 5.6 ± 0.5 mol mol^{-1}; Table 4). However, a decrease in $\delta^{13}C$ values and C:N ratios appeared between T0 and T1 at both sites. Then, the $\delta^{13}C$ values increased over time (between T1 and T3) to reach initial values at both sites. The C:N ratio remained stable and low at Comprian, whereas it increased to reach initial (T0) values at GB (Table 5).

3.4.3 Metal concentrations and metal quantities per mean individual (body burdens)

Considering metal concentrations in soft tissues, after a slight decrease between T0 and T1 for most metals and especially at GB, an increase in all metal concentrations was observed over time at both sites (Fig. 7-A). This increase was particularly high between T1 and T2 and at Comprian. Indeed, no noticeable increase in metal concentrations was observed between T2 and T3 at GB, except for Pb (Fig. 7-A), and the increase was moderate at Comprian during this period. Overall, higher metal concentrations in oysters were observed at Comprian than at GB over the study period, for all elements except Pb (similar concentrations between sites for Pb; Table 5, Fig. 7-A).

When considering bioaccumulated metal quantities per average dried individual (i.e. total body burdens), trends differed considerably (Fig. 7-B). Differences between sites were less marked over the study period, and oysters from GB even reached higher quantities of all metals except Cu at T3. Indeed, Cu was the only element for which GB oysters had higher metal quantities than Comprian oysters at T1 and T2. At T3, oysters finally reached similar Cu body burdens at both sites (Table 4, Fig. 7-B).

3.5. Bioaccumulation factors (BAFs) in oysters

BAFs in oysters (calculated from total metal concentrations in the water column, including both dissolved and particulate phases) were the highest for Zn, then Cu, Cd, Ni and/or Pb, depending on the site for the last two elements (Table 6). At GB, BAFs generally increased over time (between T1 and T3) for all metals, whereas at Comprian, the highest BAFs were calculated at T2 for all metals except Cd (i.e. similar BAFs at T2 et T3 for Cd). Interestingly, despite some exceptions at T1, BAFs were consistently lower at Comprian than at GB over the study period, except for Cu (inverse trend, Comprian > GB).

4. Discussion

Arcachon Bay is an emblematic European oyster production area of high ecological and economic importance, providing provisioning, regulating, supporting and cultural services (Newton et al., 2014). Therefore, studying and understanding metal contamination transfers from the environment to biota in this semi-enclosed maricultured ecosystem is crucial. In the present study, the multi-compartmental sampling we performed, combined with multi-elemental analysis and analyses of biometric parameters and physico-chemical parameters in sediment, seawater, particles (including phytoplankton) and oysters, allowed us to i) characterise the spatio-temporal dynamic of the metal contamination for these different matrices in Arcachon Bay over a year and ii) highlight the lower growth rate of oysters in the inner part of the bay (Comprian site), concomitantly observed with higher concentrations of particles and to higher metal concentrations measured in oysters from this site, but also in the water column (i.e. total concentrations including both dissolved and particulate phases). Finally, our results have some implications for the use of bivalves as bioindicators of chemical contamination in mussel-watch programs.

4.1. General spatio-temporal trends in the contamination of oysters' environment

The significant and positive correlations between individual metal concentrations in SPM 6–100 µm attested to the polymetallic character of the contamination that is generally documented in coastal sites (Lu et al., 2018), as fine particles have a great capacity to adsorb and transport metals in coastal environments.

Broadly, in terms of spatial variations, two patterns emerged: i) significant differences between sites were recorded for all metals except Pb in filtered seawater, while for Cd only in DGTs, and for all metals when considering total concentrations in the water column (dissolved plus particulate phases), with the consistent common pattern of higher concentrations at Comprian compared to GB; ii) metal concentrations were more homogenous between sites for sediment and particles (whether considering SPM 6–100 µm or SPM >0.45 µm), and/or site differences were metal-specific for these matrices (especially for SPM >0.45 µm, collected at low tide only). In terms of temporal variations, there was i) a common trend to lower dissolved metal concentrations during summer months, and higher concentrations observed in late winter 2018 in seawater (both filtered and DGT samples) for most metals, while ii) no clear seasonal variations were observed for metal concentrations in particles (whether considering SPM 6–100 µm or SPM >0.45 µm). Moreover, at the Comprian site in particular, higher total metal concentrations in the water column were observed (dissolved plus particulate phases) concomitantly with higher concentrations in particles and higher percentages of particulate metals in the water column (i.e. in summer 2017 and winter 2018). The accompanying parameters measured simultaneously to metal concentrations in the different matrices help to explain some of the metal variations observed, as explained hereafter.

4.2. Seawater variations

The salinity variations typically showed the impact of the Leyre flow rate over the year (i.e. higher flow rate and consequently lower salinities in January and March 2018, especially at low tide for both sites; Fig. A1). These variations also evidenced a higher continental influence at the Comprian site, located in the innermost part of the bay. This part of the bay is submitted to river inputs and underground freshwater discharges, with a major part coming from the Leyre river and the remaining being provided by secondary streams (Deborde et al., 2008; Rimmelin et al., 1998). This continental (river) influence may explain, at least in part, the higher metal concentrations measured in seawater from Comprian, especially for the metals that present a non-negligeable dissolved phase (i.e. all metals except Pb), because riverine inputs remain a major source of metal release into coastal aquatic ecosystems (Mason, 2013). Indeed, recent Cu isotope compositions revealed that closer to the coast, a major exposure to anthropogenic Cu emissions occurs (Araújo et al., 2021). Finally, the probable influence of the Leyre river likely explains the lower metal concentrations measured in seawater during summer months, i.e. time of year when the Leyre river flow is the lowest (Fig. A2), while higher metal concentrations are observed in seawater in late winter, i.e. when the Leyre river flow is the highest (Plus et al., 2009 and Fig. A2). This finding is also in agreement with previous results concerning organic compounds like herbicides, for which a peak of contamination was observed in winter in Arcachon Bay (Fauvelle et al., 2018). This phenomenon has been explained by a temporary contaminant storage in superficial groundwater prior to discharge in rivers when groundwater table level is high enough to be drained by the neighboring river in winter.

4.3. Particle and sediment variations

The turbidity variations and the concentrations of SPM <100 µm or SPM >0.45 µm highlighted the higher loads of particles in the water column of Comprian all year long, especially at low tide. Although particles did not show any clear site difference in terms of metal concentrations, the discrepancy observed between sites in particle loads generated the site difference observed for all metals in terms of total metal concentrations in the water column (taking into account both dissolved and particulate phases). This also explains why higher total metal concentrations were observed at periods where particle contents were higher.

In sediments and particles, granulometry and C and N isotope compositions highlighted a relative homogeneity of these compartments over the year at each site, and no major site difference. This is probably due to significant reworking (i.e. tidal resuspension) of fine sediment particles over time and across the whole bay, especially during spring tides generating high speed currents, as previously reported in this system (Dubois et al., 2012) where processes of sediment accretion and erosion occur at centimetric to decimetric levels (Ganthy et al., 2013). In the Arcachon Bay, about two- thirds of the lagoon's total volume is flushed in and out at each tidal cycle (Plus et al., 2009) and it is well-established that tides can remobilize metal contaminants from the sedimentary stratum into the water column in coastal areas (Bancon-Montigny et al., 2019). Fine sediment particles then become a non-negligible part of the suspended particles. The low and poorly variable percentages of OM we measured in particles indicated that they were mainly inorganic, probably resulting from recurrent tidal sediment resuspension over time and all over the bay. Therefore, all of this explains, at least in part, why no clear nor consistent site differences nor temporal variations were observed in the metal concentrations of sediments and particles.

4.4. Relationships between metal concentrations in particles and particle characteristics

Firstly, the percentages of particulate metals and their variations throughout the study period were consistent with the known distribution and partition of the study metals in transitional waters, with partition coefficients for the dissolved and particulate phase (K_D) in the order: Pb > Cu, Zn > Ni > Cd (Chiffoleau et al., 1994). Among the five metals considered, Cd presented the lowest percentage of particulate metal over the year, coherent with its low affinity for particles in saline waters, due to its tendency to form soluble chlorocomplexes (Chiffoleau et al., 1994). This non-conservative geochemical behaviour likely explains the near absence of correlations observed for Cd with SPM characteristics.

Secondly, the negative correlations observed between metal concentrations in particles (SPM 6–100 µm) and the concentrations of SPM <100 µm (except for Cd) suggested that resuspension of sediment particles in most parts of the bay and at each tidal cycle (see above) tend to dilute metal concentrations in the suspended material originally discharged from rivers (when expressed as a quantity of metals for a given mass of particles). As particles were mainly mineral (i.e. average % of OM = 16.9 ± 5.0%), this implies that metals are mainly bound to the organic phase of particles, which is indeed confirmed by the positive correlations (except for Cd) observed between metal concentrations in particles and the percentage of OM in particles. Finally, the negative correlations observed between metal concentrations and pigments (chlorophyll *a* and pheopigments) indicate that particulate metals are probably not related to fresh (or only partly degraded) phytoplankton but to another fraction of the particulate OM (POM). We may hypothesize that it corresponds to a terrestrial fraction of the POM (originating from rivers), but further analyses would be necessary to test this hypothesis.

Interestingly, Cu was the element with the highest correlation coefficients and the only one presenting a significant (although slight) correlation with the number of phytoplankton taxa (corresponding to biodiversity of phytoplankton taxa). Adorption and absorption processes onto particles, including phytoplankton, are very complex mechanisms to apprehend in situ and most metals have a marked ability to be adsorbed onto small particles (dead or alive, organic or inorganic). However, for Cu in particular these ad- and absorption processes were documented to vary widely according to microalgae species (Levy et al., 2008).

4.5. Spatial variations of transplanted oysters' growth and condition index

The biometric parameters measured for transplanted oysters first evidenced significant spatial variations in oysters' growth and condition index between the two study sites, with Comprian lower than GB. The higher C:N ratio measured in GB oysters at T3 also confirmed the better body condition of GB oysters compared to Comprian ones, as this ratio may be considered as a proxy of the lipid content or of the body condition in aquatic organisms (Post et al., 2007).

The lower growth rate of oysters from Comprian may be primarily due to both trophic and/or physiological aspects. From a trophic point of view, although the difference was not significant between sites when considering all year data, higher densities of microphytoplankton were observed at GB than at Comprian during spring 2017 and winter 2018, corresponding to periods of phytoplankton blooms in the bay during the study. Therefore, oysters from GB may have had more access to food during these periods, at least in terms of food quantity. In terms of food quality, the trophic parameters measured in oysters (i.e. C and N stable isotopes) suggested that the food assimilated by oysters was similar at the two sites. From a physiological point of view, we also showed that oysters from Comprian face higher salinity variations and higher particle loads (inorganic particles from sediment resuspension and/or terrestrial particles from river loads) throughout the year. In oysters, the amount of pseudo-faeces produced (i.e. particles that are filtered out and released without being ingested) has been shown to increase with the concentrations of particles. These pseudo-faeces contain mucus and this loss of OM can represent a significant cost in relation to nutrition (Bayne, 2017). Furthermore, above a certain concentration in particles (seston) in the water column, a sharp reduction of filtration and rejection rates was documented in *C. gigas*, leading to physical constraints limiting food acquisition (Barillé et al., 1997). All of this may contribute to the lower growth rate of oysters at the Comprian site. However, an additional possible impact of environmental metal contamination (shown to be higher in seawater from Comprian, for instance) on i) oyster growth rate directly (i.e. due to the loss of energy allocated to detoxify metals and/or counteract potential impacts; e.g. Smolders et al., 2004), or on ii) the nutritional value of the trophic resources (i.e. indirect impact, linked to the diminution of phytoplankton content in terms of essential fatty acid profiles, for instance; Filimonova et al., 2018) cannot be excluded.

4.6. Spatial variations in transplanted oysters' metal concentrations and body burdens

Following the same tendencies observed for dissolved and total metal concentrations in "bulk" seawater (including the particulate phase), concentrations were higher in oysters from Comprian than those from GB for all metals except Pb. Interestingly, the site difference was inverse when considering metal body burdens (µg instead of concentrations), with oysters from GB that reached higher body burdens than oysters from Comprian at T3, for all metals except Cu (i.e. for Cu, Comprian = GB at T3).

Broadly, the metal concentrations observed at the end of the transplantation period (and thus at the end of this one-year study) were in the same order of magnitude or even slightly lower than those reported by the ROCCH network in recent years, at least for the Comprian site (i.e. site monitored by the ROCCH network). However, the discrepancy observed between metal concentrations versus body burdens in terms of site differences was clearly linked to the site difference in oyster growth and condition index. As such, the lower growth rate of oysters from Comprian led to higher metal concentrations observed at this site for most metals. Considering concentration data only may thus induce a bias on the interpretation of metal contamination and bioaccumulation in oysters. Results of body burdens indeed rather suggested that oysters from GB are at least equally, if not more, contaminated with metals (as proven by their higher metal body burdens (i.e. higher metal quantity in an average individual) for most metals (except Cu) at T3 in particular), but their metal load is clearly more biodiluted in their tissues compared to those from Comprian (leading to lower metal concentrations at GB than at Comprian).

By considering a ratio of concentrations between compartments from the same site, the BAFs calculated for oysters finally allowed us to i) evaluate the "bioaccumulation potential" of the different metals by oysters at a given site and to ii) compare the "bioaccumulation potential" of oysters between sites for a given element. In other words, BAFs can help to assess if oysters have a higher ability to concentrate a metal relative to the concentration of that metal in the water column at one site or another, for a given exposure through the water column (dissolved plus particulate metal phases). At GB, where total metal concentrations in the water column were lower than at Comprian, oysters had higher BAFs and hence a higher "bioaccumulation potential" for all metals except Cu, as already evidenced by body burdens. As alluded to above, this may be partly explained by the fact that oysters from GB may have ingested more food and associated ad- or absorbed metals than oysters from Comprian over time, the trophic pathway remaining the major route for metal incorporation by bivalves (Ke and Wang, 2001; Metian et al., 2009a, 2009b; Wang and Fisher, 1996). However, this does not explain the opposite trend found for Cu in terms of site differences for BAFs compared to other metals.

4.7. Possible drivers of oyster Cu contamination in the Arcachon bay

Our metal body burden results indicated a metal contamination of oysters from the whole bay, even those cultivated in the outermost part (i.e. at the GB site). This was also confirmed by BAF values, evidencing a "bioaccumulation potential" of oysters from both sites in the bay, which was even higher at the GB site. Concomitantly, in the innermost part of the bay (i.e. Comprian site), i) higher concentrations of particles throughout the year, ii) higher metal concentrations in seawater and in the water column in general (i.e. "bulk" seawater including both dissolved and particulate phases) and iii) lower growth rate and condition indices were measured in oysters. As mentioned above, the higher loads of particles (and associated contamination) probably play a role in the lower growth rate and hence higher metal concentrations measured in Comprian oysters. These higher loads of particles themselves are likely due to the seagrass disappearance that has mainly been observed in the innermost part of the bay for nearly two decades (Cognat et al., 2018), as seagrass beds act a sediment stabilizer and reduce the resuspension rate of fine particles (Ganthy et al., 2013; Kombiadou et al., 2014).

Among the study metals, however, Cu showed some specificities. The difference between sites in Cu bioaccumulation by oysters was null when considering body burdens, and an opposite trend in BAF patterns was observed for Cu (higher BAFs at Comprian) in comparison with other metals. Overall, these results highlight peculiar determinants of Cu contamination for oysters from the bay, especially in the innermost part of the bay (near the Comprian site). From these results, we could hypothesize i) a higher bioavailability of Cu and/or ii) additional sources of Cu for oysters in this part of the bay. With regard to the first hypothesis, neither the filtered samples nor the DGTs deployed in seawater showed any site differences in terms of “labile” Cu in seawater, potentially more bioavailable to oysters. The Cu bound to dissolved OM or other ligands (potentially present in filtered samples, but not in DGT samples) would be less available to bivalves than the ionic forms of Cu (Crecelius et al., 1982; DePalma et al., 2011), the latter being retained on DGTs. As for the contaminants that could be brought to bivalves by the sedimentary and/or particulate routes, it remains complex to identify the factors that can affect the release and/or the bioavailability of contaminants present in these matrices, especially as these factors may be metal-specific and may be highly variable over time and space when sediments are regularly disturbed (Eggleton and Thomas, 2004). With regard to the second hypothesis, unfortunately our sampling design did not allow us to identify any evident source of Cu contamination, whether in terms of the type of analyses performed or in the spatial and temporal resolution of these analyses. Understanding the peculiar Cu contamination of oysters from the bay (and its increasing trend in the last decade) therefore remains a challenging issue, although our results in seawater (and unpublished data from the “Syndicat Intercommunal du Bassin d'Arcachon”, SIBA) suggest that local river flows (including the Leyre river) probably contribute a non-negligible part of Cu contamination (as well as other metals) in the bay. While Cu isotope signatures have recently attested an anthropogenic origin for the bioaccumulated Cu in oysters, the source(s) identification remained elusive (Araújo et al., 2021). Copper is an element with a known biological role within a narrow range of concentrations but as with all essential elements, it may induce toxic effects on aquatic organisms at high concentrations. For instance, the deleterious effects of Cu on development have been proven as early as the larval stages in oysters, at concentrations not far (although slightly higher) from the environmental levels (Sussarellu et al., 2018). In oyster spat fed with phytoplankton cultures contaminated with Cu at realistic environmental concentrations, a significant impact on their condition index was also demonstrated, probably due to changes observed in phytoplankton fatty acid profiles and increased energy demand in spat (Akcha et al., 2022).

4.8. Conclusions and prospects

This one-year field study in Arcachon Bay identified i) higher loads of particles in the water column of the innermost part of the bay (Comprian site), probably due to inputs brought by the main fluvial outlet (Leyre river) and the tidal resuspension of fine sediments accentuated by the decline of seagrass beds in the area. These two sources of particles and metals, supplemented by the reduced renewal of waters in the innermost part of the bay, likely have a major role in ii) the transfer of relatively high metal loads into the water column (dissolved plus particulate phases) and iii) may explain the higher bioaccumulation patterns observed for most metals in transplanted oysters from the innermost part of the bay (Comprian site) when considering soft tissue concentrations. These more pronounced turbidity and metal stresses experienced by oysters from this site were indeed associated with lower growth rates compared to oysters transplanted in areas under more marine influence (GB site), leading to increased oyster metal concentrations at Comprian while metal body burdens are similar to GB or present an inverse trend (for Cu: GB>Comprian). Thus, in addition to a potential impact of metal contamination on oysters that cannot be excluded, the higher loads of suspended particulate material at Comprian may have a more direct impact on oyster growth rates by inducing physical constraints, higher production of pseudo-faeces produced (loss of OM) and/or limited food and energy acquisition by oysters at this site.

In this major French oyster production area, the main findings of this study have important implications from both environmental and socio-economic perspectives. Firstly, it has implications in terms of the use of bivalves as biomonitors of chemical contamination in such coastal environments. Our results indeed demonstrate the importance of considering biometric and environmental conditions when interpreting the concentration data measured in bivalve species, especially for inter-site or inter-annual comparisons, in addition to taking into account potential differences between bivalve species in terms of bioaccumulation capacities (Cai and Wang, 2019; Hummel et al., 1997). More broadly, our findings argue in favor of complementary approaches to monitor and assess the temporal trends of metal contamination in coastal areas, including monitoring water and sediment metal concentrations as thoroughly as possible, to better understand any trends of increasing metal concentrations observed in bivalves (as did the ROCCH network for Cu in oysters from the Comprian site since the early 2000s; see introduction). In terms of mariculture production, our results highlight that human-induced changes (at least partly) in environmental conditions (i.e. natural habitat modifications and contaminant emissions) can lead to a decreased economic value of oysters by affecting growth rates and sanitary risk associated with the consumption of sea products contaminated by metals.

Finally, this study identified local and more fundamental research perspectives. Firstly, our findings evidenced peculiar observations for Cu among the different metals considered. Further studies are needed to fully address this Cu contamination issue and identify Cu sources in Arcachon Bay, using for instance novel techniques such as non-traditional metal isotopes (i.e. Cu isotopes) measured in oysters and/or in the surrounding matrices (e.g. Araújo et al., 2021). Our study also brought important knowledge in terms of environmental metal concentrations in seawater and particles (including phytoplankton) for further conduction of experimental studies on trophic transfers, for instance (Akcha et al., 2022). Although the results are sometimes difficult to extrapolate to in situ observations, experimental studies remain the most reliable way to study specific aspects of metal bioaccumulation dynamics over time in organisms, by controlling some of the environmental conditions (e.g. temperature, salinity, pH, etc.) that may affect metal speciation, bioavailability and hence metal incorporation and/or excretion by organisms.

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Declaration of interests

☒The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

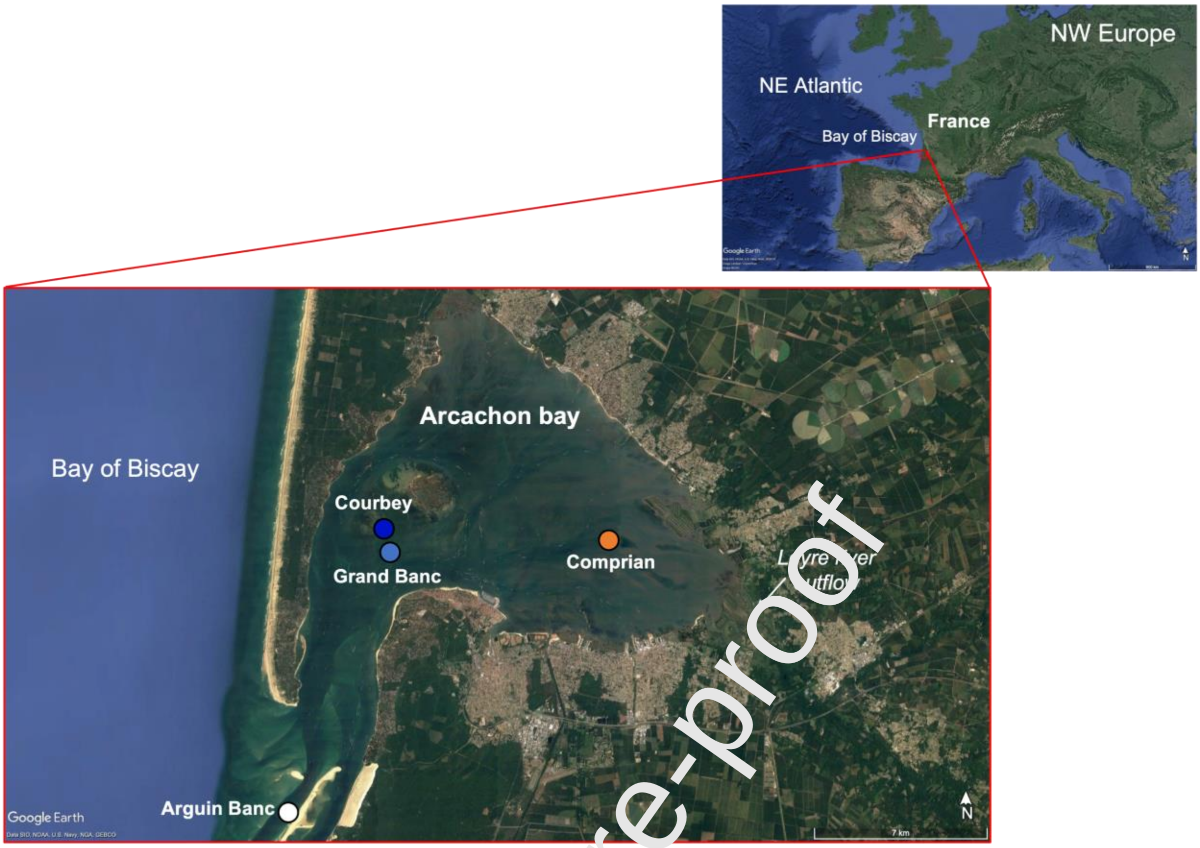


Figure 1: Maps of the study area (Arcachon bay) on the French coasts of the Bay of Biscay, NE Atlantic. The two contrasted sampling sites in the bay are indicated (Comprian and Grand Banc/Courbey) as well as the Leyre river outflow near the Comprian site, and the site outside the bay (Arguin Banc) where oysters were born and raised for 18 months before being transplanted to Comprian and Grand Banc. As also indicated in the lower left corner, maps were generated and adapted from "Google Earth, Data SIO, NOAA, U.S. Navy, NGA, GEBCO, Image Landsat / Copernicus and Image IBCAO" in January 2022.

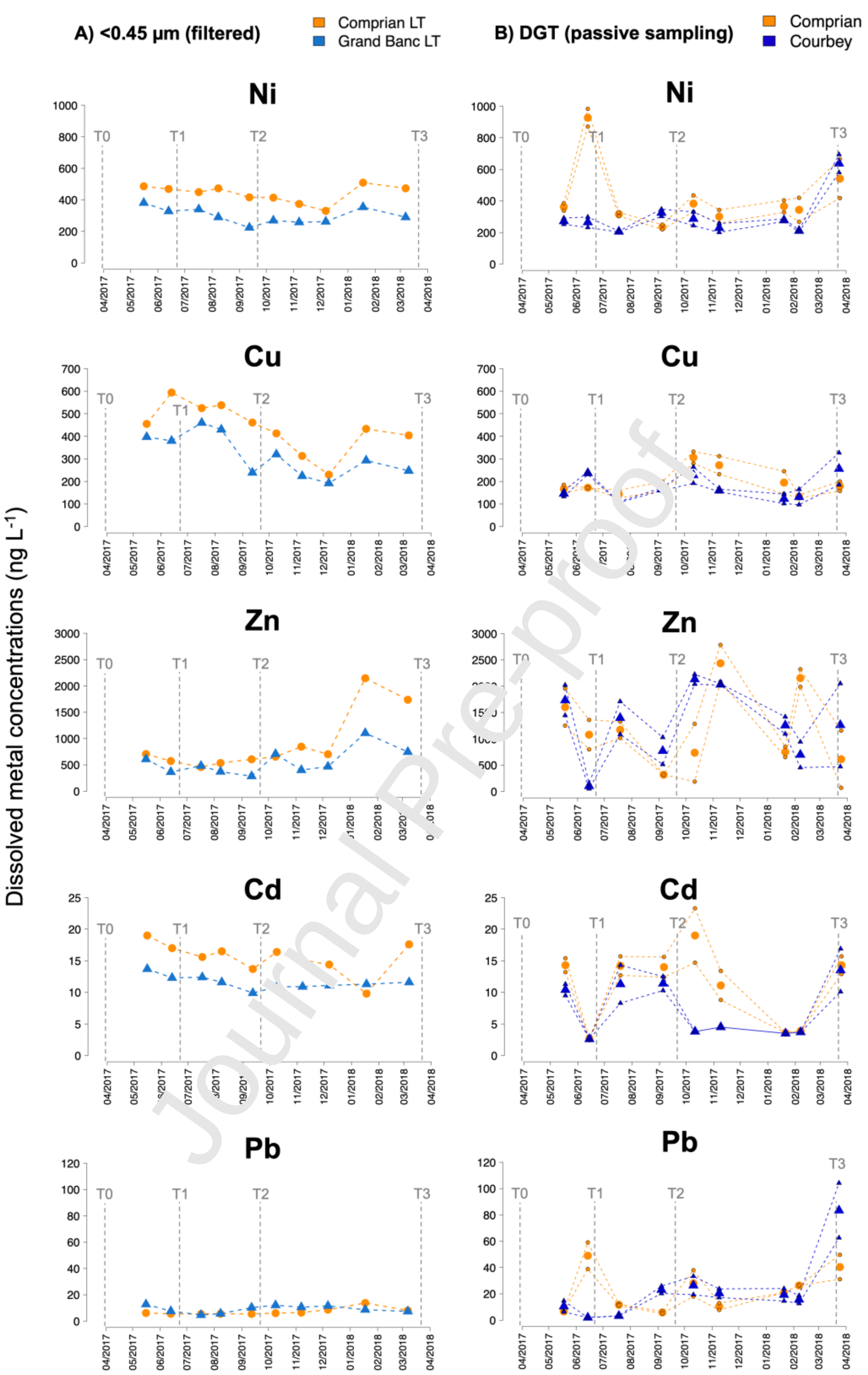
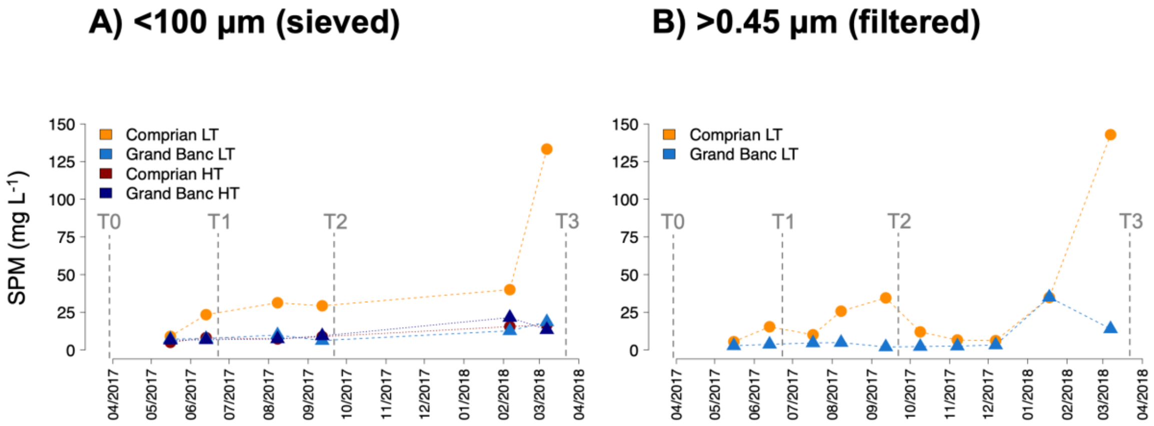


Figure 2: Dissolved metal concentrations measured over time at both sites (Comprian (orange dots) and Grand Banc or Courbey (blue triangles)), with A) concentrations in filtered samples (<0.45 µm, collected once a month at low tide only) versus B) average concentrations in Diffusive Gradient in Thin films (DGTs, monthly few days immersion), including the standard deviations of triplicate analysis for DGTs (smaller dots around mean values at each sampling date, linked with dotted lines). Sampling periods of transplanted oysters (T0, T1, T2, T3) are also indicated.



A) <100 µm (sieved)

B) >0.45 µm (filtered)

Figure 3: Concentrations of suspended particulate matter (SPM) concentrations over time at both sites (Comprian (orange dots) and Grand Banc or Courbey (blue triangles)), with A) SPM fraction <100 µm (sieved samples on the field) and B) SPM fraction >0.45 µm (laboratory filtered samples). Sampling periods of transplanted oysters (T0, T1, T2, T3) are also indicated.

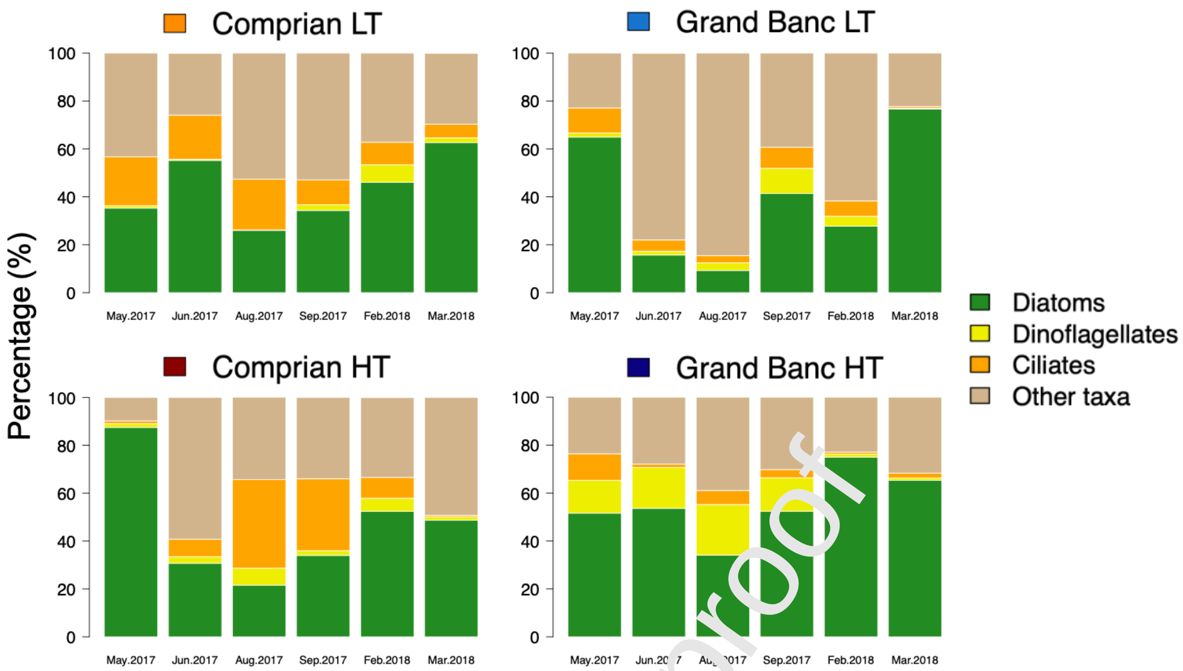


Figure 4: Taxonomic composition of the total microphytoplankton for the 6-100 µm fraction at each site and tide, in terms of percentages of the major taxa enumerated over time. LT = Low tide, HT = High tide.

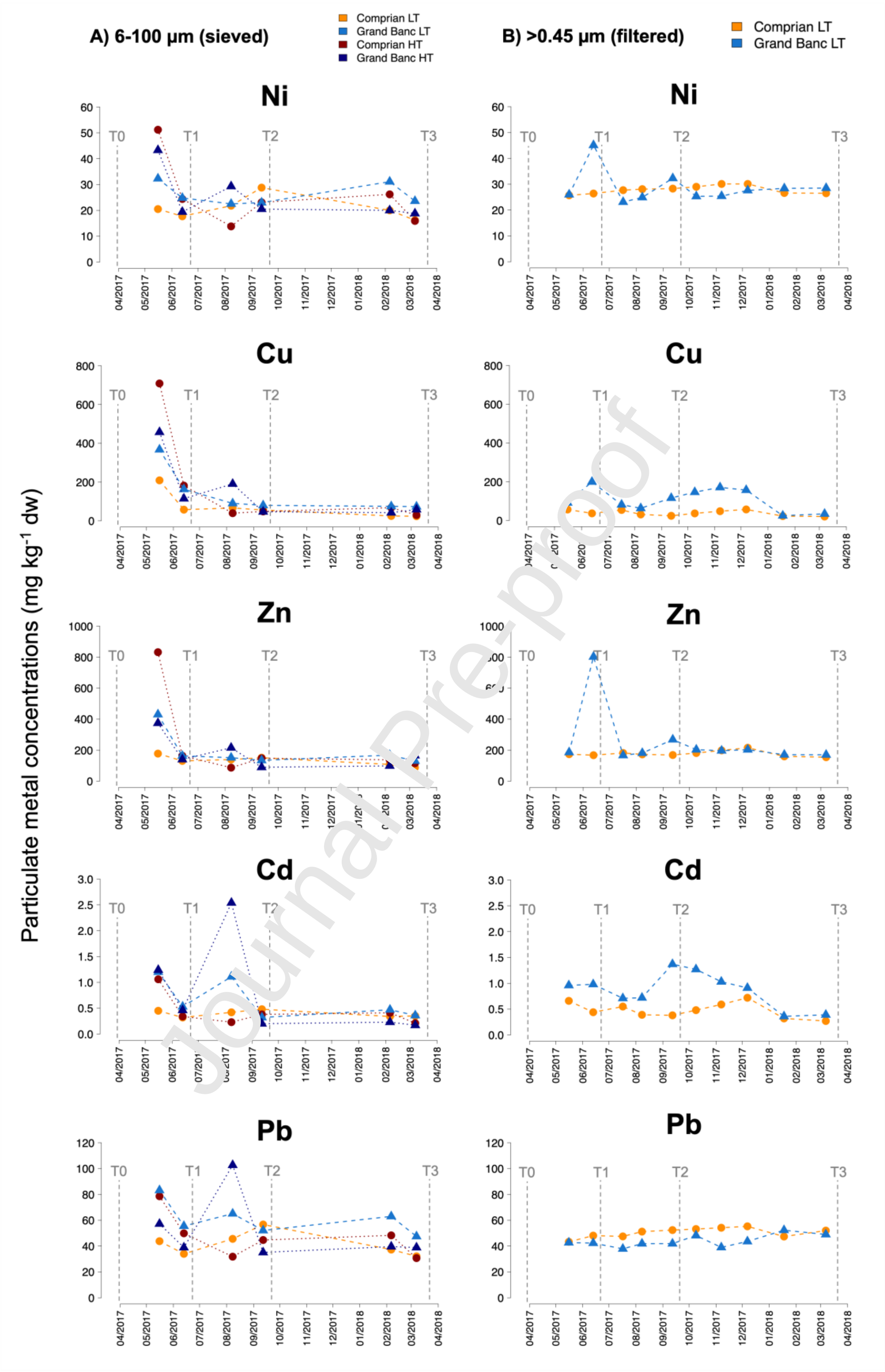


Figure 5: Particulate metal concentrations over time at both sites (Comprian (orange dots) and Grand Banc (blue triangles)), with A) concentrations in the SPM 6-100 µm fraction (sieved samples on the field) *versus* B) concentrations in the SPM fraction >0.45 µm (laboratory filtered samples). Sampling periods of transplanted oysters (T0, T1, T2, T3) are also indicated.

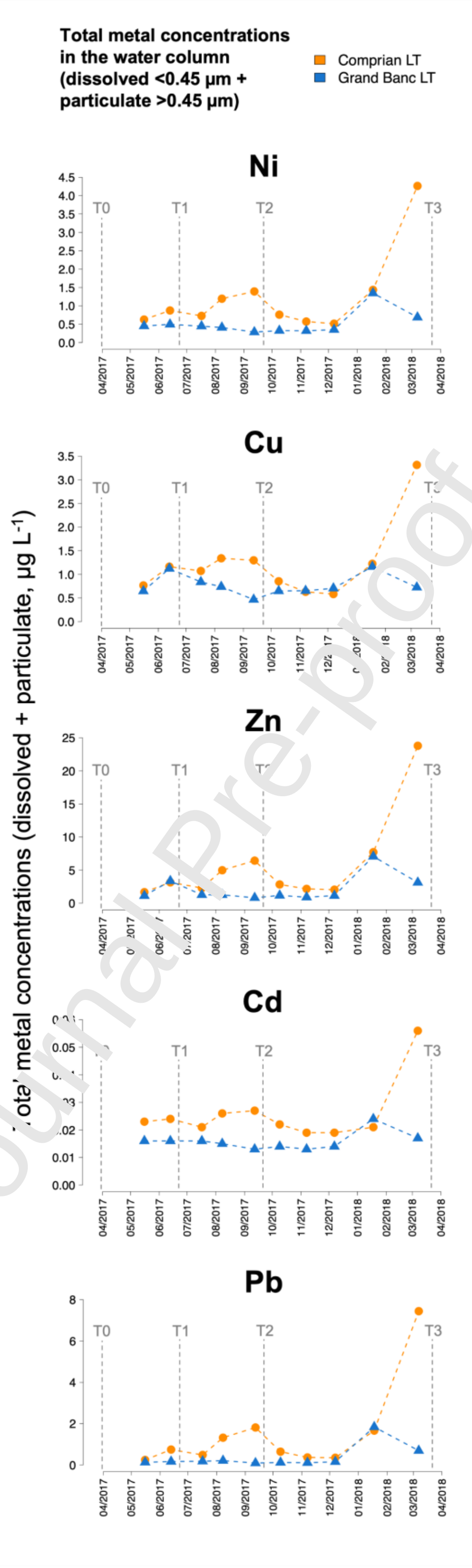


Figure 6: Total metal concentrations in the water column over time at both sites (Comprian (orange dots) and Grand Banc (blue triangles)), including both dissolved (<0.45 µm) and particulate (>0.45 µm) metals. Sampling periods of transplanted oysters (T0, T1, T2, T3) are also indicated.

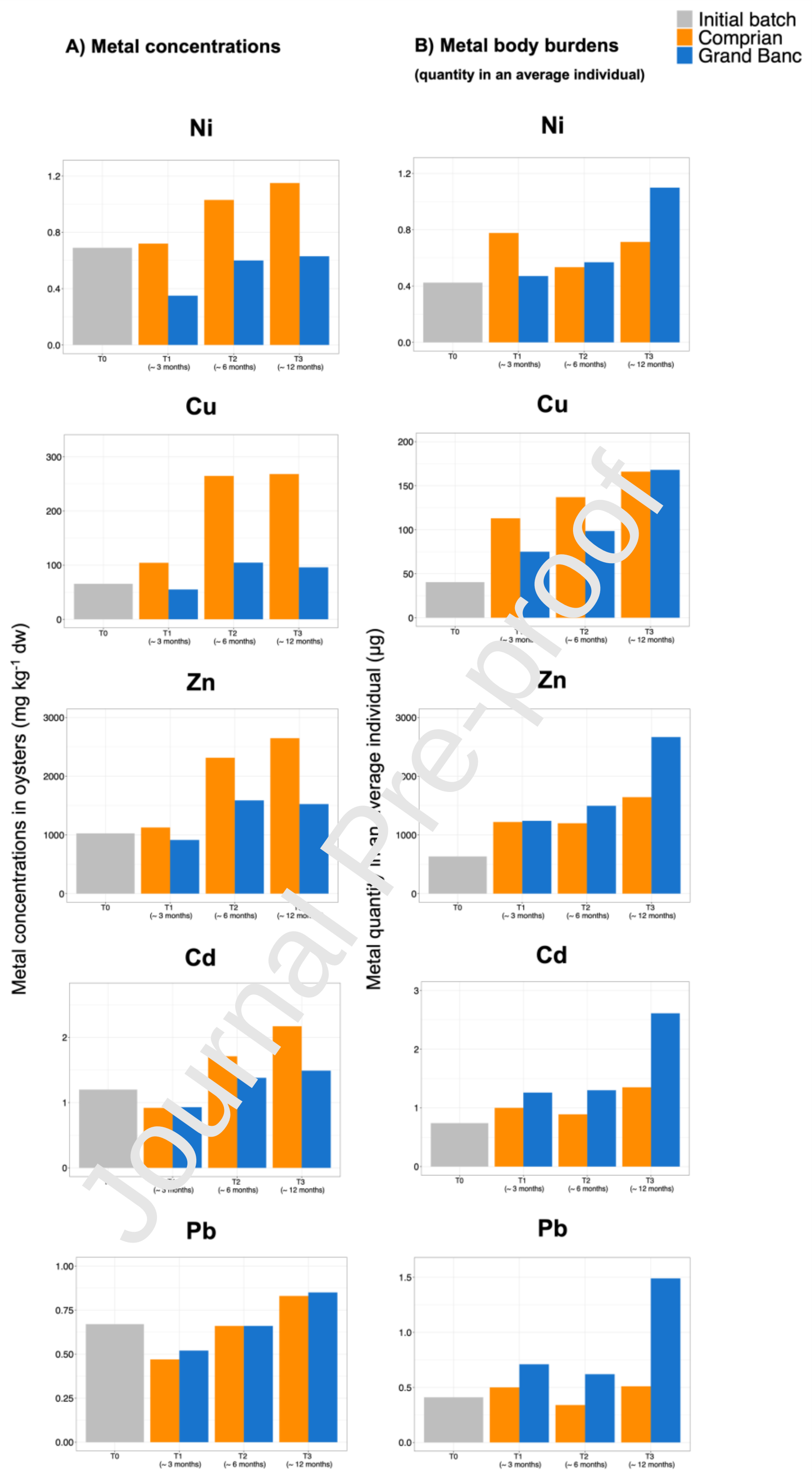


Figure 7: Metals in oysters, at each site (Comprian (in orange and on the left) and Grand Banc (in blue and on the right)) and for each sampling period (numbers of months after transplantation ; T0 =initial batch), with A) measured metal concentrations in oysters' soft tissues versus B) calculated body burdens (metal quantity in an average (dried) oyster individual, based on the mean individual flesh dry weight measured on the 30 oysters used for biometric measurements at each site and for each sampling period).

Table 1: Results obtained on sediments, at each sampling period and site: $\delta^{13}C$ and $\delta^{15}N$ values (in ‰, acquired on decarbonated samples) and metal concentrations (in mg kg^{-1} dry weight). For metals, both raw and data normalised to aluminium 5% are given.

	Beginning of the study (25/04/2017)		**End of the study (22/03/2018)**	
	Comprian	**Grand Banc**	**Comprian**	**Grand Banc**
$\delta^{13}C$	-19.6	-20.7	-20.5	-20.2
$\delta^{15}N$	5.6	5.7	6.0	5.3
Ni				
Raw data	17.6	10.3	9.9	10.2
Normalised data	17.5	15.9	13.8	15.2
Cu				
Raw data	10.1	6.5	5.3	5.2
Normalised data	10.0	9.9	[illegible]	7.8
Zn				
Raw data	96.3	52.8	53.1	51.9
Normalised data	95.4	81.1	73.7	77.6
Cd				
Raw data	0.16	0.08	0.08	0.06
Normalised data	0.16	0.12	0.11	0.10
Pb				
Raw data	34.3	25.5	22.0	21.8
Normalised data	33.9	39.2	30.5	32.6

Table 2: Dissolved metal concentrations measured in seawater at both sites (Comprian and Grand Banc or Courbey for passive sampling), in filtered samples (<0.45 µm, collected once a month at low tide only) or in Diffusive Gradient in Thin films (DGTs, monthly few days immersion). Values are means ± standard deviations over the one-year field study (minimum – maximum).

	Comprian	**Grand Banc or Courbey**	**Both sites**
Filtered samples (<0.45 µm), ng L^{-1}	*N = 10*	*N = 10*	*N = 20*
Ni	439 ± 56 (330 – 509)	300 ± 49 (224 – 381)	369 ± 88 (224 – 509)
Cu	437 ± 107 (230 – 594)	318 ± 94 (192 – 460)	377 ± 115 (192 – 594)
Zn	897 ± 569 (459 – 2146)	554 ± 246 (285 – 1104)	725 ± 461 (285 – 2146)
Cd	15.5 ± 2.5 (9.8 – 19.0)	11.6 ± 1.0 (9.9 – 13.7)	13.5 ± 2.8 (9.8 – 19.0)
Pb	7.1 ± 2.6 (5.4 – 14)	9.1 ± 2.8 (4.7 – 13)	8.1 ± 2.8 (4.7 – 14)
DGT samples (passive sampling), ng L^{-1}	*N = 10*	*N = 9*	*N = 19*
Ni	402 ± 203 (235 – 927)	301 ± 132 (206 – 637)	354 ± 176 (206 – 927)
Cu	181 ± 68 (64 – 306)	173 ± 54 (108 – 256)	177 ± 60 (64 – 306)
Zn	1126 ± 726 (319 – 2436)	1266 ± 661 (104 – 2133)	1192 ± 680 (104 – 2436)
Cd	10.8 ± 5.6 (2.7 – 19.0)	7.2 ± 4.3 (2.6 – 13.5)	9.1 ± 5.2 (2.6 – 19.0)
Pb	22.1 ± 14.4 (5.9 – 49.0)	22.7 ± 24.3 (2.0 – 83.4)	22.4 ± 19.1 (2.0 – 83.4)

Table 3: Particulate metal concentrations measured at both sites (Comprian and Grand Banc), in suspended particulate material (SPM) 6-100 µm (sieved samples on the field, collected 2 and 6 weeks before each oyster sampling at both low and high tides) or in SPM <0.45 µm (laboratory filtered samples, collected once a month at low tide only). Values are means ± standard deviations over the one-year field study (minimum – maximum); dw = dry weight.

	Comprian	**Grand Banc**	**Both sites**
SPM 6-100 µm (mg kg^{-1} dw)	*N = 12*	*N = 12*	*N = 24*
Ni	23.3 ± 9.8 (13.8 – 51.2)	25.7 ± 7.2 (18.8 – 43.3)	24.5 ± 8.5 (13.8 – 51.2)
Cu	126.0 ± 193.0 (23.6 – 708.9)	146.4 ± 133.4 (41.9 – 457.5)	136.2 ± 162.6 (23.6 – 708.9)
Zn	189.7 ± 204.2 (86.8 – 831.9)	188.2 ± 105.9 (89.4 – 430.2)	189.0 ± 159.1 (86.8 – 831.9)
Cd	0.42 ± 0.22 (0.21 – 1.06)	0.74 ± 0.69 (0.17 – 2.54)	0.58 ± 0.53 (0.17 – 2.54)
Pb	44.5 ± 13.6 (30.7 – 78.6)	56.6 ± 20.1 (35.1 – 102.7)	50.5 ± 17.9 (30.7 – 102.7)
SPM >0.45 µm (mg kg^{-1} dw)	*N = 10*	*N = 10*	*N = 20*
Ni	27.8 ± 1.6 (25.7 – 30.1)	28.7 ± 6.3 (23.1 – 45.1)	28.2 ± 4.5 (23.1 – 45.1)
Cu	38.8 ± 14.4 (20.4 – 57.1)	108.4 ± 59.2 (24.8 – 199.3)	73.6 ± 55.1 (20.4 – 199.3)
Zn	177.2 ± 18.1 (154.4 – 215.2)	254.4 ± 194.6 (166.2 – 802.1)	215.9 ± 140.2 (154.4 – 802.1)
Cd	0.48 ± 0.15 (0.27 – 0.72)	0.87 ± 0.33 (0.36 – 1.37)	0.67 ± 0.32 (0.27 – 1.37)
Pb	50.4 ± 3.7 (43.3 – 55.3)	43.9 ± 4.6 (37.9 – 52.3)	47.2 ± 5.3 (37.9 – 55.3)

Table 4: Spearman rank order correlation coefficients (r) between trace metal concentrations and accompanying (biological) parameters analyzed on the suspended particulate matter (SPM) 6-100 µm size fraction, both tides and both sites combined. The exceptions were the parameters "concentration of SPM", "% of organic matter (OM) in SPM" and the parameters relative to taxonomical compositions, for which the sampled size fraction <100 µm was considered instead of the 6-100 µm size fraction (see 2.2.4). Significant correlations at $p<0.05$ are indicated in bold characters (with * $p<0.05$; ** $p<0.01$; *** $p<0.001$).

	Ni	**Cu**	**Zn**	**Cd**	**Pb**
Ni	—				
Cu	**0.715*** **	—			
Zn	**0.740*****	**0.863*****	—		
Cd	**0.753*****	**0.716*****	**0.723*****	—	
Pb	**0.915*****	**0.772*****	**0.787*****	**0.819*****	—
Concentration of SPM <100 µm	**-0.432***	**-0.712*****	**-0.535****	-0.375	**-0.462***
% OM in SPM <100 µm	**0.484***	**0.788*****	**0.671*****	0.403	**0.482***
Chlorophyll a	**-0.569****	**-0.606****	**-0.477***	**-0.414***	**-0.555****
Phaeopigments	**-0.518****	**-0.629*****	**-0.586****	-0.355	**-0.538****
δ^{13}C	-0.279	-0.164	-0.028	-0.209	-0.170
δ^{15}N	0.291	0.237	0.201	0.024	0.229
Total microphytoplankton	-0.260	0.117	-0.015	-0.208	-0.227
Total diatoms	-0.265	0.007	-0.112	-0.350	-0.364
Total dinoflagellates	0.045	0.262	0.086	0.024	0.046
Total ciliates	-0.147	0.072	0.078	-0.034	-0.190
Number of phytoplankton taxa	0.315	**0.423***	0.367	0.180	0.274

Table 5: Results obtained on oysters, at each site and sampling period (numbers of months after transplantation): individual length (mm), total wet weight (g), shell weight (g), flesh dry weight (g) and condition index for biometric parameters (means ± standard deviation when appropriate), $\delta^{13}C$ and $\delta^{15}N$ values (‰) and bulk C:N ratios (mol mol^{-1}), metal concentrations measured in oysters' soft tissues (mg kg^{-1} dry weight), and calculated metal quantities in an average oyster individual (µg). Ratios between Comprian (COM) and Grand Banc (GB) values for biometric parameters and metal data are also indicated (in italics).

	T0 (initial batch) 30/03/2017	**T1 (~3 months) 26/06/2017**			**T2 (~6 months) 25/09/2017**			**T3 (~12 months) 22/03/2018**		
		Comprian	**Grand Banc**	***COM/GB ratio***	**Comprian**	**Grand Banc**	***COM/GB ratio***	**Comprian**	**Grand Banc**	***COM/GB ratio***
Individual length	66.7 ± 6.9	69.2 ± 8.3	73.3 ± 8.6	*0.9*	71.2 ± 7.5	76.6 ± 7.3	*0.9*	70.7 ± 6.1	87.6 ± 13.4	*0.8*
Individual total wet weight	24.3 ± 5.1	34.0 ± 6.1	38.8 ± 8.3	*0.9*	39.9 ± 6.2	49.3 ± 9.1	*0.8*	45.2 ± 7.6	73.4 ± 19.4	*0.6*
Individual shell weight	15.2 ± 2.7	21.8 ± 4.4	24.6 ± 5.6	*0.9*	26.4 ± 4.2	31.8 ± 6.3	*0.8*	30.3 ± 5.5	49.5 ± 13.5	*0.6*
Individual flesh dry weight	0.62 ± 0.21	1.08 ± 0.31	1.36 ± 0.28	*0.8*	0.52 ± 0.13	0.94 ± 0.27	*0.6*	0.62 ± 0.19	1.75 ± 0.37	*0.4*
Condition index [a]	69.2	87.9	96.1	*0.9*	38.3	53.7	*0.7*	45.7	94.8	*0.5*
C:N ratio	6.1	5.1	5.2	—	5.0	5.6	—	5.2	6.4	—
$\delta^{13}C$	-19.5	-20.5	-20.4	—	-19.7	-20.4	—	-19.2	-19.6	—
$\delta^{15}N$	7.3	7.2	7.3	—	6.8	7.4	—	7.7	7.1	—
Ni										
Concentration	0.69	0.72	0.35	*2.1*	1.03	0.60	*1.7*	1.15	0.63	*1.8*
Quantity in an average individual	0.42	0.78	0.47	*1.7*	0.53	0.57	*0.9*	0.71	1.10	*0.6*
Cu										
Concentration	65.5	104.3	55.0	*1.9*	264.7	104.6	*2.5*	268.2	95.9	*2.8*
Quantity in an average individual	40.4	112.9	74.5	*1.5*	137.0	98.5	*1.4*	166.5	167.6	*1.0*
Zn										
Concentration	1026	1127	914	*1.2*	2316	1589	*1.5*	2649	1526	*1.7*
Quantity in an average individual	633	1220	1240	*1.0*	1199	1496	*0.8*	1644	2668	*0.6*
Cd										
Concentration	1.20	0.92	0.93	*1.0*	1.71	1.38	*1.2*	2.17	1.49	*1.5*
Quantity in an average individual	0.74	1.00	1.26	*0.8*	0.89	1.30	*0.7*	1.35	2.61	*0.5*

Pb										
Concentration	0.67	0.47	0.52	*0.9*	0.66	0.66	*1.0*	0.83	0.85	*1.0*
Quantity in an average individual	0.41	0.50	0.71	*0.7*	0.34	0.62	*0.5*	0.51	1.49	*0.3*

[a] Condition Index from Lawrence and Scott (1982).

Table 6: Bioaccumulation factors (BAFs) in oysters, calculated from total metal concentrations in the water column (dissolved <0.45 µm + particulate >0.45 µm), for each sampling site and period (numbers of months after transplantation).

		Ni	**Cu**	**Zn**	**Cd**	**Pb**
Comprian	T1 (~3 months)	957	108 277	469 465	39 906	943
	T2 (~6 months)	1 417	247 043	1 013 017	81 125	1 361
	T3 (~12 months)	961	200 290	532 717	81 999	627
	Mean BAF	***1 112***	***185 203***	***671 733***	***67 677***	***977***
Grand Banc	T1 (~3 months)	733	62 191	409 353	57 433	3 558
	T2 (~6 months)	1 353	124 856	1 269 021	87 994	3 631
	T3 (~12 months)	1 529	130 105	1 219 377	98 278	4 075
	Mean BAF	***1 205***	***105 717***	***965 917***	***81 235***	***3 755***

Highlights:

- A multi-compartment and multi-parameter study to unravel oyster metal contamination
- Higher contamination found in dissolved and particulate seawater phases inside the bay
- Contamination likely explained by river flows and tidal resuspension of sediments
- Higher particle loads observed with higher oyster metal concentrations and lower growth
- Differences in oyster bioaccumulation patterns for copper compared to other metals