

Spatial and temporal variations in trace metal concentrations in surface sediments of the Marennes Oléron Bay. Relation to hydrodynamic forcing

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Abstract:

Sediments quality assessment is of priority concern to provide a comprehensible overview of ecological and chemical state of an ecosystem. The Marennes-Oléron Bay, hosting the largest oyster production in France, is influenced by the historic polymetallic pollution of the Gironde Estuary. Despite, management efforts and decreasing emissions in the Gironde watershed, Cd levels in oysters from the bay are close to consumption limit ($5 \mu\text{g g}^{-1}$ dw, EC no. 466/2001). In this context, the aim of the study was to assess the priority metal (Ni, Cu, Zn, As, Ag, Cd, Hg, Pb and Th) concentrations in sediment within the Bay, by investigating spatial and temporal distribution variations and the role of hydrodynamic forcing. For that we selected three sites (east, west and south) characterizing different environments of the Bay and we observed metal concentrations, grain size distribution, bed elevation and wave activities during a year survey. The sampling strategy pointed out both spatial and temporal metal concentrations variability in sediment. In general, metal enrichments were close to geochemical background. The eastern part of the Bay, largely influenced by the Charente river particulate deposition, presented constant concentrations over the survey. In contrast, in the western part, bed elevation was strongly influenced by hydrodynamic forcing especially wave activities, and metal distribution showed constant metal concentrations except very located Cd minor enrichment related to the Gironde influence via the Antioche Strait (north). The southern part was disconnected from the rest of the Bay and showed minor to very located moderately severe Cd enrichment, related to the Gironde water discharges via the Maumusson Strait (south). Thus, the multi-disciplinary approach was relevant to characterize the interactions between hydrodynamic forcing on the environment and sediments and their metal quality state which (i) were close to geochemical background over a year for Ni, Cu, Zn, As, Ag, Hg and Pb (i) which presented enrichment of Cd in the western and southern part.

Highlights

► Spatial and temporal trace metal concentrations (Ni, Cu, Zn, As, Ag, Cd, Hg, Pb and Th) were assessed in the Marennes Oléron Bay. ► Metal concentrations, grain size distribution, bed elevation and wave activities were surveyed during a year at three sites to characterise the role of hydrodynamic forcings. ► The pluri-disciplinary approach pointed out that the three sites were varying independently and showed different temporal patterns in terms of sedimentary processes and trace metal concentrations.

Keywords : Trace metal ; Marennes Oléron Bay ; Enrichment factor ; Sediment ; Hydrodynamic ; Altus

1. Introduction

Sediment particles have been identified as key factors in metal/contaminants transport from the continent to the ocean and play an important role in river basin and coastal area (Förstner and Salomons, 2008). They can act as a source or a sink of metal-bounded particles or dissolved metal in the interstitial water, which can significantly modify contaminant distribution, behaviour and ecotoxicological impact. In coastal zone, sediments may be considered as ultimate receptacle of marine organic/inorganic suspended particulate matter (SPM) and terrigenous inputs from estuarine and riverine watersheds. However, coastal tidal dynamic can induce sediments resuspension and remobilisation which affect trace metal cycle. The Water Framework Directive (Förstner and Salomons, 2008)(WFD) aims at attempting a good ecosystem health in aquatic systems (2000/60/EC). The ecological status is based upon the status of biological, hydromorphological and physicochemical quality elements, including priority substances (2005/105/EC), as trace metals (Borja et al., 2004). Despite sediment assessment is not mentioned in the WFD monitoring, their quality assessment is of major scientific concern (Apitz and Power, 2002; Crane, 2003; Borja et al., 2004; Apitz, 2008; Tueros et al., 2009, Corbett et al., 2009; Essien et al., 2009; Valdès et al., 2009).

The Marennes-Oléron Bay (MOB, Figure 1a) is a semi-closed bay of 156 km², with 58 % of intertidal areas and a tidal range of 5 m (Bassoullet et al., 2000; Gouleau et al., 2000) located between the Oléron Island and the Charente coast in south-western France. The Bay hosts the largest oyster production in France, as one third of its intertidal areas are covered by oyster structures several months per year (Gouilletquer and Le Moine, 2002). The annual production of 55 to 60 ton.yr⁻¹ generates important regional economy related to farming and tourism activities. However, the MOB is influenced by the historical cadmium pollution from the Gironde Estuary (Latouche, 1988; Boutier et al., 1989; Jouanneau et al., 1990; Boutier et al., 2000; Dabrin, 2009). The Cd is transported through the dissolved phase (Boutier et al., 2000; Dabrin, 2009) and through fine sediments and particles (Pouliquen, 1975; Castaing and Allen, 1981; Boutier et al., 1989; Froidefond et al., 1998; Parra et al., 1999; Boutier et al., 2000; Dabrin, 2009). The pollution of the Lot-Garonne-Gironde fluvial-estuarine system originates 350 km upstream from former mining activities, especially ore treatment, in the Decazeville watershed (Jouanneau et al., 1990; Lapaquellerie et al., 1995; Blanc et al., 1999; Schäfer and Blanc, 2002; Audry et al., 2004b; Coynel et al., 2009; Audry et al., in press). Despite management efforts and decreasing emissions in the watershed (Audry et al., 2004a; Coynel et al., 2007), dissolved and particulate Cd net fluxes exported from the Estuary to the coastal

zone were estimated of 5100 and 800 kg.yr⁻¹ respectively (Dabrin, 2009; Dabrin et al., 2009). From the 1980s, the French National Monitoring Network (Réseau National Observation, RNO), under the IFREMER responsibility, bio-monitored Cd contamination in oysters collected in the Gironde Estuary and the MOB (RNO, 2000, 2006). The Cd contents in oysters from the Gironde Estuary (RNO, 2000, 2006) were significantly higher than the consumption limit level (5 µg.g⁻¹ dw, EC No.466/2001) conducting to the interdiction of oysters' production, selling and consumption in the Estuary. The Cd concentrations in oysters from the MOB are below but still close to this consumption limit level (RNO, 2000, 2006). Assessment of sediment quality in the MOB is thus of priority concern to implement a successful ecosystem management.

This study aims at assessing sediment metal quality by evaluating trace metal (e.g. Ni, Cu, Zn, As, Cd, Hg, Pb, more Th) in surface sediments. In such environment characterized by strong tidal variations, sedimentary processes as erosion and accretion are temporally highly dynamic. It is thus essential to determine if those sedimentary variations are accompanied by or affecting trace metal concentrations in sediments. For that, we proposed and discussed an original sampling strategy combining both geochemical and hydrodynamical survey to understand and integrate hydrologic and hydrodynamic forcing over spatial and temporal scale. Thus in the same time, spatial and temporal sediment variability, bed elevation and hydrodynamic forcing were monitored at three sites over a year in order to discuss the influence of sedimentary and hydrodynamic forcing on sediment metal quality of the Marennes Oléron bay.

2. Material and methods

2. Study area

The Marennes Oléron Bay (MOB; Figure 1b) is a shallow macrotidal bay, with an 8.6 m depth average and a tidal range of 5 m. It receives the fresh waters of Charente River in the North (with a 40-470 m³.s⁻¹ flow, (Soletchnik et al., 1998) and the ones of Seudre River in the South (with a 0-40 m³.s⁻¹ flow, according to the same authors). It can be divided into three sediment areas (Tesson, 1973; Pouliquen, 1975). The north-eastern Bay is a 40 km² large mudflat composed of 92% clay and silt particles (Gouleau et al., 2000; Galois et al., 2000), from Charente River, Seudre River and salt marshes, which locally presented specific drainage systems called "ridges and runnels". The western Bay is a marine mixed sandy and muddy flat, of which sediments are partly originated from the Gironde Estuary (Hily, 1977; Sauriau et al., 1989) and characterized by shellfish farming installations in the lower part and

seasonal presence of seagrass *zoostera noltii* in the upper part (Guillaumont, 1991). The central to southern Bay, from “Coureau of Oléron” to Maumusson Strait, is composed of calcareous and siliceous sandy sediments coming from coastal limestone cliffs and dune under littoral drift action. It is characterized by important permanent oysters farming activities, inducing sandbars and silt deposition (Kusuki, 1978; Sornin, 1981; Martin *et al.*, 1989; Kervella, 2010).

Currents circulation The particular bathymetry and the difference of size between the two openings, the Antioche Strait (North) and the Maumusson Strait (South) (Figure 1) bring a residual water circulation oriented along a north-south axis, with asymmetric ebb and flood tides (Klingebliel *et al.*, 1971; Bertin *et al.*, 2005; Stanisière *et al.*, 2006). During calm weather period, marine waters penetrate by the Antioche Strait and the Charente waters flow on the eastern side of the bay (Stanisière *et al.*, 2006). Thus, the central part of the Bay called “Coureau of Oléron” is influenced by sea water mass (salinity > 30) and less saline water mass (salinity < 30) constituted by a mixing between marine waters and Charente ones (Tesson, 1973). The southern Bay is slightly influenced by the marine waters entering via the Maumusson Strait because of the higher ebb tidal prism (Bertin *et al.*, 2005; Stanisière *et al.*, 2006). Thus, the water residence time was estimated of 4-11 days in the western part, 7-16 days in the eastern part and 0.5 to 5 days in the south of the Bay (Struski, 2005). The current maxima ($1 \text{ m}\cdot\text{s}^{-1}$) are observed during spring tides in subtidal areas (Tesson, 1973) whereas currents do not exceed $0.60 \text{ m}\cdot\text{s}^{-1}$ in intertidal areas (Bassoullet *et al.*, 2000).

Wave activity: Wave propagation in the bay comes roughly from the Antioche Strait. Waves are strongly attenuated in the south because of the narrowness of the Maumusson Strait and the singular bathymetry in this area. In the MOB, waves are responsible of significant sediment transport at these openings under littoral drift action, while wind waves cause fine sediment resuspension on mudflats (Tesson, 1973; Bassoullet *et al.*, 2000). Despite few data available, Bassoullet *et al.* (2000) have recorded waves of about 0.70 m high on the Brouage mudflat (East) using pressure sensors in stormy periods

Field sites Three intertidal sites were selected according to the Bay environmental characteristics and were studied during a year (Figure 1b). The first site, called “Les Doux”, was located in the middle of the oyster area in the low part of the western flat. During this study, no oyster installations were present. The second site called “Brouage” was located in the low part of the eastern mudflat between the wild oyster area and the “Coureau” of Oléron. The third site called “Perquis” was located on the western part of the Perquis sand bank in the southern part of the Bay, in annual oyster installation area.

2.2. Methodology and analysis

2.2.1. Sampling

Surface sediments (0-5 mm) were collected monthly at each site using a plastic spatula. Sediments were immediately introduced into a 60 mL PE tube hermetically closed to avoid re-oxidation processes and kept refrigerated in the dark at 4°C until particle size, water content and metal analyses.

2.2.2. Turbidity and salinity

Salinity and turbidity data were recorded by multi-parameters probes (Ysi) located at the Agnas bank (monitoring network RAZLEC, IFREMER) and at Les Doux and Brouage sites during 16 days (06-22 April 2007). These probes were positioned ten centimetres from the bottom and temperature and salinity were recorded once every 10 minutes.

2.2.3. Sedimentary parameters

Grain size analyses were performed using Beckman Coulter laser diffraction (LS13 320; focal 780 nm; 0.4 to 2000 µm). The upper limit of clay was fixed at 2 µm and the limit between silt and sand to 50 µm according to USDA standard. Each analysis was performed with seawater to avoid deflocculation processes and take into account environmental physico-chemical characteristics. Before analyses, a blank was realised to eliminate micro organisms and salt contents. The water content (W , in %) was defined as (Kervella et al., 2009)

$$W = \frac{M_w}{M_d} \quad \text{with } M_w: \text{ wet mass (g) and } M_d: \text{ dry mass (g) after 72 H drying at } 55^\circ\text{C}$$

2.2.4. Bed-elevation and wave measurements

Bed level variations, related to waves and tide currents, were continuously recorded at each site from April 2007 to April 2008 using altimeter devices (ALTUS; NKE®). The ALTUS system is an echo-sounder recording bed elevation and water height with an accuracy of 0.2 and 20 mm, respectively (Jestin et al., 1998). During this study, wave and bed parameters were recorded during 300 seconds every 20 minutes at a frequency of 2 Hz. The transmitter was positioned at 25 to 40 cm above the bottom. The distance between transmitter and bottom is estimated on acoustic waves speed propagation in water. Therefore, a correction of the bed elevation signal is necessary, using Coppens equation, according to changes of environmental temperature and salinity (Deloffre et al., 2005). In this study, we used temperature and salinity

data from the monitoring network RAZLEC (IFREMER) located at Agnas bank (middle MOB) to correct the acoustic signal. Thus, we determined the significant wave height H_{sig} from water level measurement, according to the following equation:

$$H_{sig} = 4\sqrt{m_0} = 4\sqrt{\int S_p(f)df} = 4\sqrt{\sum S_p(f_i)\Delta f_i}$$

where S_p is the pressure spectral density (corrected from depth attenuation), f is the wave frequency and m_0 is the variance of the water surface elevation .

2.2.5. Trace metal analysis

For analysing Cr, Ni, Cu, Zn, As, Ag, Cd, Pb and Th, representative sub-samples of surface sediments (30 mg of dried, powdered and homogenized material) were digested into acid-pre-cleaned PP-tubes (SCP sciences®) using 1 mL HNO₃ (14M, suprapur®), 3 mL HCl (12M, suprapur®) and 2.5 mL HF (26M, suprapur®). The reactors were heated at 110°C for 2 H using a temperature controlled digestion system DigiPrep® (SCP sciences). After cooling, the digested solution was evaporated until dryness, and the digestate was diluted to 10 mL using 250 µL HNO₃ (14M, suprapur®) and milli-Q water®. Trace metal concentrations were measured by ICP-MS (Thermo X7), using external calibration. The analysis and measurements were quality controlled using respectively certified marine sediments (BCSS-1) and riverine water (SLEW-3). Recoveries and reproducibility are presented in Table 1.

For analysing Hg representative sub-samples of surface sediments (70 mg of dried, powdered and homogenized material) were measured by cold vapor atomic spectrometry (DMA®) and quality controlled using IAEA-405 certified sediment.

3. Results

3.1. Salinity and turbidity

Measured salinity showed net temporal and spatial variations (Figure 2a). Three situations were pointed out: (A): salinity measured at Agnas (RAZLEC, IFREMER) and Brouage are relatively low (31,5) and followed the same tendency whereas at Les Doux salinity is slightly higher (32) and varied differently; (B): salinity measured at Les Doux was close to Agnas but lower than Brouage (10-14 February 2007), showing freshwater inputs from the Gironde Estuary via the Antioche Strait; (C): same situation as (A) with higher salinity, showing decreasing freshwater in puts to the Bay. Turbidity (Figure 2b) measured at Les Doux and Agnas (RAZLEC, IFREMER) in spring tides were relatively lower ([10-150] mg.L⁻¹) then those measured at Brouage ([10-500] mg.L⁻¹).

3.2. Grain size distribution

Sediment fractions and water contents varied temporally and spatially (Figure 3). At Les Doux and Perquis sites, sediments were dominated by the marine sandy fraction throughout the year (87 and 81 % respectively) and characterized by a mean median grain size distribution of 209 and 194 μm respectively. However, a slight increase in fine fraction (silt+clay, 26 %) was observed during the summer. In those two sites, the water content was low (40 and 43 % respectively) and constant over the survey. On the other hand, Brouage muddy sediments showed a muddy sediments with a mean fine fraction of 90 % throughout the year and a mean median grain size of 9 μm . However, the water content varied, and presented low water content (< 150 %) from April 2007 to October 2007 and higher water content (> 184 %) from November 2007 to April 2008.

3.3. Bed elevation variation and wave activity

At Les Doux area, bed elevation showed strong multi-centimeter vertical variations (SVV), which were related to wave's height and period (Figure 4a). In fact, measured waves were established from a little signification of 0.20 to 0.50 meter high throughout the year survey. During storms, recorded bed elevation was stronger, corresponding to current and wave ripples on the bottom.

At Brouage area, bed elevation and wave's height temporally changed (Figure 4b). So, the mudflat was in constant erosion ($\Delta h = - 100 \text{ mm}$) from June 2007 to the end of August 2007, then stable from September 2007 to late November 2007 and in accretion (silting up) from December 2007. In the meantime, measured wave's activity was from a value of 0.20 m to 0.70 m between May 2007 to August 2007, thus until 1.10 m (important activity) from December 2007 to the end of the survey : between these two periods, the waves were not significant.

In the southern Bay (Figure 4c), at Perquis area, sediment interface was in slight accretion ($\Delta h = + 35 \text{ mm}$) from June 2007 to December 2007, whereas waves were not significant during the year ($H_s < 0.20 \text{ m}$). However, during storm period, recorded wave heights were from 0.20 to 0.60 m, in relation with higher altimetry variations (erosion and accretion), which is characteristic with current ripples propagation.

3.4. Trace metal concentrations in Marennes-Oléron Bay sediments

Particulate trace metal concentrations (Ni, Cu, Zn, As, Ag, Cd, Hg, Pb, Th) varied spatially and temporally from May 08 to April 08 in the Marennes Oléron Bay surface sediments (Figure 5a). As no similar concentrations range and temporal variation were observed between sites, trace metal variation are described for each site.

3.1.1. Les Doux

Trace metal concentration ranges observed at Les Doux were lower than at Brouage and Perquis. All trace metals significantly varied over time with a standard error of mean (SEM) varying from 10% to 30% and Cu presented a punctual elevated concentrations in September 07 (30%<SEM<50%).

3.1.2. Brouage

Trace metal concentration ranges measured in Brouage were higher than at Les Doux and Perquis. Temporal variations were observed for Zn, As, Ag and Cd (10%<SEM<30%) whereas Ni, Cu and Hg were relatively constant (SEM<10%). Pb showed medium temporal variations (30%<SEM<50%).

3.1.3. Perquis

High temporal variations were observed for Ni, Cu, Zn, Ag, Cd (SEM>50%) and for As, Pb Hg (30%<SEM<50%). The lowest metal concentrations were observed in January 08 and the highest were observed in August 07 for Cr, Ni, and As concentrations and in September 07 for Cu, Zn, Ag, Cd, and Pb. Exceptional high Cd contents exceeding Brouage and Les Doux levels were noticed in August and September 07.

Despite spatial and temporal metal variations, Cd showed punctual high concentrations at Les Doux and Perquis and was distinguished from other trace metals.

4. Discussion

4.1 Trace metal concentrations in surface sediments

The sampling strategy, adapted to the three environments of the Bay (Tesson, 1973; Pouliquen, 1975), allowed to observe and compare spatial and temporal variations of metal concentrations in the Bay. First of all, trace metal concentrations in sediments varied spatially (Figure 5a). Higher metal concentrations were observed in muddy-sediments from the eastern part than sandy sediments from the western and southern parts of the bay. Those observations are consistent with increasing metal-adsorption sites with decreasing grain size described in the literature (Loring, 1991; Horowitz et al., 1999). The sampling strategy pointed out

different temporal metal distributions between sites. In fact, in the east and western part, metal concentrations variations were closely related to sediment fractions (Figure 3) and median grain size distribution whereas in the southern part accentuated temporal variations were observed with higher concentrations in September /October 07.

In the late 1980s, a previous study on trace metal concentrations (Cu, Zn, Cd, Hg, Pb) in MOB sediments showed homogenous distribution between the eastern and western part of the MOB (Gonzalez et al., 1991). Sediments were sampled at one time, close to Brouage and Les Doux sites which were characterised at this period by homogenous grain size distribution (mean: 10 μ m; minima: 8 μ m; maxima: 15 μ m; (Gonzalez et al., 1991)). As the same analytical procedure were performed in both studies (acid digestion HNO₃+HCl+HF for Cu, Zn, Cd, Pb analysis and cold-vapor atomic spectrometry for Hg (Toth and Ingle, 1977; Sturgeon et al., 1982), we have compared this previous study to Brouage sediments (mean: 9 μ m; minima: 7 μ m; maxima: 12 μ m) in order to provide a decadal tendency of trace metal concentration variations in the MOB sediments. Thus, Cu and Hg values were close to Brouage mean concentrations, Cd and Zn appeared to decrease over time whereas Pb to increase. However, we have to keep in mind that (i) the previous study (Gonzalez et al., 1991) presented only a spatial distribution and that (ii) present temporal variations in the MOB are significant. Thus, taking into account the annual variability, (i) Cu, Hg and Zn are in the same concentration range and do not present decadal variations, (ii) Cd slightly decreased over the last two decades and (iii) Pb is in a steady state in the case of excluding its exceptional local increase in May 07. Those observations suggest a double balanced process, between (i) the removal of metal from the water column, estuaries and coastal inputs into the sediments and (ii) the metal inputs (diffusion, desorption) from the sediments to the water column. Present metal concentrations observed in the MOB are relatively low, in comparison to lagoon, bay and marine environments of world oceans reported in literature (Accornero et al., 2008), and can consequently be classified as an uncontaminated system as Vaccares and Leucate Mediterranean Lagoon in the Gulf of Lion (France) (RNO, 1998; Accornero et al., 2008). Thus, the MOB is a highly dynamic system with significant spatial and temporal metal distribution variations. The sampling strategy in such environment is a key factor to provide a comprehensible overview of the ecosystem state. To assess the sediment quality, the influence of grain size distribution on metal concentrations (Horowitz et al., 1991; Loring, 1991) needs to be removed by using the enrichment factor parameter.

4.2. Enrichment factor

The degree of metal enrichment in sediments was assessed using the enrichment factor (EF) which allow to differentiate natural geochemical background to anthropogenic inputs (Zhang and Liu, 2002). Previously, trace metal concentrations in sediments were normalized to compensate natural variability due to grain-size variations (e.g. mineral composition) and to detect any anthropogenic metal contributions e.g. (Loring, 1991). Grain size dependent effect was corrected using thorium (Th) concentrations. In fact, previous work showed that Th can be used as a conservative lithogenic element (Braun et al., 1993; Krachler and Shotyk, 2004; Coynel et al., 2007; Kipp et al., 2009), especially in the Gironde Estuary watershed (Masson et al., 2006; Coynel et al., 2007; Larrose et al., 2010), because of its low solubility (e.g. van Calsteren and Thomas, 2006) and its limited reactivity (Martínez-Aguirre et al., 1995). Then, EF was defined as the ratio of Th normalized metal concentrations in sediments over Th normalized ratio in a geochemical background reference (Feng et al., 2004). The background reference concentrations correspond to the concentrations measured at the bottom of a sediment core from the West Gironde Mud Patch (Larrose et al., 2010), located on the Aquitaine Plateau, seaward of the Gironde estuary mouth (Lesueur et al., 2001), and considered as the regional geochemical baseline for sediments derived from the Gironde Estuary (Larrose et al., 2010). Thus, a value of EF between 0.5 and 1.5 suggests natural weathering processes (Zhang and Liu, 2002). In contrary, $EF > 1.5$ values suggest enrichment from anthropogenic source. More precisely, the $1.5 < EF < 3$ interval indicates a minor enrichment, the $3 < EF < 5$ one for a moderate enrichment, the $5 < EF < 10$ one for a moderately severe enrichment until $EF > 50$ values which indicates an extremely severe enrichment (Essien et al., 2009). Enrichment factor determined in sediments from Les Doux, Brouage and Perquis were reported in Figure 5b. Three tendencies were observed (i) elements as Cu, As, Ag and Pb which are in the geochemical background in the East, West and South part of the bay (Figure 5b B-D-E-G) (ii) elements as Ni and Zn which are in the geochemical background in the East and West of the Bay and which present temporal minor enrichment in the south (Figure 5b A-C) and (iii) Cd which is in the geochemical background in the East, which present minor enrichment in the West and minor to temporally moderate severe enrichment in the south of the Bay (Figure 5bF). Thus, metal enrichment factors showed that metal sediment quality strongly spatially differed. Thus, to better understand metal distribution in the Bay, we will focus on Cd because of its variation and its high pollutant state in the Bay (Cd repercussion on oysters" production) and we will examine its temporal distribution coupled to hydrodynamic forcing for each site.

4.3. Hydrodynamics

The Marennes Oléron Bay showed spatial and temporal strong hydrodynamic and sedimentation variations. In the western part, sediment interface was rather sandy and was characterized by pluri-centimeter erosion and accretion events (Figure 4a). These sedimentary movements depend of wave's characteristics (wave's height, period and direction). The area was under strong oceanic influence as evidenced measured salinity (34 Figure 2a situation C). However, during particular periods, less salty water coming from the Gironde was directly measured (Figure 2a situation B) and were simulated after this records, using PREVIMER hydrodynamic model (IFREMER) based on daily measured salinity. These observations are in agreement with previous studies on the Gironde influence on MOB sediment via the Antioche Strait (Pouliquen, 1975; Castaing and Allen, 1981; Boutier et al., 1989; Froidefond et al., 1998; Parra et al., 1999) and with Cd minor enrichment estimated on Les Doux sediments. In fact, during the transit Gironde-Antioche-MOB, Cd-enriched SPM originated from the Gironde Estuary desorbed in marine coastal waters (Boutier et al., 2000; Masson, 2007; Dabrin, 2009, Strady et al, in revision) and deposited as low Cd-concentrated SPM in the western MOB (Figure 5c)

In contrast, the eastern MOB mudflat was largely influenced by the Charente River Estuary (Tesson, 1973; Pouliquen, 1975). Salinity, turbidity and bed elevation variations (Figure 4;7b) differed from the western part showing disconnection of hydrodynamic and sedimentary parameters. During the survey, bed elevation was closely related to Charente water discharges and wave activity (Figure 6bcd), as we observed erosion during high and low water discharges from April 07 to October 07 and repeated short wave action and accretion during mean water discharges and decreasing wave activity from December 07 to April 08. Accordingly, sediments sampled during accretion presented high water content (Figure 3) and were freshly deposited (called „mollin“) whereas during erosion, sampled sediments presented lower water content (Figure 3) and were dated of 50 to 100 years, as shown by ^{210}Pb datation (Kervella, 2010). During flood, expulsion of maximum Charente River turbidity caused deposition a few days later in the mudflat (Figure 6be), as observed in the Seine Estuary (Deloffre et al., 2005). However, Cd concentrations and enrichment were constant over time and were not related to the age of sediments and to Charente River Estuary water discharges. Thus, inputs of trace metal to Brouage mudflat were constant over the survey, meaning constant trace metal inputs from the Charente River, whatever the water discharges. Concerning the older sediment, low trace metal concentrations can be explained by sediment

desorption during regular outcrop since a century and probably already not metal-enriched SPM flowing out the Charente Estuary.

The southern part of the bay showed different hydrodynamic and sedimentation processes (Figure 4c). This area was very quiet with low waves amplitude, and did not exhibit close relation with bed elevation variations, except during major storm. During the year, the area was in net accretion despite multi-centimetre sequences of erosion/deposition observed in winter (Figure 6bcf). From December 28, 2007, oyster installations were removed and could have induced development of current ripples as they did not act as hydrodynamic barriers anymore. Before this event, sandy bed-elevation variations were not related to the Charente and Gironde water discharges or to tidal cycles (Figure 6ab). However, we observed that the outside waves were much higher (mean H_s : 3 m; Figure 6c) during winter than during summer time (mean H_s : 1.8 m; Figure 6c), which could have induced sand resuspension and transport from outside dunes to the MOB via the Maumusson Strait. Cadmium enrichments in sediments from Perquis were the most important of the MOB. Enrichments temporally varied but were not related to temporal bed elevation variations (Figure 5 and 3). Because of the north to south residual circulation, the Charente origins of enriched sediments could be considered. However, Cd enrichments in the south were significantly higher than in Brouage and Les Doux, refuting their possible origins from the northern Bay. Thus, we hypothesized Cd sediment enrichment by deposition of high Cd concentrated SPM transported by the Gironde Estuary via the Maumusson Strait. In fact, the Gironde turbid plume can extend northly to the coast during high water discharges and flood situations combined to S to SE winds (Froidefond et al., 1998; Hermida et al., 1998). The connection of the Gironde turbid plume with the Maumusson Strait was estimated from a statistical study of satellite images over four years at about 30% of time per year (Dabrin, 2009; Lafon et al., 2009). Thus, SPM from the Gironde plume can enter the Bay through Maumusson Strait during those specific conditions (Dabrin, 2009, Strady et al., in revision Chemosphere) and flooding tide (Bertin et al., 2005). This is consistent with moderately severe enriched Cd observed in January 08 (Figure 5) just after the Gironde flood situation (Figure 6b).

Bed elevation, wave activity and grain size distribution surveys at the three sites were highly relevant to characterise sediment dynamic and showed hydrodynamic forcing plays an important role on sediment bed elevation and erosion/sedimentation phases and influence trace metal distribution. In fact, sediments were in constant good quality state in the eastern part and were influence neither by the Charente river water discharges or the wave activities. In contrast, sediments from the western parts were strongly related to waves activities

whereas metal concentrations were more related to water masses inputs, especially the Gironde water influence. Finally, the southern part presented different hydrodynamic and enrichment patterns as sediment quality was punctually moderately severe enriched in Cd in relation with Gironde water discharges. To resume, a conceptual outline is presented in Figure 7 and represents the Gironde and Charente influences on metal distribution and the hydrodynamic influences via the Gironde and Charente water discharges and wave propagation on sediment dynamic in the Bay.

Globally, this pluri-disciplinary approach allows pointing out the temporal variability of sediment characteristics and their trace metal contents. In most coastal marine environments, trace metal concentrations in sediments varied spatially and temporally in an annual scale (e.g. Essien et al., 2009; Corbett et al., 2009, Valdès et al., 2009). However, in areas highly influenced by tides, currents and water discharges, we rather think that temporal variability, even on a small day scale, are important to understand the dynamic of the system. The approach described in this paper can thus be broaden to dynamic environments in order to (i) point out and characterise temporal changes and (ii) determine the factors which are controlling the temporal and spatial variability of sediments and their trace metal concentrations.

5. Conclusion

This original pluri-disciplinary study pointed out both spatial and temporal trace metal concentrations in sediments of the Marennes Oléron Bay, the sediment quality and the role of regional hydrodynamics forcing on metal distribution in sediments. The MOB can be divided into three areas, western, eastern and southern, disconnected to each other, which are characterized by specific hydrodynamic forcing and trace metal distribution. Globally, sediments were little impacted by trace metals which were in the geochemical background state and so in good quality state. However, the southern Bay was the most concerned by metal enrichment. Temporal distribution variations have shown the influence of the Gironde plume via the Maumusson Strait on Cd enrichment, which was under-estimated until now. To conclude, this study has shown the relevance of an adapted sampling strategy and multi-disciplinary approach to assess metal sediment quality and later implement an ecosystem management.

6. Acknowledgements

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Figure Captions

Figures 1: a) localisation of the Marennes Oléron Bay in southwestern France, b) zoom on the Marennes Oléron Bay and the location of three sampling sites.

Figure 2: Variability of a) salinity and b) turbidity on Brouage mudflat, the Doux and Agnas bank, from 04/06/07 to 04/24/07

Figure 3: Sediments fractions and water contents measured in sediments from Les Doux, Brouage and Perquis sites during the year survey

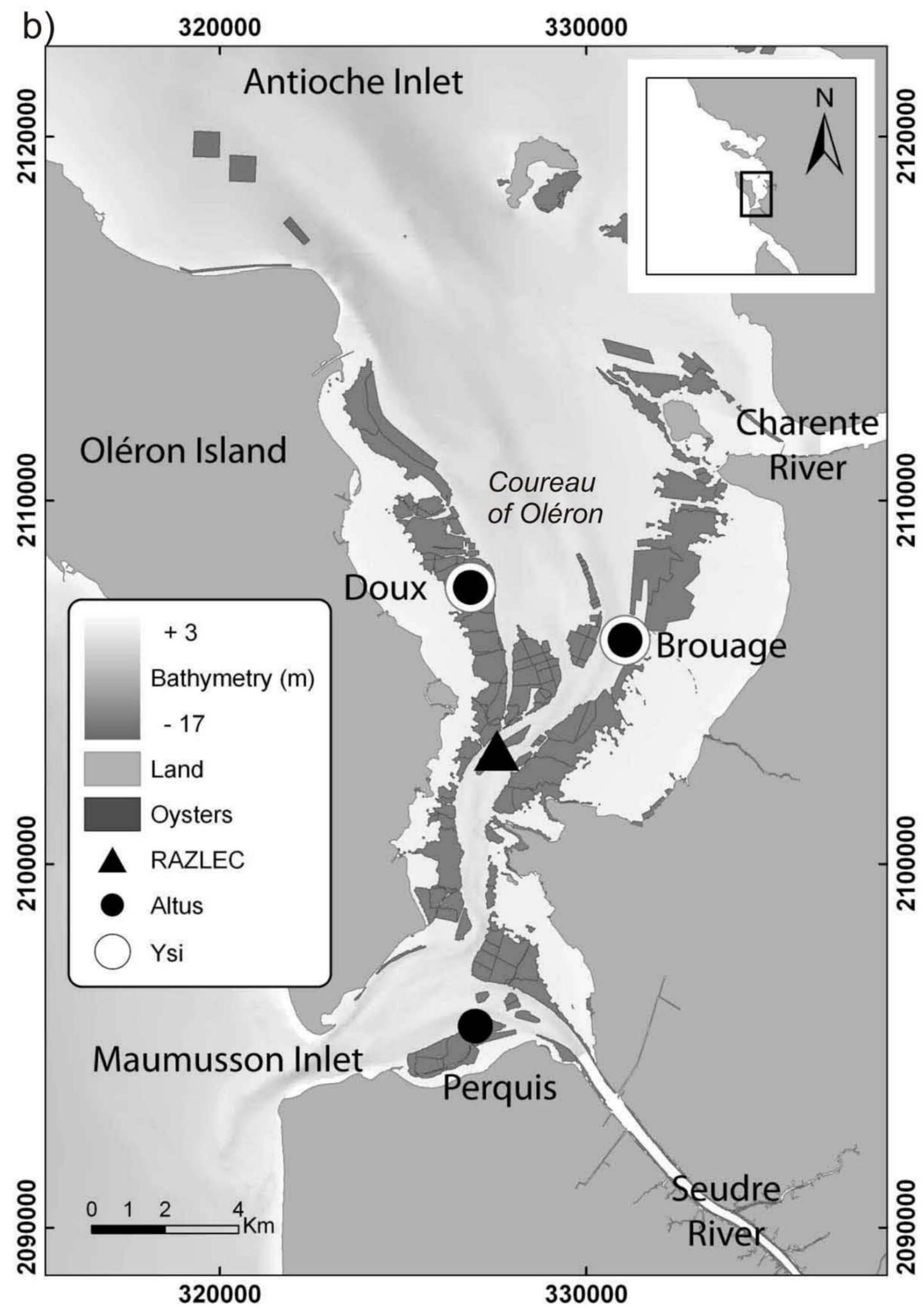
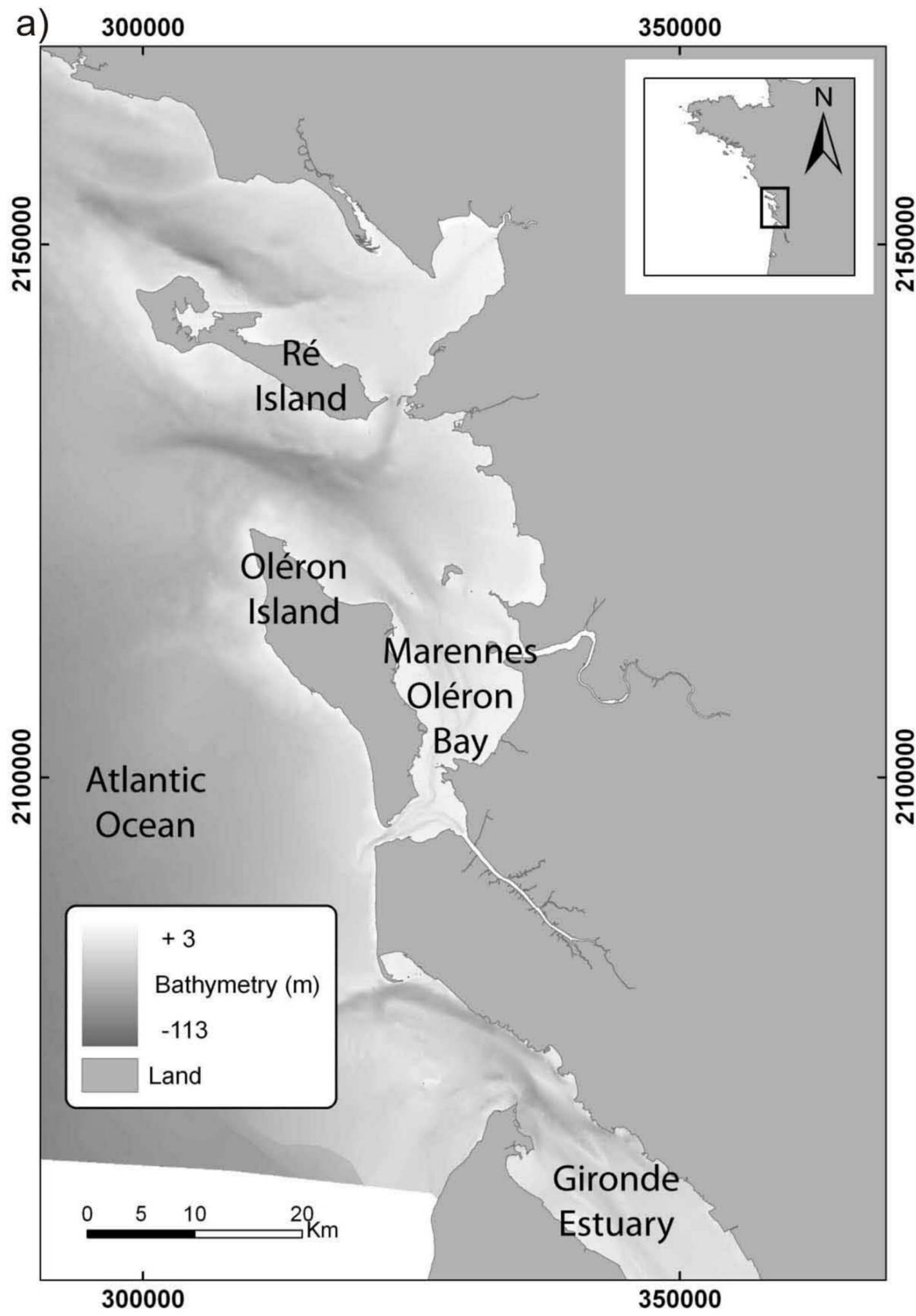
Figure 4: Variability of hydrodynamic (water level and wave height) and sedimentary (bed elevation) parameters during one year above a) the Doux, b) Brouage mudflat and c) Perquis bank

Figure 5: a) Particulate concentrations and b) enrichment factors of Ni (A), Cu (B), Zn (C) As (D), Ag (E), Cd (F), Pb (G), Hg (H) and (Th) in sediments from Les doux, Brouage and Perquis. To notice, enrichment factors data are not available for Hg and Th.

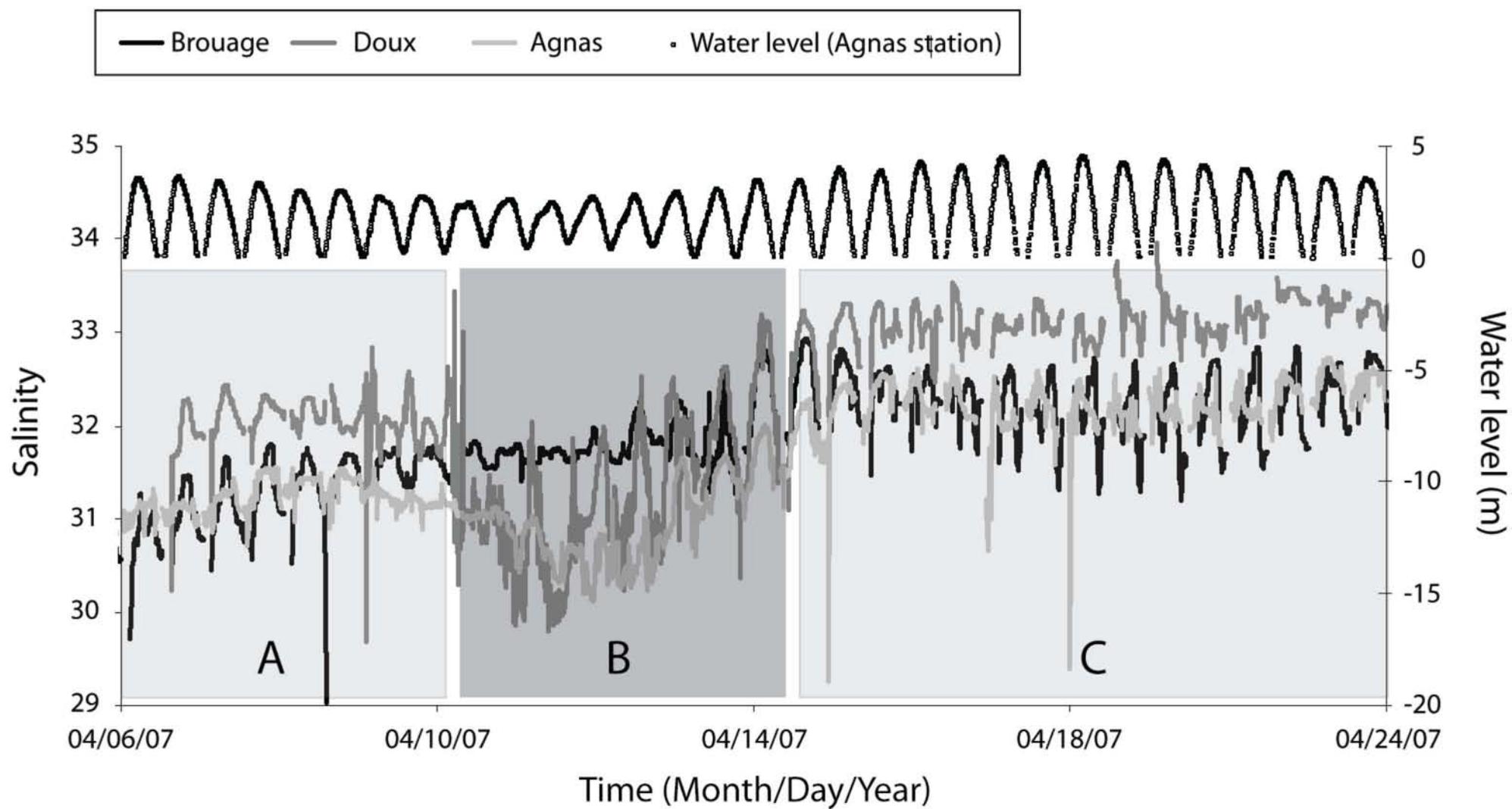
Figure 6: Relation between a) the water level, b) the Gironde and Charente water discharge and c) wave height measured in front of the Oléron island with the variability of the bed elevation on d) Brouage mudflat and e) Perquis bank, during one year

Figure 7: Conceptual outline of the influence of the Gironde Estuary, Charente Estuary and wave action on spatial and temporal variability of sediment transport and Cd concentrations in surface sediments. The thickness of the arrows is relatively proportional to the Cd contribution to the Marennes Oléron Bay.

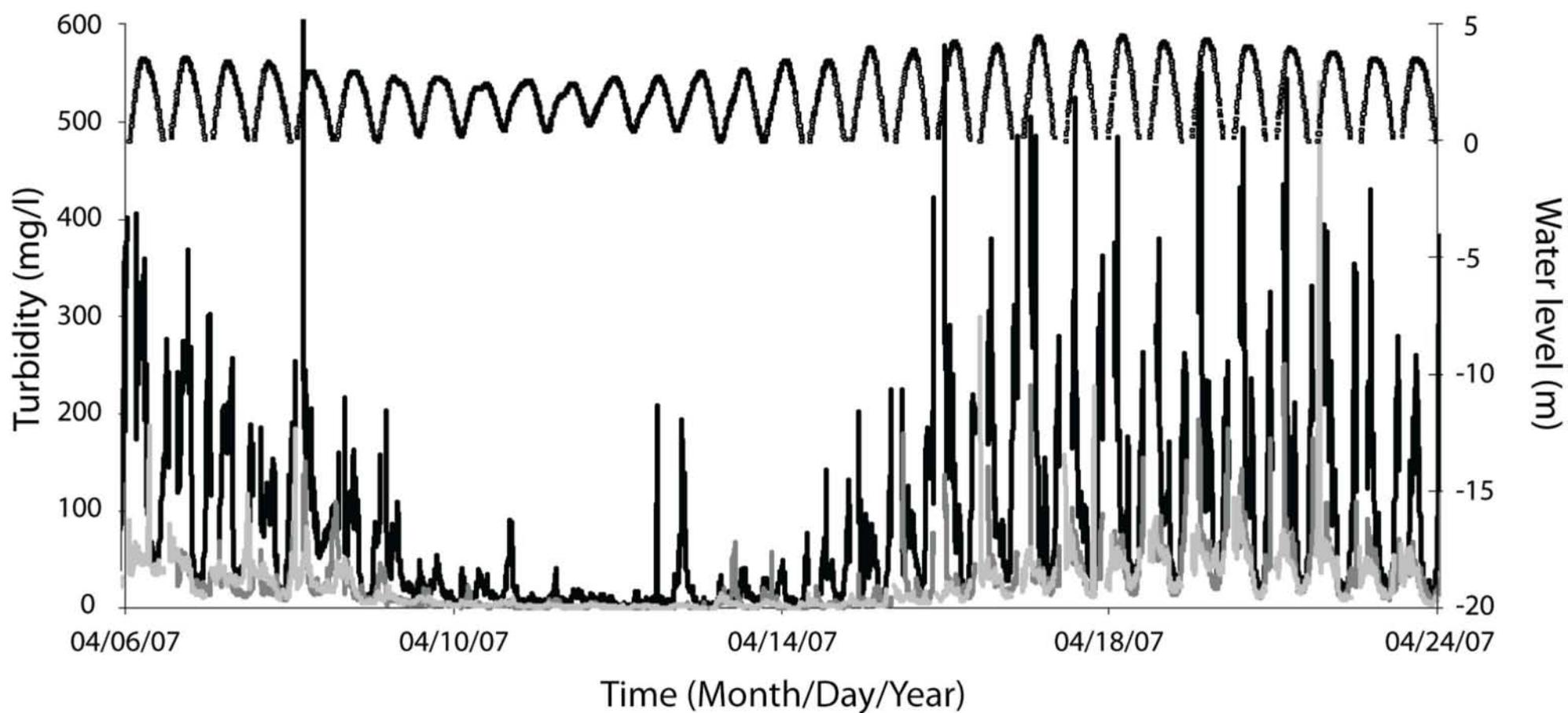
Table 1: Concentrations of certified reference material, BCSS-1 and IAEA-405 for Hg.



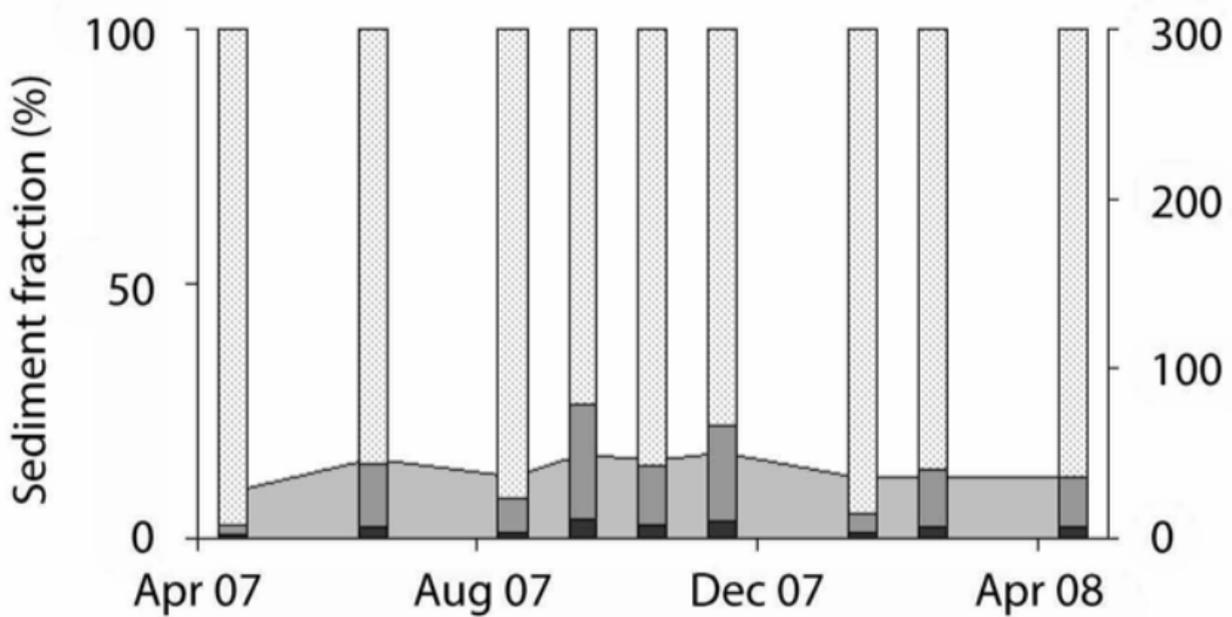
a)



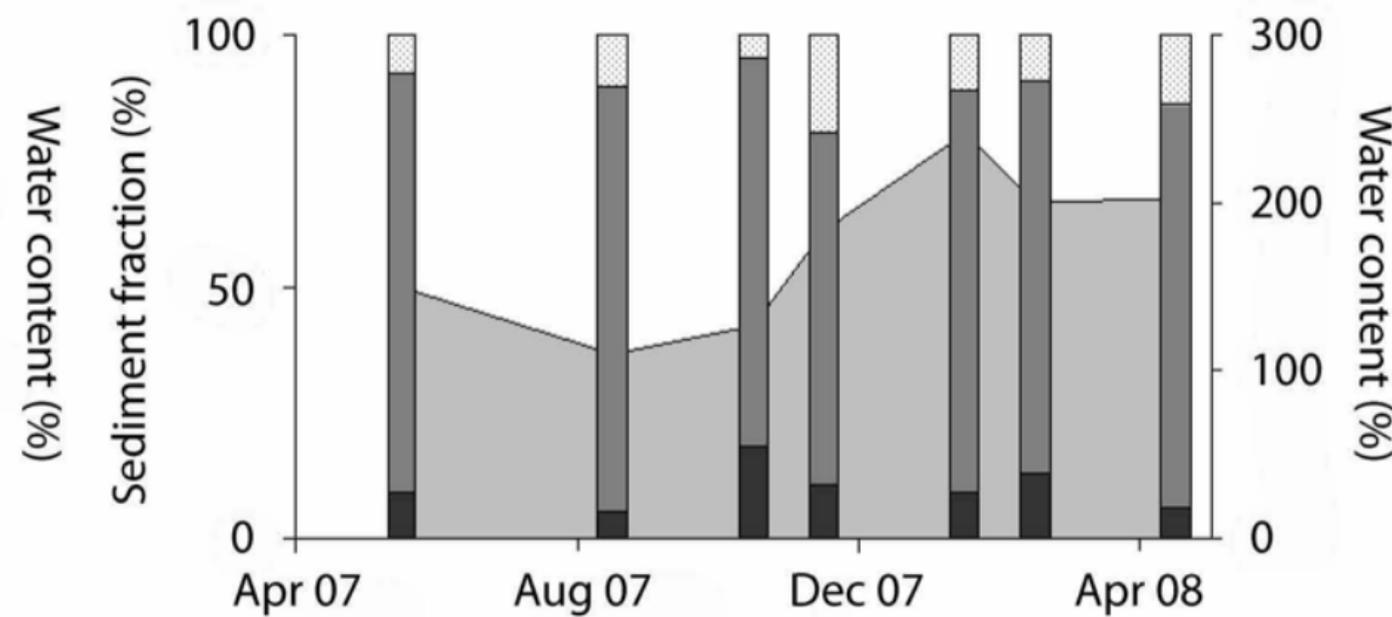
b)



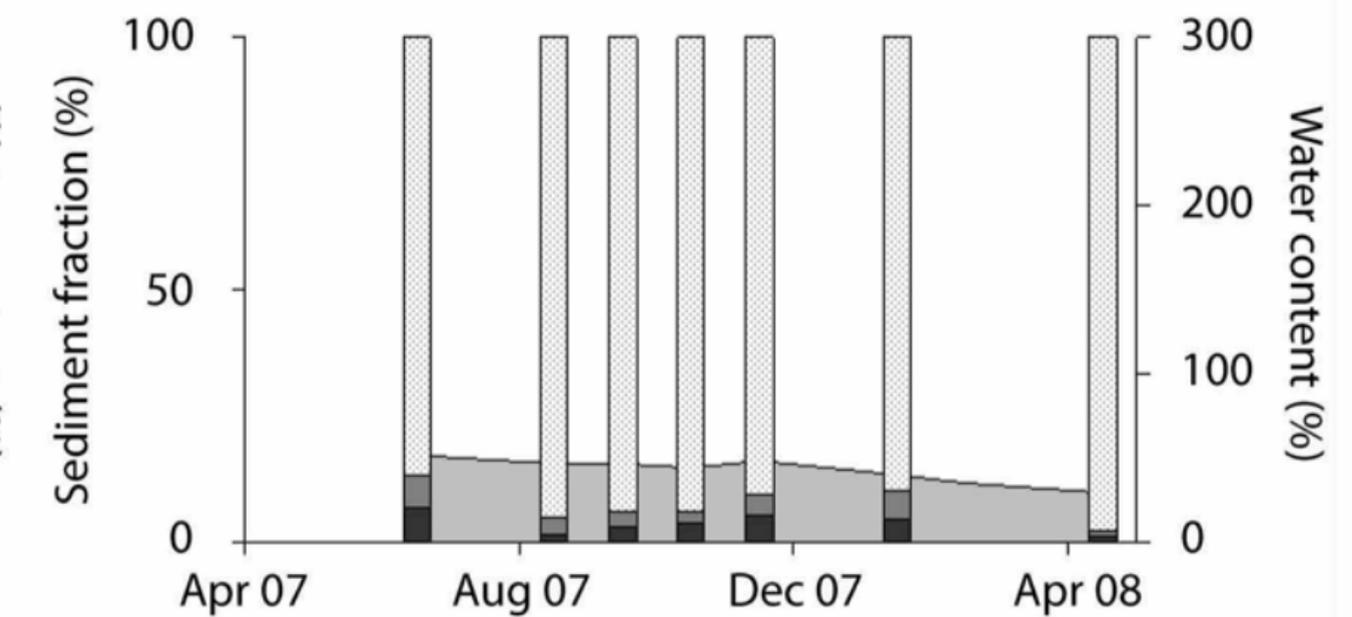
Doux site



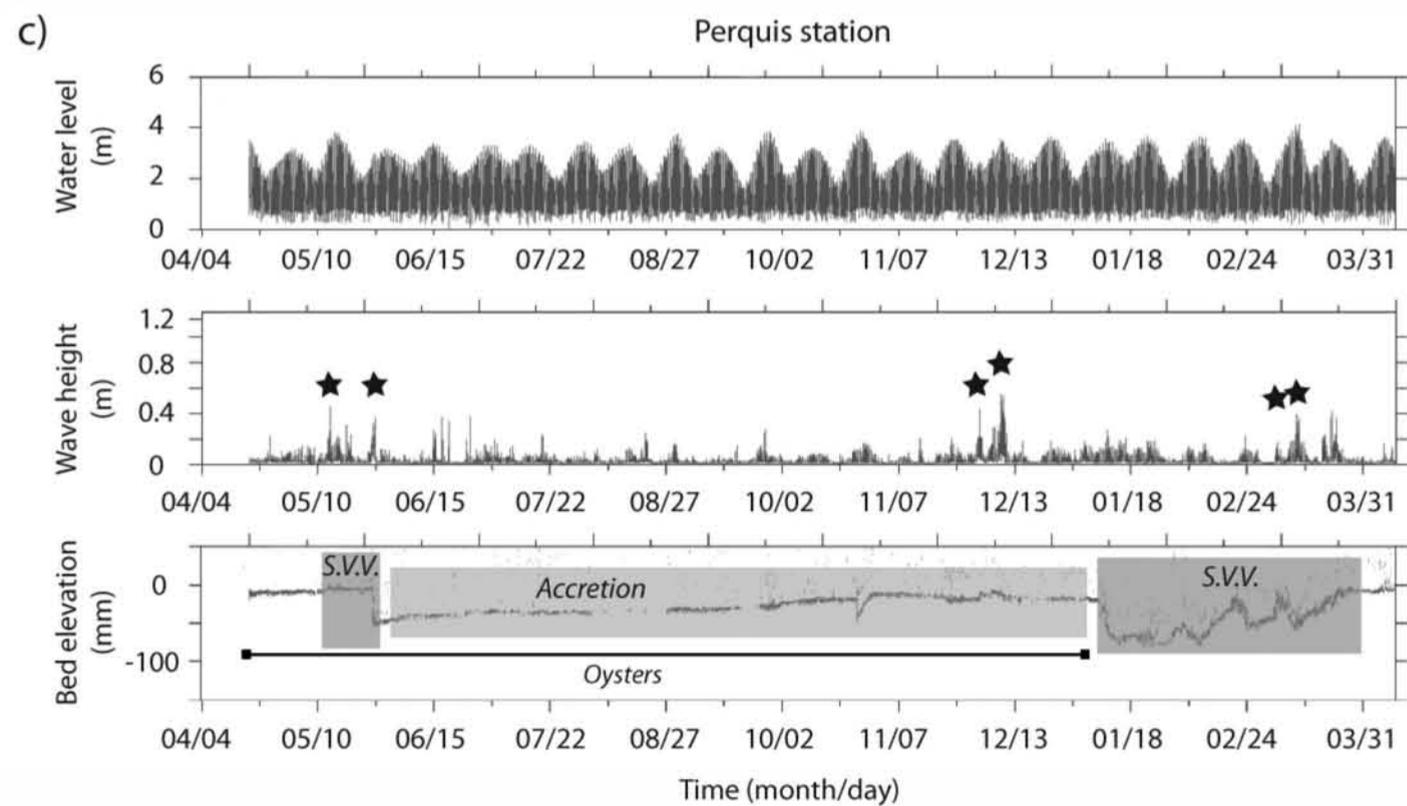
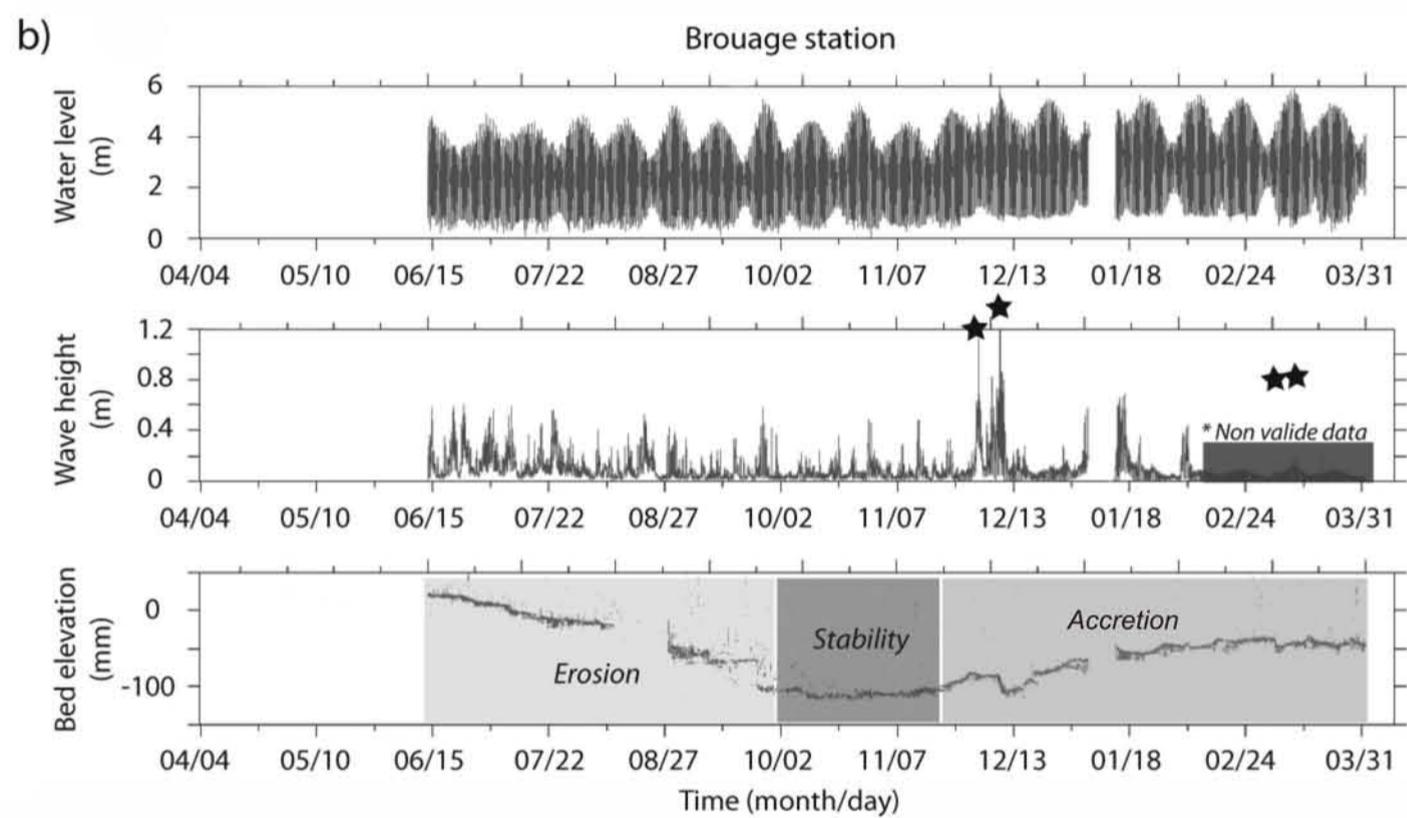
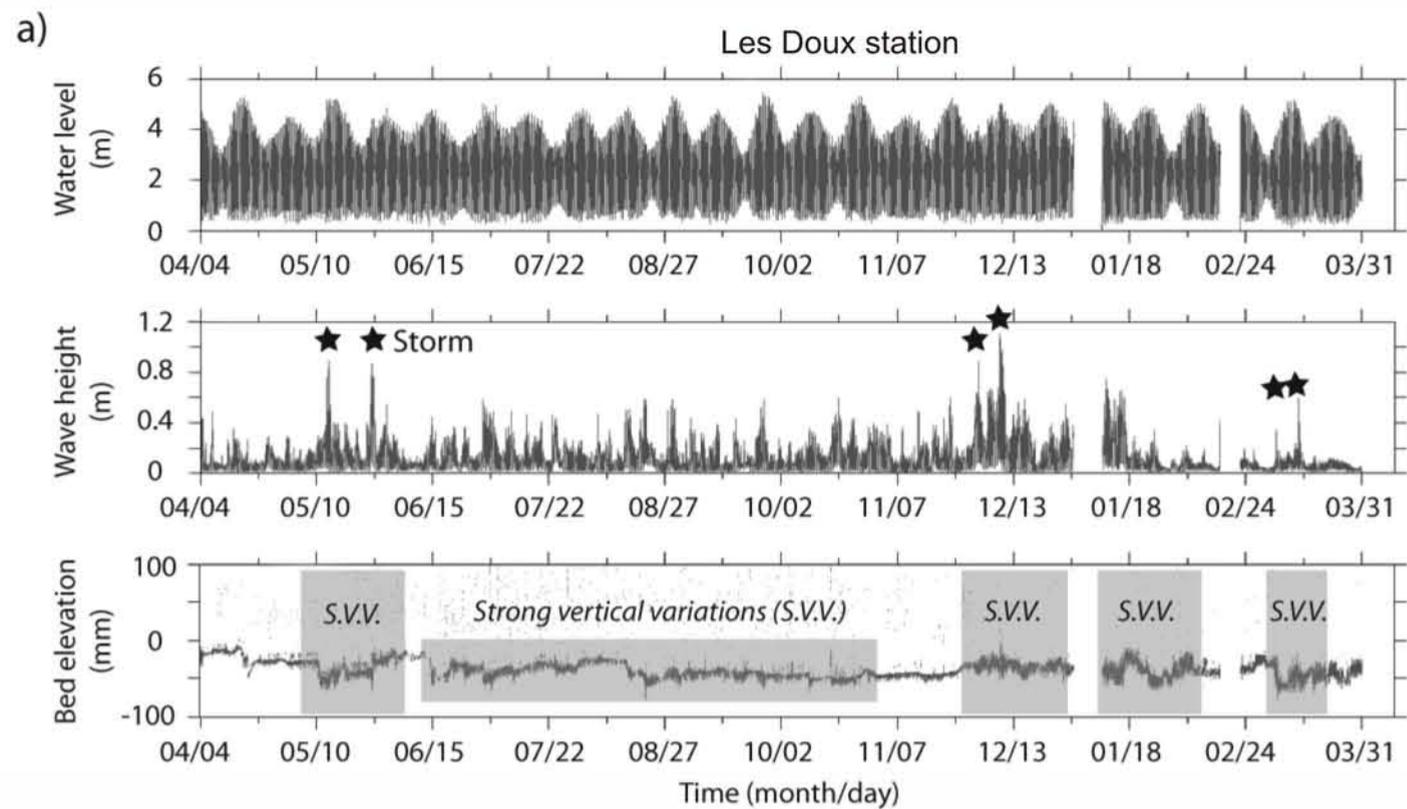
Brouage site

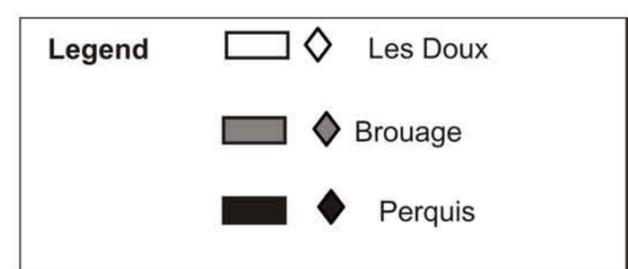
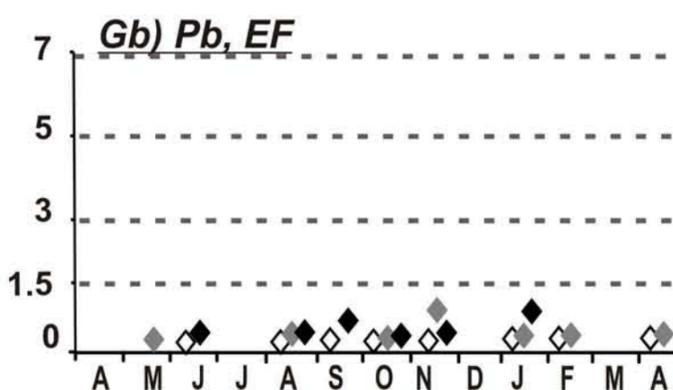
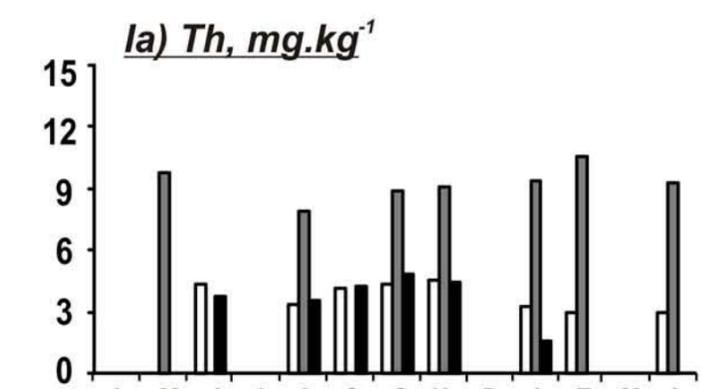
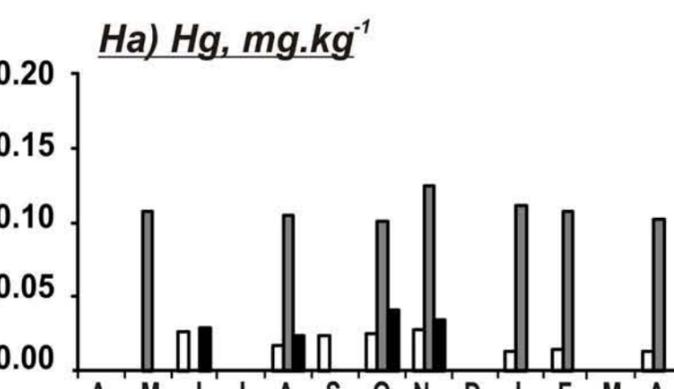
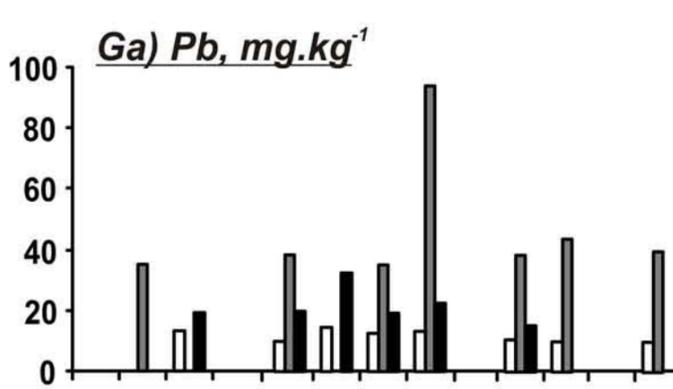
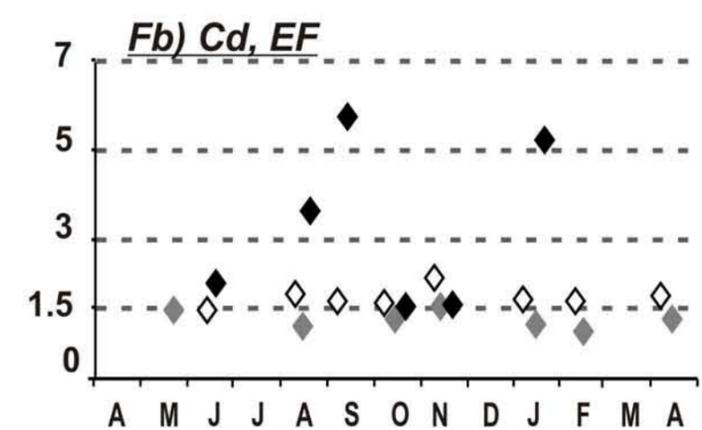
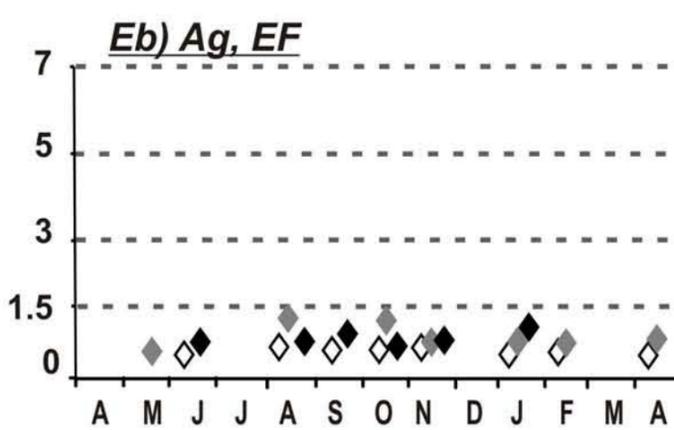
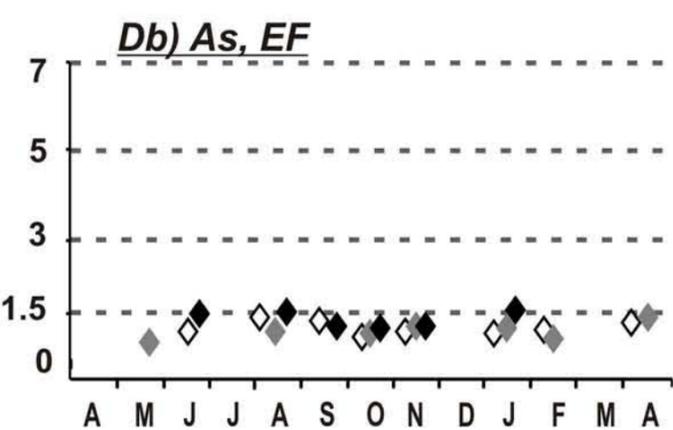
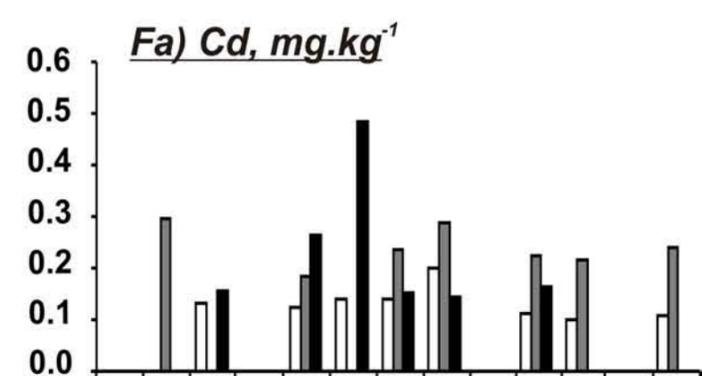
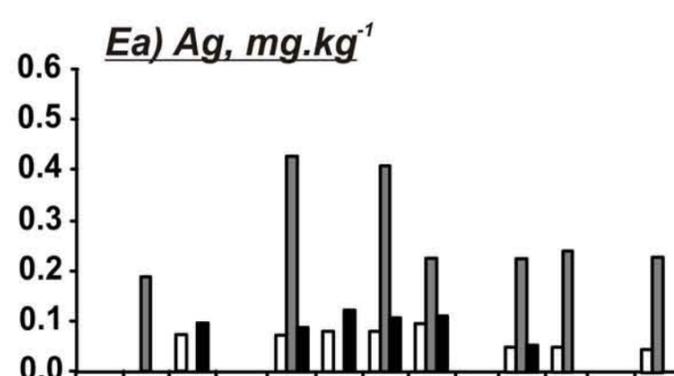
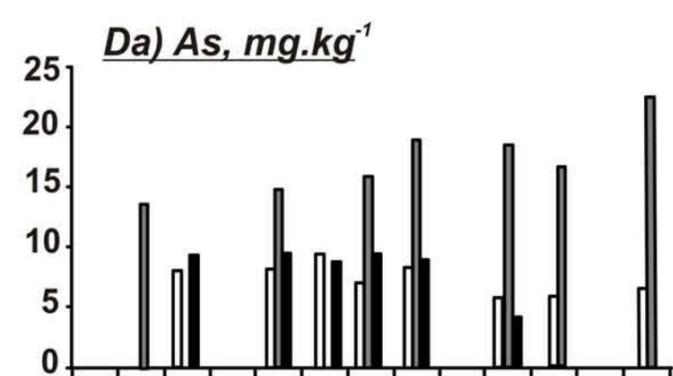
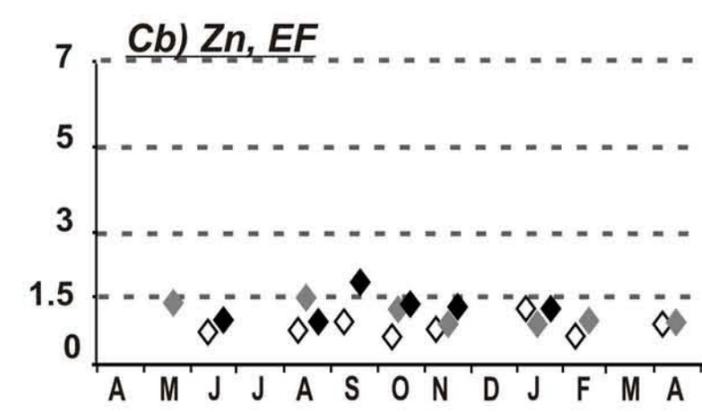
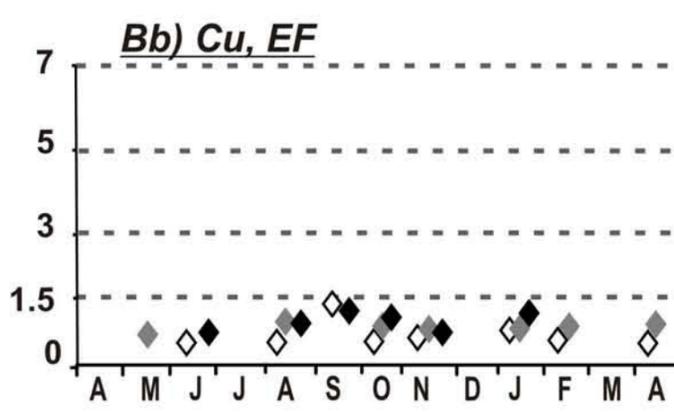
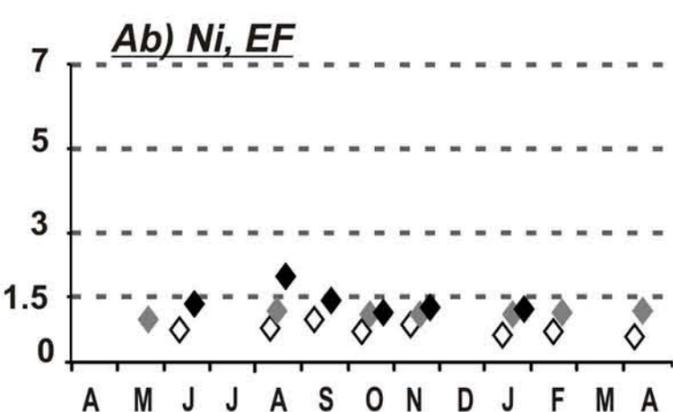
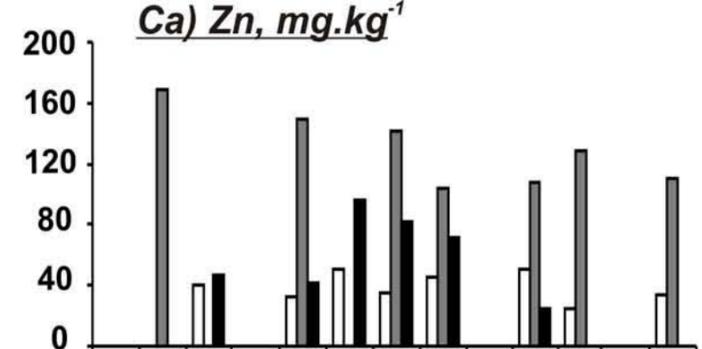
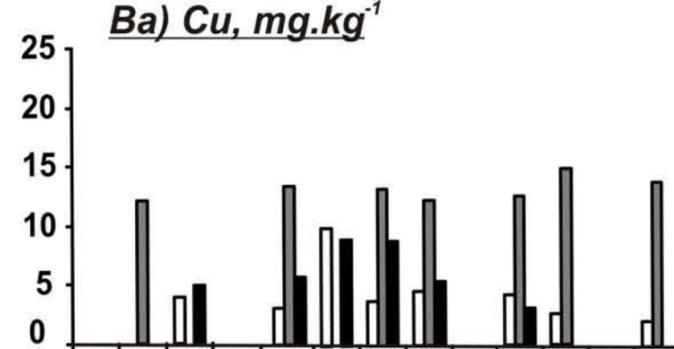
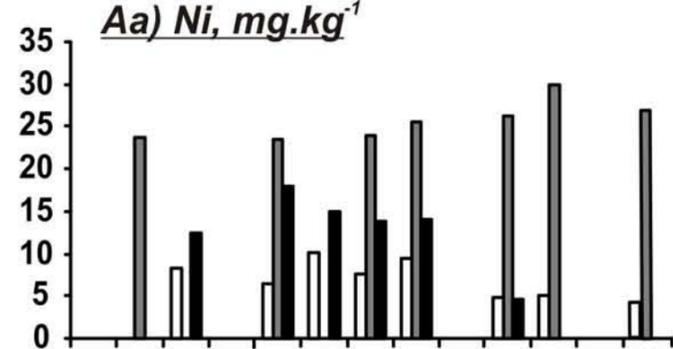


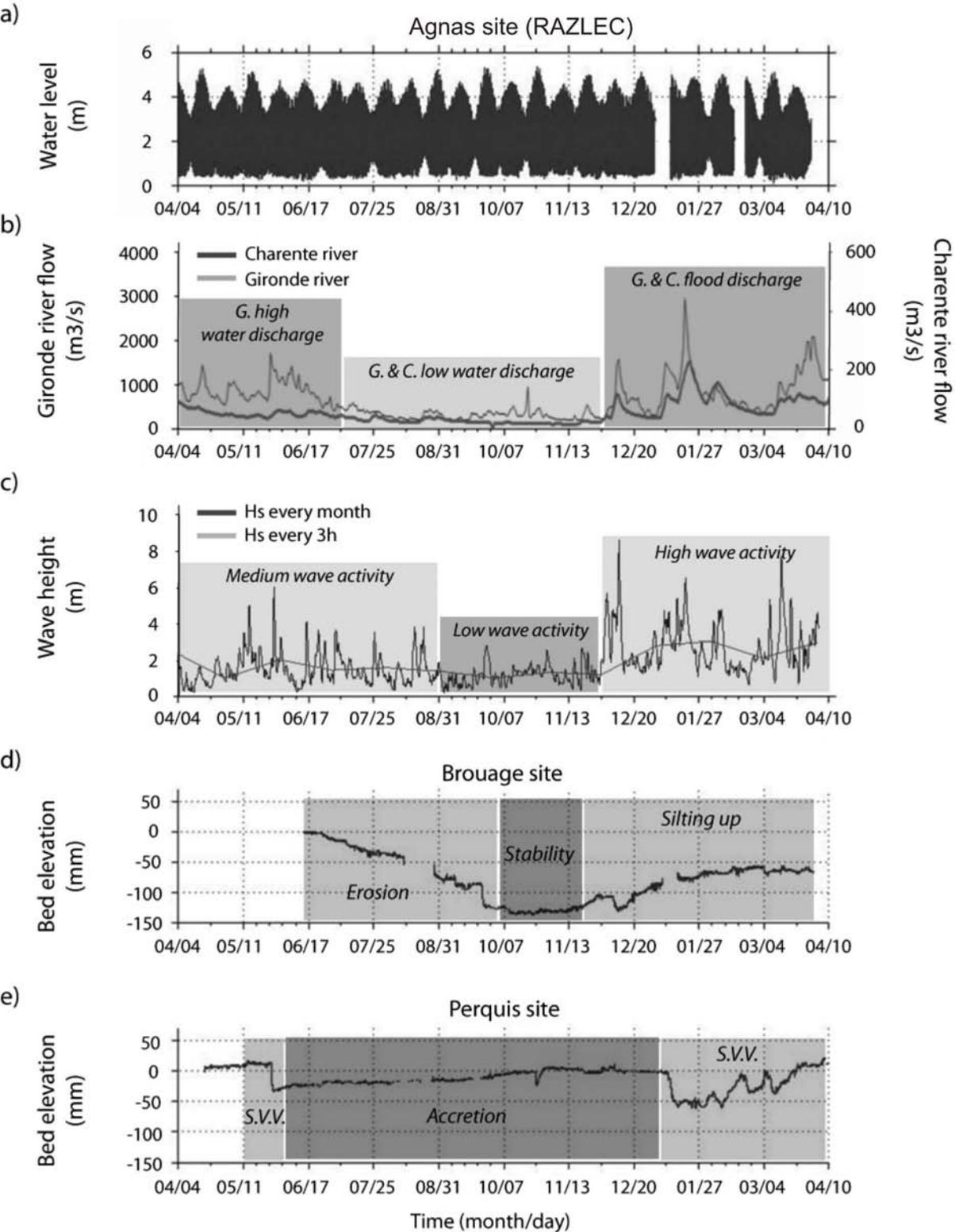
Perquis site

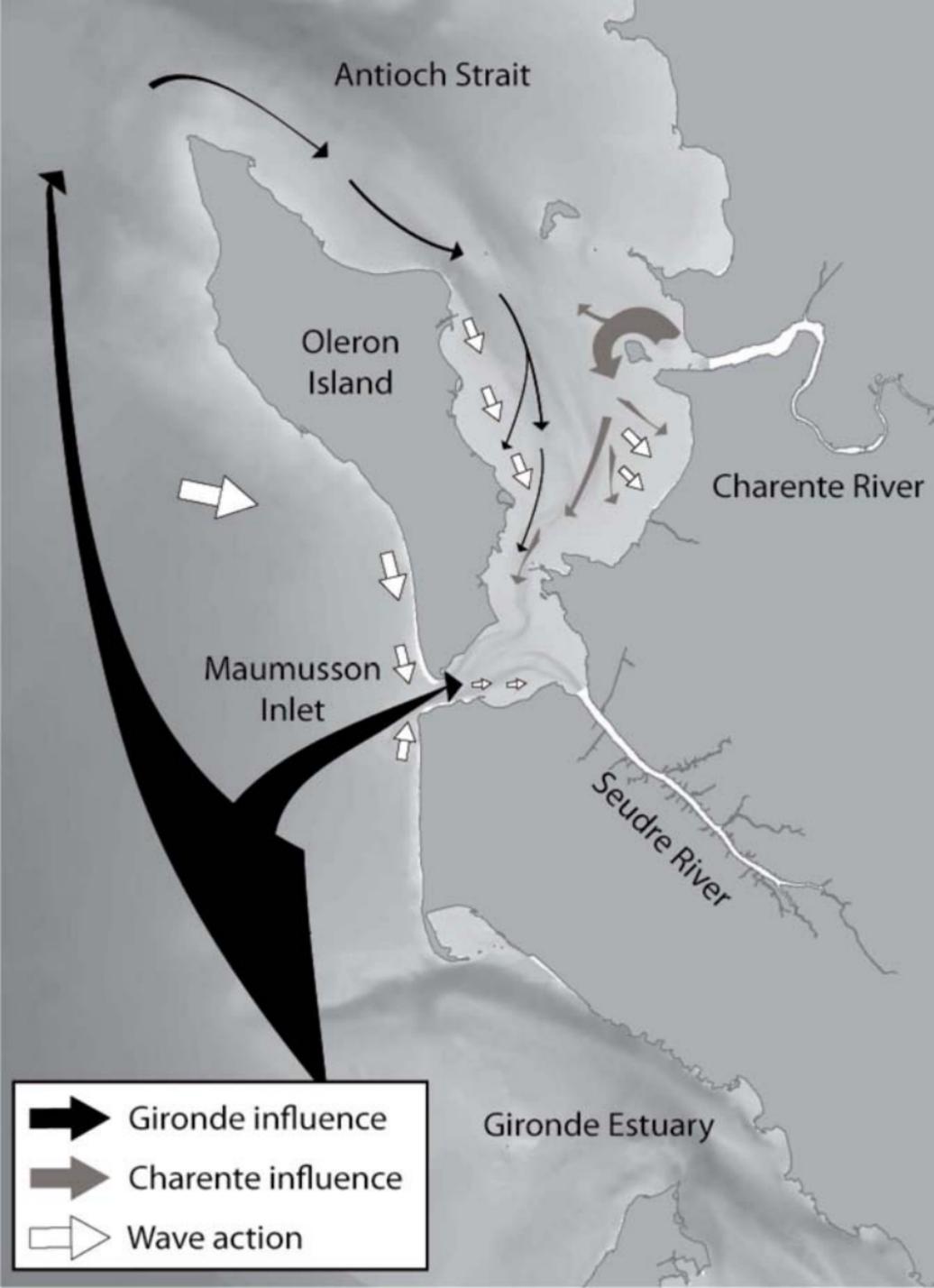


Water content
 Clay
 Silt
 Sand









	Ni	Cu	Zn	As	Ag	Cd	Pb	Th		Hg
BCSS-1	55.3 ± 3.6	18.5 ± 2.7	119 ± 12	11.1 ± 1.4		0.25 ± 0.04	22.7 ± 3.4		IAEA-405	0.81 ± 0.04
mg.kg⁻¹	56.5 ± 5.5	20.0 ± 6.1	92.6 ± 3.5	10.4 ± 0.6	0.18 ± 0.00	0.32 ± 0.01	22.3 ± 0.6	6.9 ± 0.07	mg.kg⁻¹	0.80 ± 0.06