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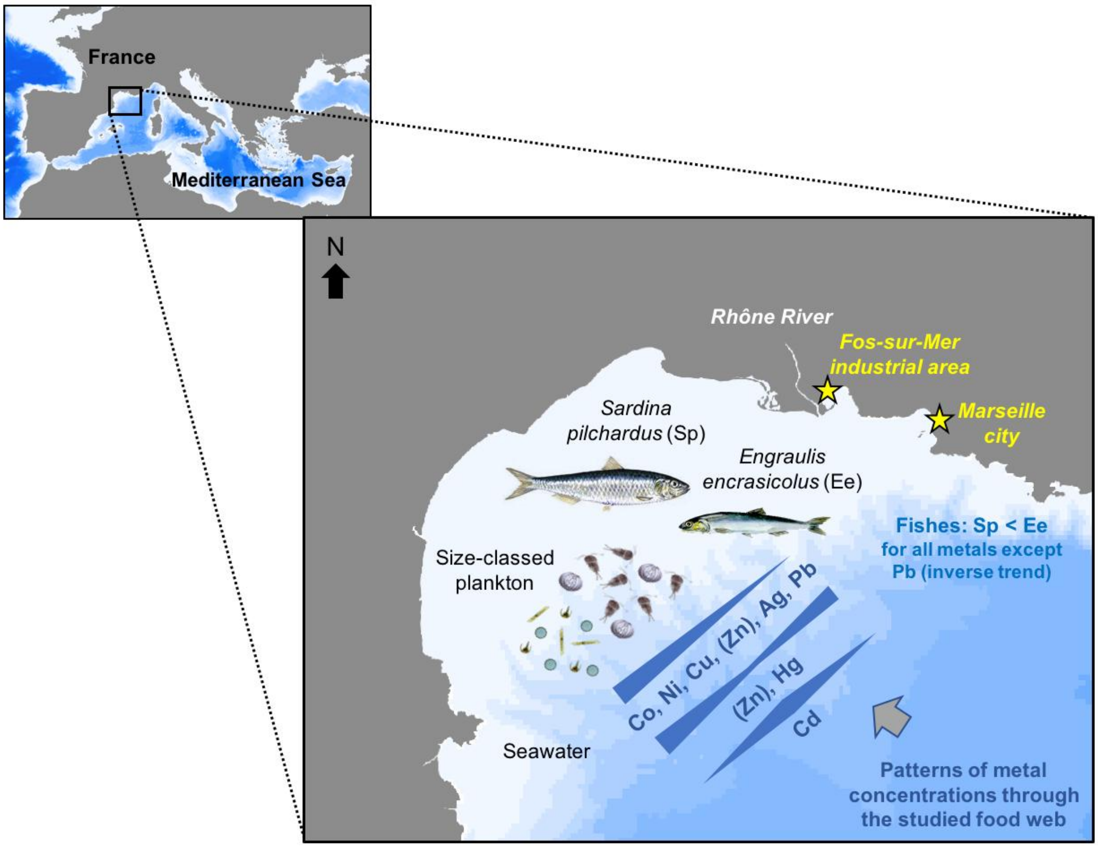
Abstract:
Trace metal contamination in the European sardine and anchovy food web was investigated in the Gulf of Lions, NW Mediterranean Sea, including seawater and size fractions of plankton. The results highlighted: i) higher and more variable concentrations in the smaller plankton size classes for all metals except cadmium; ii) higher concentrations in anchovy versus sardine for all elements except lead; iii) different patterns of metal bioaccumulation through the food web: cobalt, nickel, copper, silver, lead and zinc displayed continuously decreasing concentrations (with the exception of increased zinc in fish only), while mercury concentrations dropped considerably in larger plankton size classes and rose significantly in fish. Lastly, cadmium concentrations were found to be highest in intermediate plankton size classes, with very low levels in fish. The need to efficiently characterize the biological composition of plankton in order to fully identify its role in the mobilization and transfer of metals was highlighted.

Keywords: inorganic elements; seawater; size-classed plankton; *Sardina pilchardus*; *Engraulis encrasicolus*; Mediterranean Sea

Highlights:

- Higher concentrations were generally found in water samples from western stations
- Few or no significant spatial variations were tested or highlighted for biota
- Contrasted bioaccumulation patterns of trace metals along the food web were found
- Concentrations differed greatly among size fractions of plankton
- Anchovy presented higher concentrations than sardine for all metals except Pb

Graphical abstract:



1. Introduction

Trace metals are released into the environment from both natural sources (e.g. volcanism, soil erosion and forest fires) and anthropogenic sources (e.g. transport, harbours, industrial activities and major coastal cities). They reach the ocean through riverine and aeolian fluxes (Mason, 2013). Their increased use in human activities has led to the contamination of numerous environmental compartments and, as a result, to environmental levels with a detectable anthropic contribution (Lewis and Maslin, 2015). Some trace metals have essential biological functions within a narrow range of optimal concentrations (essential elements), while others have no known biological role (non-essential elements) and are recognized for their toxic effects on aquatic organisms, even at environmental concentrations (Mason, 2013).

Taxa- and species-specific metal regulation mechanisms (i.e. uptake, storage and/or elimination) have been described for both essential and non-essential elements (Wang and Rainbow, 2010). Their transfer between biogeochemical compartments, bioaccumulation in organisms and biomagnification in food webs depends on their concentrations and speciation in both abiotic (habitat) and biotic (food sources) environments (Neff, 2002; Rainbow, 2002). Marine organisms are hence exposed and accumulate contaminants via dissolved and trophic pathways; the latter being the main route for trace metal intake by medium to high trophic level consumers such as fish (Mathews and Fisher, 2009; Pouil et al., 2016). Understanding the mechanisms that lead to the bioaccumulation of trace metals in consumers and the interpretation of their metal burden thus requires good knowledge of feeding habits and trophic ecology, as well as metal levels in diets.

In the Gulf of Lions, in the northwestern (NW) Mediterranean Sea, small pelagic planktivorous fish such as European sardine (*Sardina pilchardus*) and anchovy (*Engraulis encrasicolus*) are fishery resources of major economic importance (Palomera et al., 2007). Both species play a major ecological role in food web functioning (Bănaru et al., 2013) by transferring energy and nutrients from lower trophic levels (plankton) to upper levels (large pelagic fish, marine mammals and seabirds). In a broader context of maintaining the functional integrity of ecosystems and associated ecosystem services, thorough knowledge of global anthropogenic impacts on this pelagic compartment, including contamination pressures, appears crucial. The Mediterranean Sea is notoriously faced with various pollution threats, including chemical contamination (Danovaro, 2003; Durrieu de Madron et al., 2011). Moreover, the UNEP recently highlighted the lack of data on pollutant impacts on Mediterranean marine ecosystems (UNEP/MAP, 2012). This data may be of particular importance with regards to the Mediterranean Sea, where contaminant levels observed in predatory species are significantly higher than in the Atlantic Ocean (Bodiguel et al., 2009; Cossa and Coquery, 2005). Although this difference may be explained by higher contaminant concentrations in abiotic compartments, it may also be due to the enhanced ability of Mediterranean pelagic food chains to bioaccumulate certain chemical elements or substances, as documented for mercury (Chouvelon et al., 2018; Cossa and Coquery, 2005;

Harmelin-Vivien et al., 2009). Therefore, contamination in the planktonic compartment must also be studied in order to properly assess contamination pressures on small pelagic fish.

Plankton and, in particular, phytoplankton, forms the first link between abiotic (seawater) contamination and pelagic fish, hence playing a major role in contaminant transfer into marine food webs. The contamination dynamics of plankton must therefore be assessed in order to properly apprehend contaminant bioaccumulation at the secondary trophic levels of small pelagic fish. This issue remains a challenge and has been relatively poorly-investigated *in-situ*, probably due to the difficulties in sampling representative fractions of plankton and obtaining sufficient material to perform trace level chemical analyses. Despite an abundance of studies on the transfer of metals in upper trophic levels, none address the problem in its entirety from the water column to small pelagic fish and most consider a limited number of fish organs (muscle, liver and sometimes gonads or gills). This failure to address wide-ranging food web compartments and analyse contaminants in certain organs/tissues (with tissue-specific bioaccumulation properties) may constitute a considerable bias with regards to bioconcentration/bioaccumulation calculations, for which trophic levels must be considered in their entirety (Gray, 2002; Wang, 2002). As a result, although the biomagnification of mercury in aquatic ecosystems is undisputed, zinc is sometimes thought to bioaccumulate in fish food chains (Mathews and Fisher, 2008; Wang, 2002), while conclusions regarding cadmium, lead and silver differ (Cheung and Wang, 2008; Luoma and Rainbow, 2005; Reinfelder et al., 1998).

In this general context, the specific objectives of our study were to: (i) characterize the trace metal burden of the plankton-sardine-anchovy short food web (including seawater) in the Gulf of Lions, NW Mediterranean; (ii) assess (whenever possible) the spatial and seasonal variability of this burden; (iii) identify the potential links between the contamination of anchovies and sardines and their respective trophic ecology; (iv) define trace metal pattern(s) in terms of bioaccumulation, behavior and transfer within the studied small pelagic fish food web.

Both essential (cobalt (Co), nickel (Ni), copper (Cu), and zinc (Zn)) and non-essential elements (silver (Ag), cadmium (Cd), mercury (Hg), and lead (Pb)) were considered.

2. Material and Methods

2.1. Study area

The Gulf of Lions (GoL) in the NW Mediterranean Sea is characterized by complex hydrological dynamics with: (i) a cyclonic Northern Current flowing along the continental slope; (ii) a combination of wind-driven processes such as coastal upwelling and dense shelf water formation; and (iii) freshwater dynamics associated with the large Rhône River discharge (Millot, 1999). The Rhône accounts for the highest mean annual discharge (*ca.* 1700 m^3 s^{-1}) into the Western Mediterranean basin

(Launay et al., 2019), including 95% of suspended particulate matter (SPM) fluxes to the French Mediterranean coast (Sadaoui et al., 2016) and 50% of GoL primary production (Lochet and Leveau, 1990). The Mediterranean Sea's very low tidal range allows the Rhône riverine plume to expand westwards into the GoL (Boudet et al., 2017; Many et al., 2018). This plume is particularly apparent in the first two meters of the water column (Lorthiois et al., 2012). The influence of SPM from the Rhône River on both surface water and sediment is observed throughout the western Gulf (Durrieu de Madron et al., 2000; Espinasse et al., 2014a).

Various zooplankton habitats exist in the GoL, characterized by different biological and physical variables: species composition, size structure, depth, salinity, wind and currents (Espinasse et al., 2014a). Differences in zooplankton and pelagic fish isotopic signatures and radionuclide contamination have already been observed in the eastern and the western areas of the GoL (Espinasse et al., 2014b; Strady et al., 2015a). Phytoplankton and zooplankton communities display conspicuous seasonal variations in composition and structure, reflected in their respective carbon and nitrogen isotopic signatures (Bănaru et al., 2013; Espinasse et al., 2014a, 2014b; Harmelin-Vivien et al., 2008). Phytoplankton spring bloom generally occurs between March and June in the GoL (Alekseenko et al., 2014).

2.2. Seawater and plankton sampling

Seawater and plankton were sampled in May 2010 (spring) and February 2011 (winter) using the RV "L'Europe" at six to seven stations (depending on the season) along an East-West transect of the GoL (Fig. 1). As described in previous publications related to this study (Strady et al., 2015a, 2015b; Tiano et al., 2014), the sampling strategy was adapted to the compartment and size of the sampled organisms. A chlorophyll *a* (Chl-a) concentration profile (measured continuously) was obtained at each sampling site using a CTD probe fitted with a fluorimeter. Plankton sampling was performed at the maximum Chl-a concentration depth (generally around 10-15m water depth). Seawater for trace metal analysis was pumped from the surface and at depths of 10, 20, 30, 40 and 50 m using an all-Teflon tube and surface pump system. The water was pressure-filtered on board through 0.45 µm mesh pre-cleaned with HNO_3 acid and pre-weighed polycarbonate filters (Nucleopore®) under a clean laminar flow hood installed in a trace metal-clean van. A sub-sample (~500 mL) of the filtered seawater was transferred into acid-cleaned Teflon® (FEP) bottles and acidified with ultrapure (SupraPur® quality from Merck) HCl (0.4%) for further dissolved total Hg analyses. The remaining filtered seawater (~1L) was transferred into acid-cleaned polyethylene bottles and acidified with ultrapure HNO_3 (0.1%) for further dissolved trace metal (other than Hg) analyses. Both sub-samples of filtered seawater were hermetically sealed, double-bagged and stored in the dark at 4° C pending analytical processing.

Plankton was collected by pumping or trawling according to the target size. Small planktonic organisms were sampled by pumping seawater *in situ* (nominal pumping rate 320 L/min) at the Chl-a maximum depth using an 8-cm diameter tube and filtered on board using a series of sieves made out of plankton net, mesh size 200, 60 and 6 µm. Two small plankton size fractions were thus retained ([6-60 µm] and [60-200 µm]). The trawling system was used to collect larger plankton (larger than 200 µm mesh) and towed at a speed of 2-3 knots for around 30 minutes near the Chl-a maximum depth. The samples were immediately sieved on board in the trace metal laboratory (i.e. clean van) using a sieve column with four different filter mesh size: 2000, 1000, 500 and 200 µm. All plankton fractions were kept in acid pre-cleaned polyethylene tubes and frozen at -18°C on board. They were then freeze-dried and kept in the dark at room temperature in the laboratory pending analyses.

2.3. Fish sampling

European sardines (*S. pilchardus*) and anchovies (*E. encrasicolus*) were collected in July 2010 (summer) during the yearly PELMED pelagic surveys conducted by the French Institute for the Exploitation of the Sea (Ifremer) and in March 2011 (winter) by professional fishermen, in two areas of the GoL corresponding to eastern and western plankton stations (Fig. 1). Immediately after collection, they were identified according to species and sampling area or station, then stored in a freezer in plastic bags at -18°C. The fish (sardines: n = 280 individuals in total; anchovies: n = 265 individuals in total) were then dissected in the laboratory in clean and contamination-free conditions. The sampled tissues/organs included pieces of white muscle, liver, gonads (females only) and “remaining tissue” (including remaining muscle, skin, head, skeleton, viscera, etc.). They were placed back in the freezer immediately after dissection. The dissected sardines measured 8.0-13.9 cm in length; anchovies measured 9.9-13.3 cm in length. In order to collect enough biological material for analysis, sample pools were constituted according to sampling area/station, species, gender (males vs. females), and tissue/organ type. For sardines and anchovies, 27 and 23 pools of individuals were considered respectively, together with 27 and 23 pools of tissues/organs respectively per species. The pools contained 11 individuals on average. Each pool was homogenised, re-frozen, freeze-dried and ground into a fine powder using an agate mortar or stainless-steel blade mill (for “remaining tissue”) pending further chemical analyses. The agate mortar, grinding bowls and stainless-steel blades were thoroughly washed with milli-Q water after grinding each sample.

2.4. Trace metal analyses

Total trace element concentrations in seawater, size-classed plankton and fish tissue were analysed at the Ifremer LBCM laboratory in Nantes, France. This laboratory regularly performs inter-calibration studies (www.quasimeme.org).

Mercury in seawater samples and biological compartments was assessed using a different approach to the other study metals (Co, Ni, Cu, Zn, Ag, Cd, Pb) in terms of both sample treatment and analytical techniques. All Hg analyses on seawater were performed within 3 months of sampling using an Atomic Fluorescence Spectroscopy detector (AFS, Tekran, model 2500®) as described in Cossa et al.

(2011) coupled to an LBCM-built front end, according to the US-EPA method N°1631 (U.S. Environmental Protection Agency, 2002). Total Hg in solids (biological compartments) was assessed by atomic absorption spectrophotometry on aliquots of sample powder (10–50 mg) using an Advanced Mercury Analyser (ALTEC AMA-254, Altec Ltd), according to the standard operating procedure described in the US-EPA method N°7473 (U.S. Environmental Protection Agency, 1998).

Dissolved seawater concentrations of Co, Ni, Cu, Zn, Ag, Cd and Pb were determined with a Quadrupole Inductively Coupled Plasma Mass Spectrometer (Q-ICP-MS, Thermo Electron Corporation, Element X Series®) on acidified filtrates treated according to an adapted protocol from Danielsson et al. (1982) and described in detail by Chiffoleau et al. (2002) and Guesdon et al. (2016), after pre-concentration using a liquid/liquid extraction procedure. Biological compartment samples were analysed according to an in-laboratory approved method. Briefly, dried samples (~200 mg dry mass wherever possible) were placed in microwave Teflon® bombs and mineralized using a mixture of ultrapure HNO_3 and HCl acids. The digests were then diluted to 50 mL with milli-Q water. Total metal concentrations were also determined using Q-ICP-MS.

The quality assurance of all metal analyses relied on blank controls and the accuracy and reproducibility of data relative to the certified reference materials (CRMs) used in each analytical run. Blank values were systematically below the detection limits and CRM values concurred with certified concentrations. Details of the CRM analyses are reported in Table S1, together with the limits of quantification (LOQs) for each metal and each matrix (seawater and biological compartments).

2.5. Data treatment and statistical analyses

All data submitted to statistical tests were first checked for normality (through a Shapiro-Wilks test) and/or homogeneity of variances (Bartlett's test). If these conditions were fulfilled, parametric tests were then used in the subsequent analyses; otherwise, non-parametric analogues were used. All statistical tests were performed with the software R version 3.4.3 (R Development Core Team, 2017). Detailed data per station for seawater (dissolved concentrations) and size-classed plankton are provided in Supplemental Material (Tables S2 and S3).

In order to apprehend variations throughout the water column, trace element concentrations in seawater (dissolved metals) and salinity were first measured according to sampling depth, station and season (Figs. 2, 3 and 4, Table S2). Within each season, variations in seawater concentrations were then assessed by calculating coefficients of variation (Table 1). Non-parametric Spearman correlation coefficient tests were applied to identify potential relationships between salinity and concentrations of elements in seawater in the GoL per element and per season, across all stations (Table 2). The potential enrichment of surface waters in metals (<10 m) versus the remaining water column (i.e. 10-20 m and ≥20 m) was also tested using parametric ANOVA tests followed by a Tukey's HSD post-hoc pairwise comparison test, or non-parametric Kruskal-Wallis (KW) tests followed by a multiple comparison test with Holm's adjustment method, per element and per season, across all stations. In order to assess the effect of season and geography on dissolved trace metal concentrations in the GoL, parametric Student t-tests or non-parametric Mann-Whitney-Wilcoxon (MWW) tests were performed per element across all stations to ascertain seasonal variations, and per season to ascertain geographical variations (Table 3). To avoid bias, the tests used data obtained at 10 to 40 m, i.e. the depth range at which most sampling was performed, as few samples were collected from surface waters and at 50 m. Moreover, previous statistical tests revealed no significant differences between the 10-20 m and ≥20 m depth ranges across all study elements and in either season (see Results).

The homogeneity and consistency of biological compartments were first improved by solely taking into account pools containing all three tissues, i.e. liver, muscle and "remaining tissue" (plus gonads for females), enabling the calculation of concentrations in "whole organisms" using the following formula:

[Whole organism] = (([Liver]*Liver mass) + [Muscle]*Muscle mass + [Remaining tissue]*Remaining tissue mass (+ for females: [Gonads (for females)]*Gonad mass)) / (Liver mass + Muscle mass + Remaining tissue mass (+ for females: Gonad mass))

where "[Tissue/organ]" is the metal concentration determined in the relevant tissue/organ (in mg kg^{-1} dry mass), weighted by the mass of each tissue/organ comprising the whole organism or total body weight (in mg dry mass). Indeed, metal burden (or concentration) values in whole organisms are more relevant data than concentrations measured in certain tissues/organs in terms of assessing the trophic transfer of biogeochemical elements between two trophic levels and through food webs (e.g. Cherel et al., 2005; Lahaye et al., 2005).

The statistical procedure adopted for biological compartments was as follows: due to the relatively low number of samples available per compartment in a given season (Table 4), statistical seasonal differences and variations among biological compartments could not be tested. Seasonal concentration variations measured within each compartment were therefore only considered in separate samples collected in spring (plankton) or summer 2010 (fish) and in winter 2011 (Figs. 5 and 6). Spatial variations in selected compartments in summer (plankton) and spring (fish) 2010 (i.e. season(s) and biological compartments with a minimum number of available samples ($n \geq 4$ per zone), Table 4) were statistically analysed using Student t-tests or MWW tests (Table 5). In fish species that did not display any spatial statistical variations (see results), variations according to tissue/organ were tested using KW tests (Fig. 7) and variations according to gender (for a given tissue/organ) were examined using Student t-tests or MWW tests (Table 6), using samples collected in summer 2010 only (i.e. with enough fish samples for statistical tests, Table 4). Variations according to species (for a given tissue/organ except gonads) were also tested using Students t-tests or MWW tests (Table 7) using fish collected in summer 2010.

Finally, bioaccumulation factors (BAFs) were calculated for each biological compartment of the considered theoretical food web (different plankton size fractions and fish species) and each trace element, according to the following equation of Griboff et al. (2018):

$$BAF = C_{ssbo}/C_{sw}$$

whereby C_{ssbo} is the element concentration in biological organisms at steady state (in mg kg^{-1} dry mass), and C_{sw} is the element concentration in seawater (in mg L^{-1}). As planktonic organisms and fish live at different depths throughout the day and throughout their lifecycle, selected seawater concentrations included all of the sampled water column (Fig. 8, Table S2). Finally, the correlation between BAFs calculated for the different metals (Table S5) was tested using non-parametric Spearman correlation coefficient tests and seasonal differences (per element) were tested using Student-t tests or MWW tests.

3. Results

3.1. Metals in seawater (dissolved metals)

Spatial and temporal variations of dissolved trace metal concentrations were recorded in the GoL. Larger fluctuations occurred in spring for all elements except Pb, as indicated by higher coefficients of variation in spring (Table 1). Variations in dissolved trace metal concentrations were also analyzed according to depth, from the surface to 50 m in both seasons (Figs. 3 and 4). Surface waters (<10 m depth) were significantly enriched in Co, Ni, Cu and, to a lesser extent, in Zn in spring (i.e. no statistical difference between the <10 m and 10-20 m depth ranges for Zn, but statistical difference between the <10 m and ≥20 m depth ranges) and in Zn alone in winter (ANOVA or KW tests followed by post-hoc multiple comparison tests, $p < 0.05$). Mean concentrations of all considered elements (Co, Ni, Cu, Zn) did not differ between the 10-20 m and ≥20 m depth ranges in either season (post-hoc multiple comparison tests, $p > 0.05$). No significant differences in the three other study elements (Cd, Hg and Pb) were observed according to depth in either season (ANOVA or KW tests followed by post-hoc multiple comparison tests, $p > 0.05$). Cobalt and Ni were significantly negatively correlated with salinity in both seasons (Table 2), while Cu and Zn were significantly negatively correlated with salinity in spring only, and Cd and Pb were significantly positively correlated with salinity in winter only. No correlation with salinity was observed for Hg in spring (Table 2).

Higher mean concentrations of Co and Cu were determined in the water column (10-40 m depth) in spring (MWW tests, $p = 0.008$ and $p = 0.026$ respectively), with higher mean concentrations of Cd in winter ($p < 0.001$), while no seasonal differences were observed for Ni, Zn and Pb (MWW tests, all $p > 0.05$). In both seasons, significantly higher dissolved concentrations of Co and Ni were determined in the western part of the GoL, with higher concentrations of Pb in the eastern part (Table 3). Higher concentrations of Cd and Cu were also determined in the western part of the GoL in spring, but not in winter. No spatial differences were observed for Zn and Hg and no clear pattern emerged at a station level (Figs. 3 and 4; Table S2).

3.2. Metals in size-classed plankton

Overall, fraction size appeared to be a major factor in trace metal concentration variations measured in plankton (Table 4, Figs. 5 and 6). The highest values of all metals (Co, Ni, Cu, Zn, Ag, Hg and Pb), except Cd, were found in the smallest size fraction [6-60 µm] and, to a lesser extent, in [60 - 200 µm]. These two size fractions also displayed the greatest concentration variability. The highest concentrations of Cd were determined in intermediate size fractions [200-500 µm], [500-1000 µm] and [1000-2000 µm]. The lowest concentrations of all metals were generally recorded in the largest size fraction [>2000 µm], especially in spring (Table 4, Figs. 5 and 6). Mean concentrations of trace metals in plankton were generally more variable in spring than in winter (Figs. 5 and 6). In the four compartments in which spatial differences could be tested (i.e. [6-60 µm], [60-200 µm]; sardines and anchovies collected in spring (plankton size fractions) or summer 2010 (fish), Table 5), no variations in Co, Ni, Cu, Zn or Ag were observed. However, significant spatial differences in Cd, Hg and Pb were revealed in the [60-200 µm] size fraction only, with significantly higher mean concentrations in plankton in the eastern part of the GoL (Table 5).

3.3. Metals in fish

Trace metal analyses and statistical tests were performed on fish collected in summer 2010 (i.e. season with sufficient samples) in order to appreciate variations according to zone, tissue/organ, gender and species. Spatial variations were investigated separately for each species considering whole individuals and no significant differences were revealed (Table 5). Conversely, trace metal concentrations in tissues/organs were significantly different in the two species, with identical patterns observed in both anchovies and sardines (Table 6, Fig. 7). The highest concentrations of all elements except Ni and Zn were systematically found in the liver and the lowest concentrations in muscle (except Hg). The highest concentrations of Ni and Zn were found in female gonads in both fish species, with higher values in sardines than anchovies (Fig. 7). Variations according to gender were tested separately on the basis of tissues/organs and species (Table 6). In sardines, significant gender-related variations were only found in Ag in “remaining tissue”, with females showing slightly lower Ag concentrations than males. These variations were more conspicuous in anchovies: females displayed significantly lower liver concentrations of all metals except Ni (Cu, Zn, Ag, Cd, Hg and Pb) versus males and significantly lower concentrations of Cu in whole individuals (Table 6).

Finally, significantly higher mean concentrations of Ni, Cu, Zn, Ag, Cd and Hg were found in anchovy “remaining tissue” or whole individuals versus sardines (Table 7, Figs. 5 and 6). Significantly higher concentrations of Ni, Cu, Cd and Hg were also found in anchovy liver versus sardine and significantly higher concentrations of Ni and Hg were found in anchovy muscle versus sardine. Among the trace elements analysed, the only exception was hence Pb, which was found in higher concentrations in sardines across all tissue types (Table 7).

3.4. Bioaccumulation factors (BAFs)

Bioaccumulation factors (BAFs) were calculated for each plankton fraction and fish species in both seasons, taking into account dissolved metal concentrations measured throughout the sampled water column (Fig. 8; Table S4). BAF variation patterns in the food web were fairly similar for given metals regardless of season, but differed among metals (Fig. 8; Table S4). The highest BAFs were obtained on the two smallest fractions [6-60 µm] and [60-200 µm] for all metals except Cd. Four BAF profiles were differentiated using Spearman rank correlation coefficient tests. Firstly, Co, Ni, Cu and Pb were significantly correlated, with Spearman rank correlation coefficients (r) varying from 0.909 to 0.986 (Table S5). All four elements exhibited similar profiles, with a continuous BAF decrease along the food web from the smallest phytoplankton fraction [6-60 µm] up to fish (Fig. 8). Secondly, Zn BAFs were also significantly correlated with Co, Ni, Cu and Pb BAFs, but with lower r values (from 0.579 to 0.700). The Zn profile was similar to those of Co, Ni, Cu and Pb in plankton, but increased slightly in fish: this pattern was particularly apparent when BAF was expressed in log (Fig. 8; Table S4). Thirdly, Hg displayed a particular BAF profile, with a strong decrease in plankton according to size, particularly between phytoplankton [6 - <200 µm] and zooplankton [200 to >2000 µm] and a sharp increase in both fish species (Fig. 8). Lastly, Cd exhibited a completely different BAF profile, which was not significantly correlated with any other element (Table S4). Cadmium was the only metal to show the highest values in zooplankton [200 to >2000 µm] rather than small phytoplankton size classes [6 - <200 µm], with low BAF values in fish too (Fig. 8; Table S4).

Seasonal differences were highlighted in some compartments, with slightly higher BAFs in winter for Co, Cu, Zn and Pb, in particular Co in the smallest fractions and Cu, Zn and Pb in the intermediate fractions (Table S4). However, no significant seasonal variations in the study elements were found across compartments (Student-t tests or MWW tests, $p < 0.05$), except for Zn (MWW test, $p = 0.015$), which showed a higher mean BAF value in winter (*ca*. 773 00 ± 144 000 dm) than in spring (plankton) and summer (fish) (*ca*. 570 000 ± 375 000 dm).

4. Discussion

Our study enabled the characterization of trace metal burdens in seawater, plankton and two major small pelagic fish species from the GoL in the NW Mediterranean Sea. A consistent and original database was obtained on trace metal contamination in the study area, including its short, small pelagic fish food web, hence reinforcing available data on radionuclides and rare earth elements (Strady et al., 2015a, 2015b). This geographical area is of major economic and ecological importance and has been widely investigated in recent years, in particular in the aim of understanding the potential drivers of change observed in the small pelagic fish community (e.g. Brosset et al., 2017, 2016, 2015, 2015; Le Bourg et al., 2015; Van Beveren et al., 2017, 2016). However, thorough information on the chemical contamination of pelagic compartments (from seawater to pelagic fish) by trace elements was lacking.

4.1. Variability of metals in seawater and vertical distribution patterns

Although a relatively large number of trace metal studies on the Mediterranean Sea were performed in the 1980s and 1990s (see Yoon et al., 1999), recent dissolved trace metal measurements remain scarce (however, see Battuello et al., 2016 and Heimbürger et al., 2011 for recent data reported in the northwestern Mediterranean Sea). Overall, surface and sub-surface (i.e. 0-15 m depth) concentrations determined in the GoL versus the range of surface metal concentrations reported in recent decades in the Mediterranean Sea (see Heimbürger et al., 2011; Lacan et al., 2006; Morley et al., 1990; Riso et al., 1994; Yoon et al., 1999; Zeri and Voutsinou-Taliadouri, 2003) are as follows: (i) in the same range for Zn, (ii) in the same range or higher for Ni, Cu, Cd, Pb, and (iii) higher than previously-reported for Co and Hg. However, in direct comparison with a recent study in the northwestern Mediterranean Sea (Ligurian Sea), the seawater concentrations we measured in the GoL were lower than those reported by Battuello et al. (2016).

Dissolved trace metal concentrations in Mediterranean surface waters are generally higher than in the Atlantic Ocean (Boyle et al., 1985; Morley et al., 1997), mainly due to atmospheric inputs (including Saharan dust events and European anthropogenic emissions) and riverine outflows on the continental shelves (Durrieu de Madron et al., 2011 and references therein). In the Mediterranean Sea, surface-enriched concentrations of Co, Cu, Ni and Zn, along with their significant negative correlations with salinity, suggest that concentrations are influenced by the Rhône River plume, as already demonstrated in the GoL (e.g. Radakovitch et al., 2008; Cossa et al., 2017). However, as this pattern of surface-enriched waters is only observed for essential elements (Co, Cu, Ni and Zn) and not non-essential elements (Cd, Hg and Pb), we cannot exclude a potential uptake by plankton in surface waters (Battuello et al., 2016).

Below the surface, significantly higher concentrations of most elements (Co, Ni, Cu and Cd) were observed in the western area of the GoL; only Pb showed higher concentrations in the eastern area. This spatial variation is mainly due to the dynamics of the Rhône River plume, which is generally directed westward by the Northern Current and prevailing winds (Gangloff et al., 2017) and to the existence of small rivers to the West (Sadaoui et al., 2016). Our study revealed that Co, Ni, Cu and Zn were more concentrated in surface desalinated waters, in particular in spring. The East-West influence of Rhône river water inputs in the GoL has already been demonstrated with regards to particulate organic matter and sediment transfer (Durrieu de Madron et al., 2000). In contrast, the higher dissolved Pb concentrations we found in the eastern area of the GoL are coherent with the results of

Strady et al. (2015a), which recorded higher ^{210}Pb concentrations in the eastern area. As Pb did not display any relationship with depth and salinity, its concentration is probably linked to the industrial and urban activities of the nearby city of Marseille and town of Fos-sur-Mer, together with inputs from the Rhône River. Dissolved Pb concentrations were indeed significantly higher at St1 and St10 adjacent to Marseille (Fig. 1) than at other stations.

In our study, seasonal variations in dissolved element concentrations were limited to Cd (higher in winter), Co and Cu (higher in spring). No seasonal-dependent variations in Ni, Zn and Pb were observed in the water column (10-40 m depth), in contrast to the study of Battuello et al. (2016), which found high seasonal variations in Ni and Zn in the Ligurian Sea, allocated to bioaccumulation by zooplankton and changes in the abundance of zooplankton taxa. In the GoL, the lack of seasonal variations in dissolved element concentrations in seawater, in particular in Ni and Zn, but also Pb, may be due to the predominant role of highly variable river and atmospheric contaminant inputs (Desboeufs et al., 2018; Dumas et al., 2015; Sadaoui et al., 2016).

4.2. Variability of metal concentrations in size-classed plankton

Recent data on trace metal concentrations in plankton in the Mediterranean Sea are also relatively scarce (however, see Battuello et al., 2016; Rossi and Jamet, 2008; Strady et al., 2015a, 2015b for recent data reported in the northwestern Mediterranean Sea) in comparison to the numerous Mediterranean Sea studies conducted in the 1980s (see in Roméo et al., 1992). Different plankton species are known to show significant variations in terms of metal bioaccumulation (Battuello et al., 2017; Bhattacharya et al., 2014; Levy et al., 2008). Therefore, variations in metal concentrations among plankton size fractions are probably related to their specific composition.

As indicated by (Espinasse et al., 2014b) and Strady et al. (2015a), the plankton composition of our samples (i.e. same samples as cited authors) was related to the size of the considered fraction: the two smallest fractions [6-60 µm] and [60-200 µm] were mainly composed of phytoplankton and detritus (detritus decreased with particle size). The larger fractions [200-500 µm], [500-1000 µm] and [1000-2000 µm] were mainly composed of copepods and crustacean larvae of increasing size. The largest fraction [>2000 µm] consisted mainly of large gelatinous organisms (salps, siphonophores, pteropods, chaetognaths), with some copepods and euphausiids (Espinasse et al., 2014b; Strady et al., 2015a).

The marked ability of most metals to be adsorbed onto small particles (dead or alive, organic or inorganic) may partly explain the highest values we observed in the smallest size fractions (i.e. higher surface/volume ratio hence higher potential for metal adsorption and absorption). Moreover, sorption processes can vary widely according to microalgae species (e.g. Levy et al., 2008 for Cu); this may also explain the highly-varied concentrations observed in the smallest fractions composed of phytoplankton and detritus. Alternatively, the relatively-high Cd, Zn and, to a lesser extent, Ag contents found in intermediate size fractions could be related to predominant copepods. Previous studies have documented that the assimilation of these metals by copepods depends on their prey and is particularly efficient when copepods feed on protozoa rather than phytoplankton (Twining and Fisher, 2004). This is due to the fact that Cd, Zn and Ag occur in higher proportions in the cytoplasmic fraction of protozoan cells, and are therefore more easily assimilated by consumers (Reinfelder et al., 1998; Reinfelder and Fisher, 1991). Our sampling campaign did not allow an assessment of protozoa proportions in the various fractions, but the study results clearly highlight Cd, Zn and Ag biomagnification in part of the trophic chain, as suspected by Reinfelder et al. (1998). Finally, the low concentrations we observed in the largest size fraction [>2000 µm] could be related to a predominance of gelatinous organisms, which concentrate metals less efficiently than crustaceans (Roméo et al., 1992), together with a possible "bio-dilution effect" due to size. Cadmium, Cu, Pb and Zn concentrations recorded in salps and copepods sampled in the Mediterranean Sea in the 1980s (e.g. Krishnaswami et al., 1985; Roméo et al., 1992) were in the same range as in our study fractions ([>2000 µm] and [200-500 µm], respectively). However, Co, Ni, Cu, Zn, Cd and Pb concentrations reported in zooplankton (>300 µm) collected from the Ligurian Sea (Battuello et al., 2016) were far lower than those found in corresponding fractions in the GoL ([200-500 µm], [500-1000 µm] and [1000-2000 µm]). As decreased metal concentrations in zooplankton (vesus phytoplankton) can be partly explained by the possible excretion of metals through faecal pellets (Rossi and Jamet, 2008), metal concentration patterns observed in the various plankton size fractions are likely to depend on both their size and species composition. While increasing cell and organism size is probably the main driver behind decreasing direct sorption process (ad- and ab-sorption) in phytoplankton, the diet, physiological characteristics and detoxification mechanisms the different zooplankton species (Battuello et al., 2017) are probably of prime importance in explaining the metal concentrations found in our zooplankton samples (corresponding to the >200 µm plankton fractions according to Espinasse et al., 2014b and Strady et al., 2015a).

4.3. Variability of metal concentrations in fish

Trace metal concentrations measured in anchovies and sardines collected in the GoL allowed us to pinpoint bioaccumulation variations according to geographical zone, organ/tissue (i.e. organotropism), gender and species. Seasonal-dependant variations could not be tested. Broadly, (i) no spatial differences were recorded (considering whole individuals); (ii) liver showed the highest concentrations (except for Zn and, to a lesser extent, Ni), while muscle showed the lowest concentrations (except for Hg); (iii) few gender-related differences were observed (more so in anchovies than sardines), with lower concentrations in females when significant; (iv) concentrations in reconstructed whole individuals were correlated with concentrations in "remaining tissue" comprising the majority of body mass; (v) anchovies were generally more contaminated by all metals except Pb in comparison to sardines (inverse trend).

The highest concentrations of most metals in liver versus muscle is a well-documented pattern, especially in fish (Durrieu et al., 2005; Le Croizier et al., 2018; Metian et al., 2013; Pouil et al., 2017), due to the direct role of the liver in metal storage and/or detoxification further to trace element incorporation, in particular through the trophic pathway/diet (Roesijadi, 1992; Siscar et al., 2014; Wang and Rainbow, 2010). Conversely, the relatively high concentrations of Hg (versus other metals) observed in muscle may be due to its high affinity with muscular protein sulfhydryl groups (-SH) (e.g. Bloom, 1992). The high Zn concentrations recorded in female gonads are probably due to its vital role in fish gonad development (Fletcher and King, 1978). Finally, on the organism scale, the liver represented less than 2% of wet body mass on average in the studied small pelagic fish species, while "remaining tissue" (including remaining muscle, viscera, gills, kidneys, bones, skin, etc.) accounted for 85-95% of body mass. Therefore, whole (reconstructed) organisms showed very similar metal concentrations to "remaining tissue".

Dietary exposure is widely considered as the main route for contaminant incorporation and assimilation (both inorganic and organic) in consumers such as fish (Fisk et al., 2001; Mathews and Fisher, 2009; Wang, 2002). However, no differences in anchovy diet according to gender were reported in the GoL by Pethybridge et al. (2014), or by Karachle and Stergiou (2014) in another area of the Mediterranean Sea (Agean Sea), suggesting that the contaminant variations we found are probably not related to differing diets. Other factors such as substantial contaminant elimination through reproduction (i.e. through spawning by female anchovy) may explain gender-related variations in anchovies; this topic has been well-documented in terms of organic contaminants in fish (Bodiguel et al., 2009). However, similar variations were not reported in sardines, while variations in anchovies were mainly found in liver. This probably indicates poor elimination of trace metals versus organic contaminants by small pelagic female fish during reproduction, as already suggested for large pelagic fish such as tuna (Chouvelon et al., 2017).

Variations in metal concentrations found in the two study fish species may also be due to differing in trophic ecologies (trophic level, prey preferences, etc.). Both anchovies and sardines were recently shown to have dietary overlaps in the GoL, with the main targeted prey being small copepods such as *Microsetella*, *Oncaea* and Corycaeidae copepods (Le Bourg et al., 2015). However, sardine have a more diverse, temporally variable and seasonally-specific feeding strategy than anchovies (Le Bourg et al., 2015; Pethybridge et al., 2014), confirming that the two species do not feed on exactly the same food sources or at the same trophic level in the GoL (Costalago et al., 2014, 2012). Moreover, anchovies tend to feed on the continental shelf and in the western GoL, whereas sardines remain nearer the coast and feed more in the eastern area (Le Bourg et al., 2015; Saraux et al., 2014). Sardines can also capture smaller prey than anchovies (Blaxter and Hunter, 1982; Costalago et al., 2014, 2012). The differing trophic ecologies of anchovies and sardines can hence account, at least in part, for the variations observed in trace metal concentrations: each species probably feeds on planktonic prey species affected by different levels of contamination.

Finally, differences in body condition and/or proximate composition could also explain some of the variations observed in the two fish species, although these parameters were not analysed in our study.

Indeed, recent studies have demonstrated that variations in metal content may be attribuable to the specific proximate composition (i.e. proteins, lipids, ash content) of fish species (e.g. Marval-León et al., 2014; Sofoulaki et al., 2018). This is due to the fact that most metals, including all the trace elements studied here, have a high affinity with the cysteine amino-acid of certain proteins, such as metallothioneins (e.g. Capdevila et al., 2012). Similarly, certain organic pollutants have a well-known high affinity with lipids (e.g. Munschy et al., 2016). Sardines have a far higher total lipid content than anchovies in the GoL (Pethybridge et al., 2014), although specific lipid content may vary according to season. If a lower lipid content theoretically corresponds to a higher protein content, this could explain, at least in part, the higher metal concentrations measured in anchovies versus sardines. However, it does not explain the exception we observed for Pb. Nonetheless, in direct comparison, our results were similar to those found by Sofoulaki et al. (2018) on individuals collected from six Greek sites (Mediterranean Sea), with higher levels of most of the study trace elements observed in anchovies versus sardines, with the exception of Pb (i.e. same as the inverse trend found in our study).

4.4. Patterns of metal bioaccumulation in the study food web (BAFs)

Field-based bioaccumulation factors (BAFs) were calculated as a ratio of chemical concentration in organisms versus seawater (e.g. DeForest et al., 2007; Gobas et al., 2009), using the dissolved metal concentrations measured throughout the sampled water column. Patterns of BAFs calculated on the basis of dissolved concentrations measured at 10-15 m depth only (i.e. plankton sampling depth) were rigorously identical (results not shown), confirming the probable night and day migration of organisms in the water column, at least at the sampling depths (0-50 m).

The higher BAFs observed in the two smallest plankton fractions were probably linked to two factors: firstly, the higher surface/volume ratio of small versus large cells/organisms, which may enhance dynamic metal sorption processes and secondly, the relatively-large proportion of detritus in these fractions (Strady et al., 2015a), which may efficiently adsorb metals onto their large, particle-specific surface area. Most metals showed a decreasing BAF in higher chains of the food web, from phyto- to zooplankton, then fish. Only two metals among those considered, Zn and particularly Hg, showed increased BAF values in fish. This pattern is coherent with the well-known biomagnifying properties of Hg through food webs, especially in its methylated forms (Chen et al., 2008; Cossa et al., 2012). The slight increase in Zn BAF in fish is also consistent with the biomagnifying potential of Zn in marine fish food chains (Wang, 2002). Conversely, the higher Cd bioaccumulation we observed in zooplankton fractions (mainly composed of copepods and crustacean larvae) versus fish may be linked to efficient copepod Cd assimilation (Twining and Fisher, 2004) and the ability of crustaceans to accumulate high quantities of Cd in their exoskeleton (Sarkar et al., 2016).

Generally speaking, the BAFs calculated in our study were lower than those calculated in the Ligurian Sea (NW Mediterranean Sea) by Battuello et al. (2016), probably due to the significant differences in dissolved metal concentrations found in the two studies (see above). Moreover, field BAFs tend to be inversely-related to exposure concentrations (DeForest et al., 2007), i.e. lower when seawater concentrations are higher in the field. This could also explain BAF differences in studies performed in environments with potentially different contamination levels and hence the differences in our results in the GoL versus those of Battuello et al. (2016) in the Ligurian Sea.

4.5. Synthesis on the seawater-plankton-fish continuum

When adequate material was available for statistical testing, slightly higher metal concentrations were found in seawater samples from western stations (especially below the surface and with the exception of Pb) and in biological compartment samples from eastern stations (i.e. for the [60-200 µm] fraction, and for Cd, Hg and Pb only). The slightly higher seawater concentrations found at western stations may be due to the influence of small river outflows in this area combined with the Rhône River plume, which is directed westward by the Northern Current and prevailing winds (Gangloff et al., 2017; Sadaoui et al., 2016). The inverse spatial trend observed in some biological compartments (i.e. higher concentrations at eastern part when significant) should be confirmed by testing larger numbers of samples per compartment, in particular plankton. The role of Rhône River loads on the overall contaminant burden of GoL organisms has already been reported with regards to radionuclides such as ^{210}Po (Strady et al., 2015a) and organic contaminants such as Polychlorinated biphenyls (PCBs) in plankton (Alekseenko et al., 2018). Small pelagic planktivorous fish in the eastern areas of the GoL are therefore probably affected by more prevalent/efficient exposure to chemical contamination via trophic pathways (i.e. plankton). However, this hypothesis should be supported by an additional experimental design for metals.

No common patterns were established for essential elements (Co, Ni, Cu, Zn) or non-essential elements (Ag, Cd, Hg, Pb). Instead, our results showed different metal uptake/level fingerprints in the GoL for the different study elements, with: (i) Co, Cu, Ni, Pb and, to a lesser extent, Zn and Ag, displaying the highest concentrations in the smallest investigated plankton fractions ([6-60 µm] and [60-200 µm]. Metal levels decreased considerably in intermediate plankton sizes and, finally, in fish (with the exception of Zn); (ii) Hg, which also displayed high concentrations in the smallest plankton fractions, far lower levels in intermediate fractions and enhanced concentrations in fish; and (iii) Cd, which showed higher bioaccumulation in intermediate zooplankton fractions versus both the smallest phytoplankton fractions and fish. These findings are globally consistent with studies previously conducted in other areas, which have reported general trends of lower metal concentrations in larger plankton (e.g. Ho et al., 2007), and/or in phyto- versus zooplankton (e.g. Rossi and Jamet, 2008), probably corresponding to small versus large plankton fractions according to the composition of our size fractions as described by Espinasse et al. (2014b) and Strady et al. (2015a). Only Hg has been documented as biomagnifying in upper trophic levels such as small planktivorous pelagic fishes (e.g. Cossa et al., 2012; Nfon et al., 2009). However, as fish live far longer than planktonic organisms, they are exposed to contaminants over a longer period: this may also explain the peculiar trend found for Hg and, to a lesser extent, Zn. Indeed, as previously stated, Hg is notoriously poorly-excreted by organisms over time versus other trace metals (Maulvault et al., 2016; Wang and Wong, 2003).

Finally, the analysis of different fish tissues revealed that metal concentrations in whole organisms and, to a lesser extent, liver, reflect potential differences between fish species more accurately than muscle tissue.

4.6. Future work and prospects

First and foremost, further studies on the topic of trace metal bioaccumulation and trophic transfer in planktonic compartments and pelagic food webs in general would greatly enhanced by a better biological-chemical coupling of the various parameters analysed on each sample. Where possible, this should include: (i) a thorough identification of the taxonomic composition of each plankton size fraction analysed for contaminants and (ii) the systematic analysis of indirect tracers of autotrophic and heterotrophic components of these fractions, together with their average trophic level (e.g. analysis of stable carbon and nitrogen isotopes, fatty acid profiles). This would improve our interpretation of concentrations determined in plankton fractions; (iii) assessment of metal fractions adsorbed/absorbed onto/into plankton (i.e. using chelating agents) to improve our understanding of metal fractions that are actually “bioaccumulated” in plankton; iv) assessment of insoluble versus soluble metal fractions in plankton, or subcellular compartmentalization, to better assess which metal fractions are actually available to upper trophic levels. Regarding fish, further studies on this topic would also be largely improved by an analysis of proximate composition and biological/trophic parameters using the same samples studied for contaminants. Moreover, our analyses of the various body parts showed that whole individuals more accurately reflect differences in "global” metal contamination among species within a food web. In terms of larger species, which are difficult to analyze whole, our results suggest liver as an alternative tissue for Ni, Cu, Cd, Hg and Pb analysis and/or muscle for Ni, Zn, Hg and Pb analysis.

More broadly, future studies on this topic would be improved by (i) fine-tuning research on the smallest plankton size, i.e. <60 µm; (ii) analysing the physical-chemical form of metals, which determines their bioavailability, transfer and bioaccumulation in organisms and food webs (e.g. methylated forms for Hg); (iii) comparing eco-regions with different trophic functioning, e.g. oligotrophic vs. mesotrophic areas, or areas subject to different anthropogenic pressures. This would enable a better consolidation of the processes we observed in terms of contaminant bioaccumulation in plankton and transfer to upper trophic levels.

Acknowledgements

This study was backed first and foremost by the “COSTAS” project (2009-2012) funded by the French “Agence Nationale de la Recherche” (ANR-007/CES 2009). Additional financial backing was provided by the CNRS/INSU through the EC2CO/DRIL “POTOMAC” project (2009-2010), ONEMA (now “Agence Française pour la Biodiversité”) and MISTRALS-MERMEX-MERITE 2016-2020 program, with institutional support from Ifremer. The authors are particularly grateful to Dominique Auger, Jane Bretaudeau-Sanjuan et Bernard Averty who took part in the sampling campaign and part of the analyses. We also thank the chief scientists and crew of the R/V “L’Europe” for assisting with sampling during COSTEAU 3 (https://doi.org/10.17600/10060040), COSTEAU 5 (https://doi.org/10.17600/11060010) and PELMED 2010 (https://doi.org/10.17600/10060060) campaigns. Finally, the authors would like to thank the professional fishermen who enabled us to collect fish samples in winter and the three anonymous reviewers of an earlier version of this manuscript for suggesting improvements. We acknowledge Laura Valentine from “English Assistance for Industry” for the English corrections made to the manuscript.

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Table 1: Mean concentrations ± standard deviation (in ng L^{-1}) of dissolved trace metals in seawater from the Gulf of Lions, in spring 2010 (N = 27) and winter 2011 (N = 28), with the range of values indicated into brackets. CV = Coefficient of variation (in %).

	Co	**Ni**	**Cu**	**Zn**	**Cd**	**Hg**	**Pb**
Spring 2010	13 ± 5 (7-30) CV = 41	236 ± 32 (205-374) CV = 13	203 ± 72 (137-488) CV = 36	256 ± 160 (144-894) CV = 63	11 ± 2 (9-18) CV = 19	0.65 ± 0.32 (0.41-1.79) CV = 50	29 ± 9 (21-60) CV = 32
Winter 2011	9 ± 2 (7-13) CV = 16	231 ± 14 (205-253) CV = 6	172 ± 37 (126-323) CV = 21	202 ± 40 (140-297) CV = 20	13 ± 2 (10-16) CV = 14	— — —	29 ± 11 (20-75) CV = 39

Table 2: Results of the Spearman correlation coefficient (r) tests and associated probability (p-value) between concentrations of dissolved elements in seawater and salinity in the Gulf of Lions (N = 27 for each metal in spring 2010; N = 28 in winter 2011). Significant correlations are in bold.

		Co	**Ni**	**Cu**	**Zn**	**Cd**	**Hg**	**Pb**
Spring 2010	r	-0.859	-0.816	-0.754	-0.549	-0.372	0.205	0.232
	r^2	0.738	0.666	0.569	0.301	0.138	0.042	0.054
	p-value	**<0.001**	**<0.001**	**<0.001**	**0.003**	0.056	0.325	0.245
Winter 2011	r	-0.577	-0.593	-0.289	-0.301	0.488	—	0.651
	r^2	0.333	0.352	0.084	0.091	0.238	—	0.424
	p-value	**0.001**	**<0.001**	0.136	0.120	**0.008**	—	**<0.001**

Table 3: Results of the Student t-tests (t) or of the Mann-Whitney-Wilcoxon tests (W) and associated probability (p-values) for the statistical comparison of dissolved trace metal concentrations between East and West parts in the Gulf of Lions (N = 21 in spring 2010; N = 20 in winter 2011). Only the data corresponding to the depths 10-40 m were considered here (see section 2.5). E = East; W = West. Significant differences are in bold.

		Co	**Ni**	**Cu**	**Zn**	**Cd**	**Hg**	**Pb**
Spring 2010	t or W	(W) 4	(t) -5.5	(W) 19	(W) 32	(t) -3.7	(W) 69.5	(W) 87
	p-value	<0.001	<0.001	0.012	0.129	0.002	0.137	0.018
		E < W	**E < W**	**E < W**	E = W	**E < W**	E = W	**E > W**
Winter 2011	t or W	(W) 17	(t) -5.8	(W) 27	(t) -0.9	(t) 1.9	—	(W) 74
	p-value	0.016	<0.001	0.115	0.374	0.075	—	0.047
		E < W	**E < W**	E = W	E = W	E = W	—	**E > W**

Table 4: Trace metal concentrations (in mg kg^{-1} dry mass) determined in biological compartments (size-classed plankton and fish), reported per element type (essential vs. non-essential), per season (spring (plankton) or summer (fish) 2010 vs. winter 2011) and per sampling zone (East vs. West). Values are mean ± standard deviation (SD). N = number of stations (for size-classed plankton) or number of pools of individuals (for fish), for which total metal concentrations could be determined within each area and at each season. Nd = Not determined.

		Essential elements				Non-essential elements			
		Co	Ni	Cu	Zn	Ag	Cd	Hg	Pb
	N	Mean ± SD	Mean ± SD	Mean ± SD	Mean ± SD	Mean ± SD	Mean ± SD	Mean ± SD	Mean ± SD
Spring or Summer 2010 - East									
6-60 µm	4	6.7 ± 2.8	45.4 ± 4.2	58.8 ± 11.1	293 ± 72	0.56 ± 0.37	0.37 ± 0.16	0.570 ± 0.513	44.0 ± 15.1
60-200 µm	4	4.3 ± 3.0	22.2 ± 8.9	30.0 ± 11.6	142 ± 30	0.43 ± 0.36	0.61 ± 0.18	0.365 ± 0.535	38.9 ± 14.3
200-500 µm	3	0.22 ± 0.06	1.9 ± 0.5	7.0 ± 0.5	154 ± 14	0.14 ± 0.03	0.89 ± 0.08	0.037 ± 0.017	0.93 ± 0.09
500-1000 µm	3	0.24 ± 0.05	2.1 ± 0.5	8.1 ± 2.2	172 ± 18	0.16 ± 0.04	0.99 ± 0.01	0.042 ± 0.026	0.96 ± 0.46
1000-2000 µm	3	0.30 ± 0.07	3.4 ± 1.7	8.7 ± 2.5	186 ± 55	0.19 ± 0.05	1.0 ± 0.1	0.043 ± 0.023	1.3 ± 0.5
> 2000 µm	3	0.34 ± 0.19	1.8 ± 0.7	4.6 ± 2.2	63 ± 41	0.10 ± 0.04	0.36 ± 0.18	0.024 ± 0.019	1.3 ± 0.6
Sardine (Wh*)	8	Nd	0.52 ± 0.17	4.8 ± 0.7	85 ± 6	0.02 ± 0.00	0.06 ± 0.01	0.130 ± 0.036	0.21 ± 0.04
Anchovy (Wh*)	8	Nd	0.77 ± 0.11	5.9 ± 0.6	116 ± 17	0.03 ± 0.01	0.12 ± 0.03	0.275 ± 0.039	0.14 ± 0.04
Spring or Summer 2010 - West									
6-60 µm	4	4.7 ± 1.1	65.6 ± 40.5	41.9 ± 31.9	439 ± 115	0.74 ± 0.74	0.38 ± 0.15	0.049 ± 0.039	72.4 ± 90.9
60-200 µm	4	3.5 ± 1.8	17.2 ± 6.8	20.3 ± 16.0	76 ± 54	0.11 ± 0.04	0.14 ± 0.04	0.031 ± 0.021	17.4 ± 5.9
200-500 µm	4	0.53 ± 0.15	3.0 ± 0.6	7.2 ± 1.9	114 ± 43	0.13 ± 0.04	0.58 ± 0.14	0.013 ± 0.009	2.4 ± 0.6
500-1000 µm	3	0.50 ± 0.25	3.0 ± 1.1	7.2 ± 0.7	138 ± 9	0.14 ± 0.02	0.49 ± 0.06	0.014 ± 0.007	2.8 ± 2.2
1000-2000 µm	4	0.54 ± 0.28	2.8 ± 1.1	6.3 ± 1.2	102 ± 20	0.11 ± 0.03	0.36 ± 0.10	0.015 ± 0.007	2.3 ± 1.4
> 2000 µm	4	0.36 ± 0.23	1.8 ± 0.9	3.3 ± 1.9	41 ± 33	0.06 ± 0.03	0.16 ± 0.13	0.008 ± 0.008	1.3 ± 0.8
Sardine (Wh*)	16	Nd	0.54 ± 0.11	5.1 ± 0.8	90 ± 12	0.02 ± 0.01	0.06 ± 0.01	0.141 ± 0.038	0.23 ± 0.07
Anchovy (Wh*)	11	Nd	0.65 ± 0.15	5.8 ± 0.7	123 ± 11	0.03 ± 0.01	0.11 ± 0.02	0.236 ± 0.054	0.13 ± 0.02
Winter 2011 - East									
6-60 µm	2	8.6 ± 0.4	44.0 ± 6.5	35.2 ± 13.0	158 ± 0	0.26 ± 0.07	0.14 ± 0.04	1.318 ± 1.459	53.2 ± 11.5
60-200 µm	2	1.8 ± 0.8	38.1 ± 29.9	27.9 ± 6.2	305 ± 92	0.20 ± 0.04	1.0 ± 0.3	0.264 ± 0.283	33.6 ± 14.2
200-500 µm	3	0.43 ± 0.14	4.4 ± 0.8	25.0 ± 16.4	198 ± 45	0.15 ± 0.02	1.2 ± 0.1	0.068 ± 0.007	9.2 ± 7.8
500-1000 µm	3	0.62 ± 0.49	5.9 ± 2.8	13.6 ± 2.3	218 ± 89	0.17 ± 0.05	0.92 ± 0.18	0.067 ± 0.011	22.5 ± 31.2
1000-2000 µm	2	0.39 ± 0.04	4.2 ± 1.3	18.6 ± 7.0	186 ± 60	0.16 ± 0.08	0.77 ± 0.02	0.063 ± 0.015	8.0 ± 3.1
> 2000 µm	1	0.95	6.9	9.1	137	0.14	0.78	0.042	11.0
Sardine (Wh*)	2	Nd	0.66 ± 0.10	4.8 ± 0.7	131 ± 5	0.02 ± 0.00	0.07 ± 0.01	0.290 ± 0.054	0.46 ± 0.0
Anchovy (Wh*)	1	Nd	0.66	6.3	131	0.05	0.11	0.371	0.17
Winter 2011 - West									
6-60 µm	3	7.5 ± 1.0	66.8 ± 18.7	42.8 ± 19.5	200 ± 40	0.31 ± 0.04	0.28 ± 0.12	0.115 ± 0.025	74.4 ± 45.1
60-200 µm	3	2.3 ± 0.5	27.2 ± 14.9	16.6 ± 3.1	126 ± 7	0.15 ± 0.02	0.65 ± 0.26	0.046 ± 0.011	13.6 ± 5.2
200-500 µm	3	0.28 ± 0.05	2.4 ± 0.7	10.8 ± 1.0	125 ± 8	0.13 ± 0.03	1.0 ± 0.1	0.040 ± 0.005	0.95 ± 0.30
500-1000 µm	3	0.30 ± 0.10	2.5 ± 0.3	9.7 ± 0.3	129 ± 10	0.14 ± 0.02	0.82 ± 0.14	0.038 ± 0.003	1.5 ± 1.1
1000-2000 µm	2	0.37 ± 0.03	4.0 ± 1.1	12.8 ± 1.4	122 ± 4	0.17 ± 0.01	0.61 ± 0.14	0.040 ± 0.003	2.5 ± 0.6
> 2000 µm	3	1.1 ± 0.4	5.5 ± 3.8	16.3 ± 6.5	107 ± 71	0.11 ± 0.06	0.43 ± 0.29	0.031 ± 0.021	8.6 ± 5.4
Sardine (Wh*)	1	Nd	0.84	6.3	121	0.02	0.06	0.335	0.62
Anchovy (Wh*)	3	Nd	0.64 ± 0.07	5.7 ± 0.4	134 ± 16	0.03 ± 0.01	0.10 ± 0.01	0.379 ± 0.066	0.13 ± 0.03

*Wh = whole individuals (reconstructed data).

Table 5: Results of the statistical tests for the differences between zones for the biological compartments: [6-60 µm], [60-200 µm], sardine and anchovy (whole individuals), collected in spring (plankton) or summer 2010 (fish). This corresponded to season(s) and biological compartments with a minimum number of samples (n ≥ 4 per zone) for testing the spatial differences (see Table 4). Results are reported per element type (essential vs. non-essential) and per biological compartment. Significant differences are in bold and the results and p-values of the statistical tests performed (Student t-tests (t) or Mann-Whitney-Wilcoxon (W) tests) are indicated (with * p <0.05; ** p <0.01; *** p <0.001).

Biological compartment	**Differences between zones**	**Biological compartment**	**Differences between zones**
Essential elements		**Non-essential elements**	
Co		**Ag**	
6-60 µm	t = 1.4; p = 0.224; East = West	6-60 µm	t = -0.4; p = 0.677; East = West
60-200 µm	t = 0.4; p = 0.670; East = West	60-200 µm	t = 1.8; p = 0.127; East = West
Sardine (Wh*)	—	Sardine (Wh*)	W = 61; p = 0.840; East = West
Anchovy (Wh*)	—	Anchovy (Wh*)	W = 58; p = 0.229; East = West
Ni		**Cd**	
6-60 µm	t = -1.0; p = 0.359; East = West	6-60 µm	W = 6; p = 0.663; East = West
60-200 µm	t = 0.9; p = 0.402; East = West	60-200 µm	**t = 5.2; p = 0.002**; East > West**
Sardine (Wh*)	W = 45.5; p = 0.269; East = West	Sardine (Wh*)	W = 62.5; p = 0.949; East = West
Anchovy (Wh*)	t = 1.9; p = 0.077; East = West	Anchovy (Wh*)	W = 52; p = 0.518; East = West
Cu		**Hg**	
6-60 µm	t = 1.0; p = 0.356; East = West	6-60 µm	t = 2.0; p = 0.090; East = West
60-200 µm	t = 1.0; p = 0.367; East = West	60-200 µm	**W = 15.5; p = 0.042*; East > West**
Sardine (Wh*)	W = 49; p = 0.375; East = West	Sardine (Wh*)	W = 46.5; p = 0.294; East = West
Anchovy (Wh*)	t = 0.2; p = 0.868; East = West	Anchovy (Wh*)	t = 1.8; p = 0.093; East = West
Zn		**Pb**	
6-60 µm	t = -2.2; p = 0.074; East = West	6-60 µm	W = 10; p = 0.686; East = West
60-200 µm	W = 13; p = 0.200; East = West	60-200 µm	**t = 2.8; p = 0.032*; East > West**
Sardine (Wh*)	t = -1.2; p = 0.250; East = West	Sardine (Wh*)	W = 50.5; p = 0.424; East = West
Anchovy (Wh*)	t = -1.2; p = 0.262; East = West	Anchovy (Wh*)	t = 0.6; p = 0.564; East = West

*Wh = whole individuals (reconstructed data).

Table 6: Trace metal concentrations (in mg kg^{-1} dry mass) determined in the different tissues of fish collected in summer 2010, reported per element type (essential vs. non-essential) and per fish species (sardine vs. anchovy). Values are mean ± standard deviation (SD), and N = number of pools of individuals considered for organotropism (i.e. metal concentrations in the different tissues). For each tissue, the results of the statistical tests for gender differences (females (F) vs. males (M)) are also given. To test gender differences, only pools of individuals whose sex could be determined were considered (i.e. F vs. M; no consideration of pools of sexually undetermined (U) individuals). Also, as no spatial differences were evidenced for fish during the summer season (see Table 5), individuals from the different zones were combined. Significant differences are in bold, and only the p-values of the statistical tests performed (Student t-test or Mann-Whitney-Wilcoxon test) are indicated (with * p <0.05; ** p <0.01; *** p <0.001).

Essential elements	Ni		Cu		Zn	
	Mean ± SD	**Differences between sexes**	**Mean ± SD**	**Differences between sexes**	**Mean ± SD**	**Differences between sexes**
Sardine (N= 24 / F: n= 9, M: n= 7; U: n= 8)						
Gonads (F)	2.9 ± 4.3	—	3.6 ± 1.3	—	481 ± 149	—
Liver	0.41 ± 0.29	F = M (p=0.077)	9.7 ± 3.2	F = M (p=0.314)	115 ± 19	F = M (p=0.935)
Muscle	0.09 ± 0.02	F = M (p=0.322)	1.9 ± 0.3	F = M (p=0.623)	48 ± 12	F = M (p=0.535)
Remaining tissue	0.56 ± 0.14	F = M (p=0.560)	5.2 ± 0.9	F = M (p=0.841)	91 ± 11	F = M (p=0.470)
Whole*	0.53 ± 0.13	F = M (p=0.686)	5.0 ± 0.8	F = M (p=0.791)	88 ± 11	F = M (p=0.620)
Anchovy (N= 19 / F: n= 9, M: n= 9; U: n= 1)						
Gonads (F)	0.71 ± 0.32	—	4.3 ± 0.4	—	168 ± 18	—
Liver	0.64 ± 0.29	F = M (p=0.077)	11.8 ± 2.1	**F < M (p=0.012*)**	127 ± 23	**F < M (p=0.001**)**
Muscle	0.12 ± 0.04	F = M (p=0.770)	2.1 ± 0.5	F = M (p=0.508)	46 ± 13	F = M (p=0.114)
Remaining tissue	0.77 ± 0.17	F = M (p=0.246)	6.2 ± 0.7	F = M (p=0.145)	129 ± 15	F = M (p=0.367)
Whole*	0.70 ± 0.15	F = M (p=0.528)	5.8 ± 0.6	**F < M (p=0.035*)**	120 ± 14	F = M (p=0.052)

Non-essential elements	Ag		Cd		Hg		Pb	
	Mean ± SD	**Differences between sexes**	**Mean ± SD**	**Differences between sexes**	**Mean ± SD**	**Differences between sexes**	**Mean ± SD**	**Differences between sexes**
Sardine (N= 24 / F: n= 9, M: n= 7; U: n= 8)								
Gonads (F)	0.03 ± 0.01	—	0.12 ± 0.04	—	0.104 ± 0.040	—	0.17 ± 0.20	—
Liver	0.10 ± 0.10	F = M (p=0.260)	0.46 ± 0.15	F = M (p=0.981)	0.270 ± 0.091	F = M (p=0.710)	0.23 ± 0.06	F = M (p=0.884)
Muscle	0.02 ± 0.01	F = M (p=0.815)	0.004 ± 0.003	F = M (p=0.439)	0.179 ± 0.059	F = M (p=0.481)	0.05 ± 0.01	F = M (p=1.000)
Remaining tissue	0.02 ± 0.01	**F < M (p=0.038*)**	0.06 ± 0.01	F = M (p=0.817)	0.134 ± 0.035	F = M (p=0.676)	0.24 ± 0.07	F = M (p=0.593)
Whole*	0.02 ± 0.01	F = M (p=0.069)	0.06 ± 0.01	F = M (p=0.753)	0.138 ± 0.037	F = M (p=0.690)	0.23 ± 0.07	F = M (p=0.710)
Anchovy (N= 19 / F: n= 9, M: n= 9; U: n= 1)								
Gonads (F)	0.08 ± 0.02	—	0.17 ± 0.04	—	0.102 ± 0.019	—	0.04 ± 0.01	—
Liver	0.10 ± 0.05	**F < M (p=0.004**)**	0.91 ± 0.25	**F < M (p=0.017*)**	0.704 ± 0.197	**F < M (p=0.012*)**	0.15 ± 0.06	**F < M (p=0.008**)**
Muscle	0.01 ± 0.01	F = M (p=0.458)	0.01 ± 0.00	F = M (p=0.514)	0.268 ± 0.067	F = M (p=0.863)	0.04 ± 0.02	F = M (p=0.513)
Remaining tissue	0.03 ± 0.01	F = M (p=0.773)	0.12 ± 0.03	F = M (p=0.686)	0.248 ± 0.052	F = M (p=0.934)	0.15 ± 0.04	F = M (p=0.348)
Whole*	0.03 ± 0.01	F = M (p=0.362)	0.12 ± 0.02	F = M (p=0.780)	0.252 ± 0.051	F = M (p=0.727)	0.14 ± 0.03	F = M (p=0.660)

*Whole = whole individuals (reconstructed data).

Table 7: Results of the statistical tests for the differences between species for the fish collected in summer 2010. The results are reported per element type (essential vs. non-essential) and per fish tissue (except gonads, collected from females only). Significant differences are in bold and the results and p-values of the statistical tests performed (Student t-tests (t) or Mann-Whitney-Wilcoxon (W) tests) are indicated (with * p <0.05; ** p <0.01; *** p <0.001).

Tissue/organ	**Differences between species**	**Tissue/organ**	**Differences between species**
Essential elements		**Non-essential elements**	
Ni		**Ag**	
Liver	**W = 103; p = 0.002**; S < A**	Liver	W = 201; p = 0.514; S = A
Muscle	**t = -2.6; p = 0.014*; S < A**	Muscle	W = 255.5; p = 0.453; S = A
Remaining tissue	**t = -4.4; p < 0.001***; S < A**	Remaining tissue	**W = 50.5; p < 0.001***; S < A**
Whole*	**t = -3.8; p < 0.001***; S < A**	Whole*	**W = 71; p < 0.001***; S < A**
Cu		**Cd**	
Liver	**W = 105.5; p = 0.003**; S < A**	Liver	**W = 20; p < 0.001***; S < A**
Muscle	W = 148.5; p = 0.053; S = A	Muscle	W = 168; p = 0.094; S = A
Remaining tissue	**t = -4.0; p < 0.001***; S < A**	Remaining tissue	**W = 3; p < 0.001***; S < A**
Whole*	**t = -3.7; p < 0.001***; S < A**	Whole*	**W = 1; p < 0.001***; S < A**
Zn		**Hg**	
Liver	t = -1.9; p = 0.066; S = A	Liver	**t = -9.6; p < 0.001***; S < A**
Muscle	W = 259.5; p = 0.448; S = A	Muscle	**t = -4.7; p < 0.001***; S < A**
Remaining tissue	**W = 13; p < 0.001***; S < A**	Remaining tissue	**t = -8.3; p < 0.001***; S < A**
Whole*	**W = 21; p < 0.001***; S < A**	Whole*	**t = -8.6; p < 0.001***; S < A**
		Pb	
		Liver	**W = 375; p < 0.001***; S > A**
		Muscle	**W = 313.5; p = 0.032*; S > A**
		Remaining tissue	**W = 420; p < 0.001***; S > A**
		Whole*	**W = 430; p < 0.001***; S > A**

*Whole = whole individuals (reconstructed data).

Figure captions

Fig. 1: Location of sampling sites in the Gulf of Lions (northwestern Mediterranean Sea). Black squares = seawater and plankton sampling stations; dotted circles = pelagic fish sampling areas; dashed line = separation between the eastern and western areas (East, West) considered in this study.

Fig. 2: Vertical profiles of seawater salinity determined at each station sampled in spring 2010 and winter 2011. Data are expressed in practical salinity unit (psu). The depth range of plankton samples (i.e. 10-15 m depth) is indicated. E = East; W = West.

Fig. 3a: Vertical profiles of (dissolved) seawater concentrations (in ng. L^{-1}) of Co, Ni, Cu and Zn (i.e. essential metals) determined at each station sampled in spring 2010 (left panel) and winter 2011 (right panel). The depth range of plankton samples (i.e. 10-15 m depth) is indicated. E = East; W = West.

Fig. 4: Vertical profiles of (dissolved) seawater concentrations (in ng. L^{-1}) of Hg, Cd and Pb (i.e. non-essential metals) determined at each station sampled in spring 2010 (left panel) and winter 2011 (right panel). The depth range of plankton samples (i.e. 10-15 m depth) is indicated. E = East stations; W = West.

Fig. 5: Boxplots of concentrations (in mg kg^{-1} dry mass) of Co, Ni, Cu and Zn (i.e. essential metals) determined in the various biological compartments, reported per element and per season (spring (plankton) or summer (fish) 2010 (blue boxes) vs. winter 2011 (red boxes)). Stations are combined. Boxplots for fish are enlarged in the upper right corner. The box length represents the interquartile, the bar length represents the range and the horizontal lines in bold are median values.

Fig. 6: Boxplots of concentrations (in mg kg^{-1} dry mass) of Ag, Cd, Hg and Pb (i.e. non-essential metals) determined in the various biological compartments, reported per element and per season (spring (plankton) or summer (fish) 2010 (blue boxes) vs. winter 2011 (red boxes)). Stations are combined. Boxplots for fish are enlarged in the upper right corner. The box length represents the interquartile, the bar length represents the range and the horizontal lines in bold are median values.

Fig. 7: Histograms of trace metal concentrations (in mg kg^{-1} dry mass) determined in different tissues (i.e. organotropism) of fish collected in summer 2010, reported per element (essential (left panel) vs. non-essential elements (right panel)) and per fish species (sardine vs. anchovy). Values are mean ± SD per tissue. N = 24 and N = 19 for each sardine and anchovy tissue type, respectively (except gonads, collected from females only, N = 9 for each species). The results of statistical tests to ascertain concentration variations among tissue types are also indicated (with numbers for sardines, and letters for anchovies). An identical number or letter indicates that tissue concentrations were not significantly different within a species (i.e. results of the post-hoc multiple comparison test with Holm adjustment method after a Kruskal-Wallis test, at $\alpha = 0.05$). F = Females; RT = Remaining Tissue.

Fig. 8: Plots of mean bioaccumulation factors (BAFs) calculated per element type (essential vs. non-essential), reported per season (spring (plankton) or summer (fish) 2010 vs. winter 2011) and per biological compartment. Concentrations used for calculations were mg L^{-1} for seawater and mg kg^{-1} dry mass for biological compartments. Top panel = BAF values $*10^3$; Bottom panel = log-transformed BAF values. For exact values see Table S4 (Supplemental Material). S = Sardine; A = Anchovy.

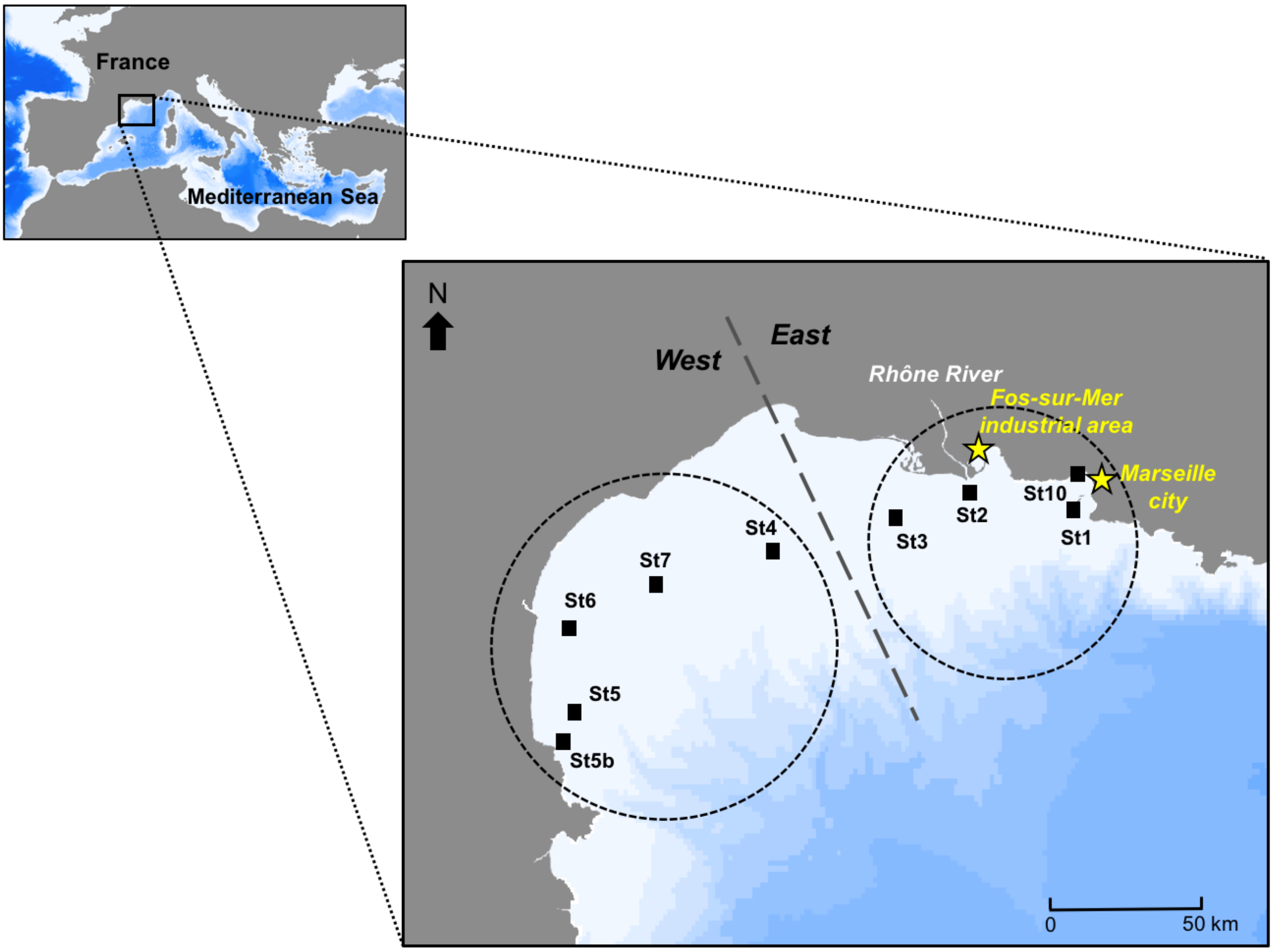
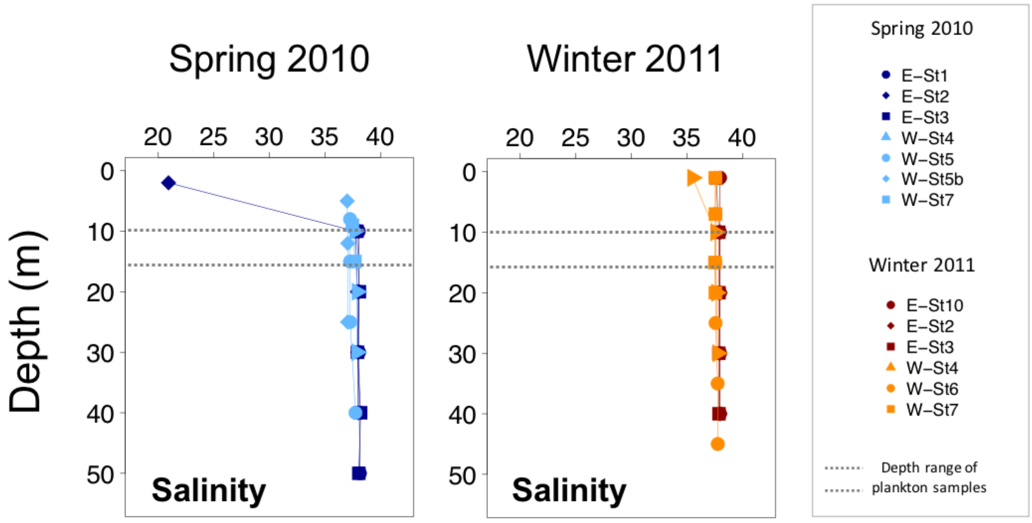

France
Mediterranean Sea
N
West
East
Rhône River
Fos-sur-Mer
industrial area
Marseille
city
St10
St2
St3
St1
St4
St7
St6
St5
St5b
0
50 km

Fig. 1



Spring 2010
Winter 2011
Depth (m)
Salinity
Salinity
Spring 2010
E–St1
E–St2
E–St3
W–St4
W–St5
W–St5b
W–St7
Winter 2011
E–St10
E–St2
E–St3
W–St4
W–St6
W–St7
Depth range of
plankton samples

Fig. 2

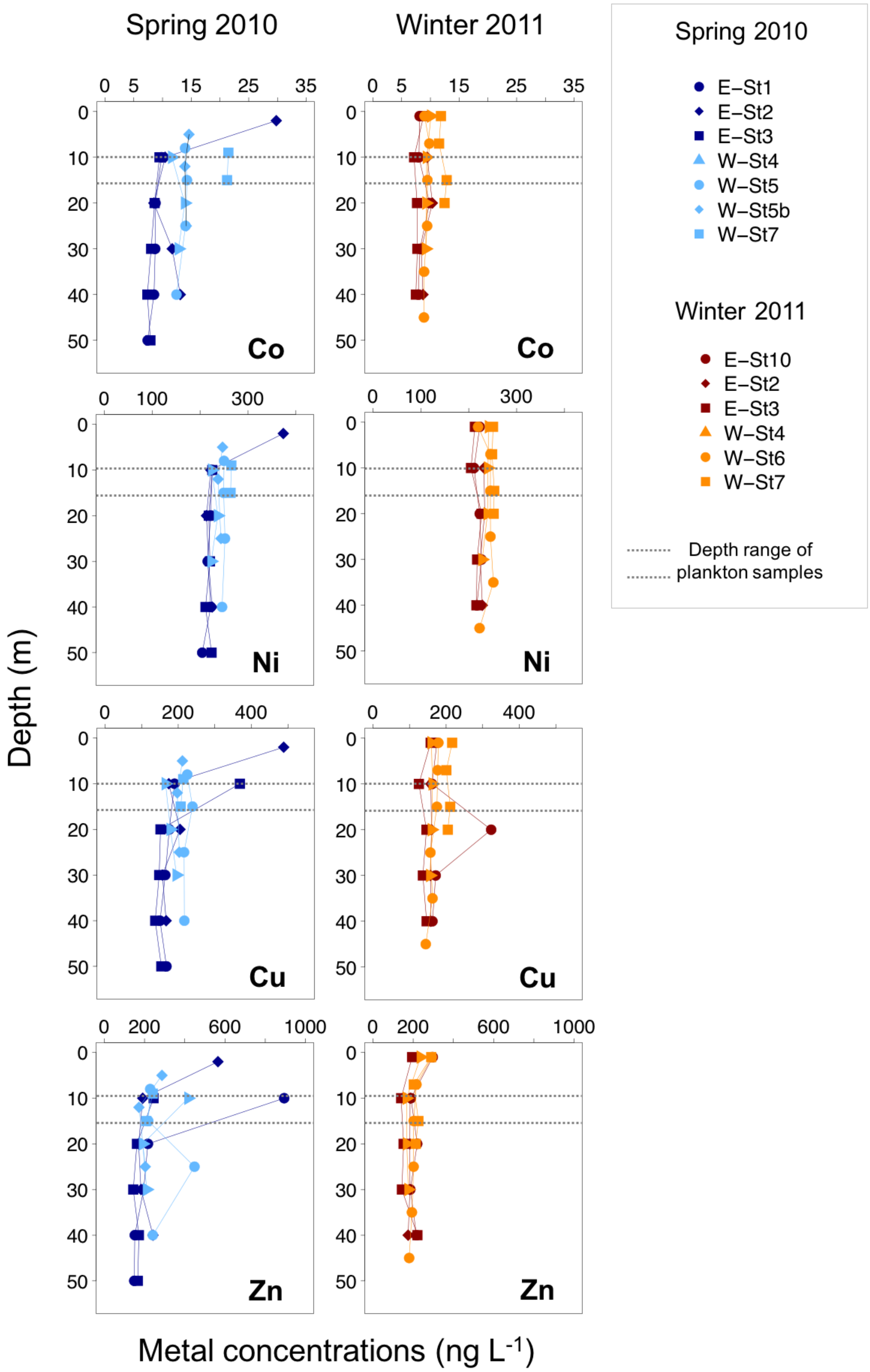
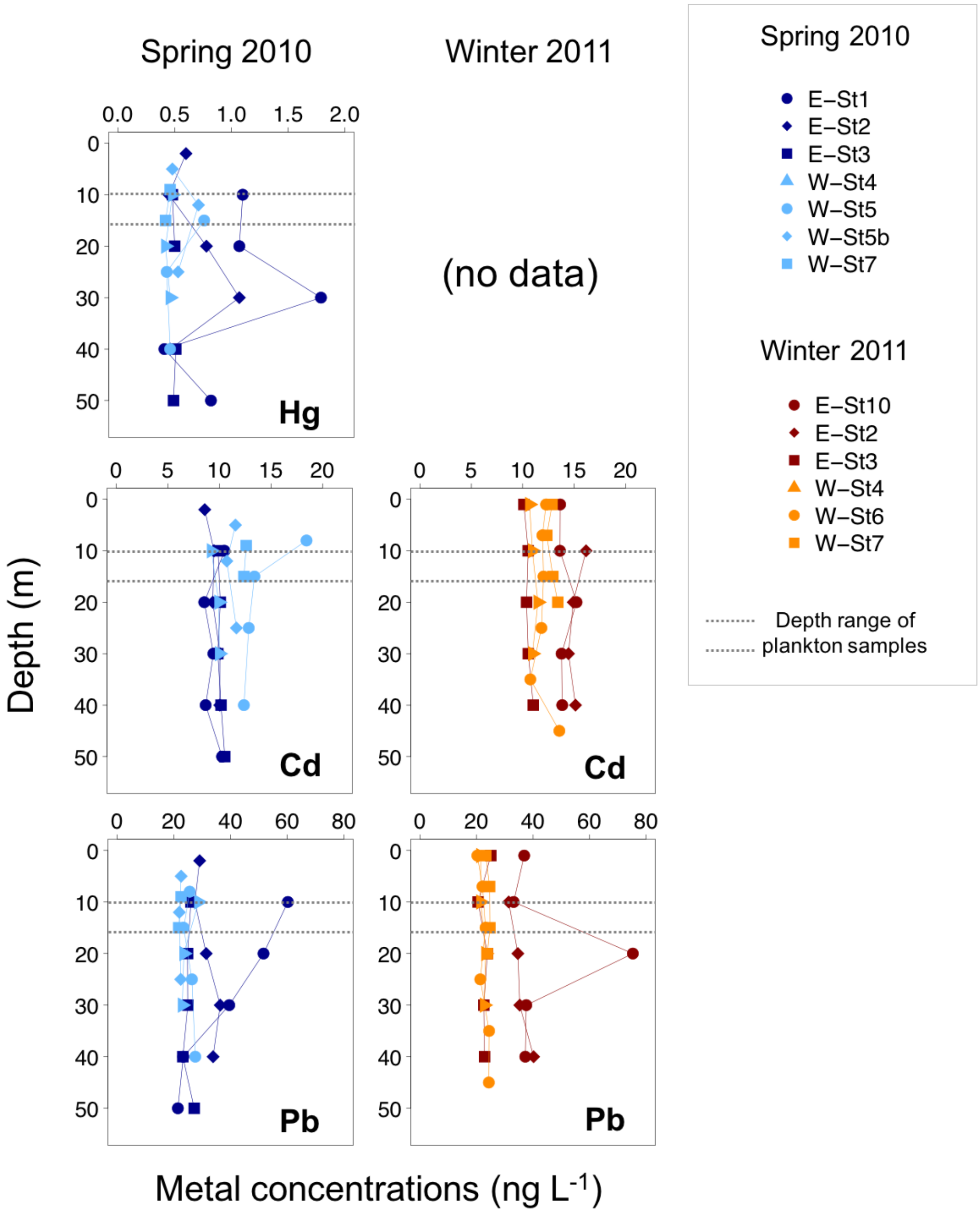
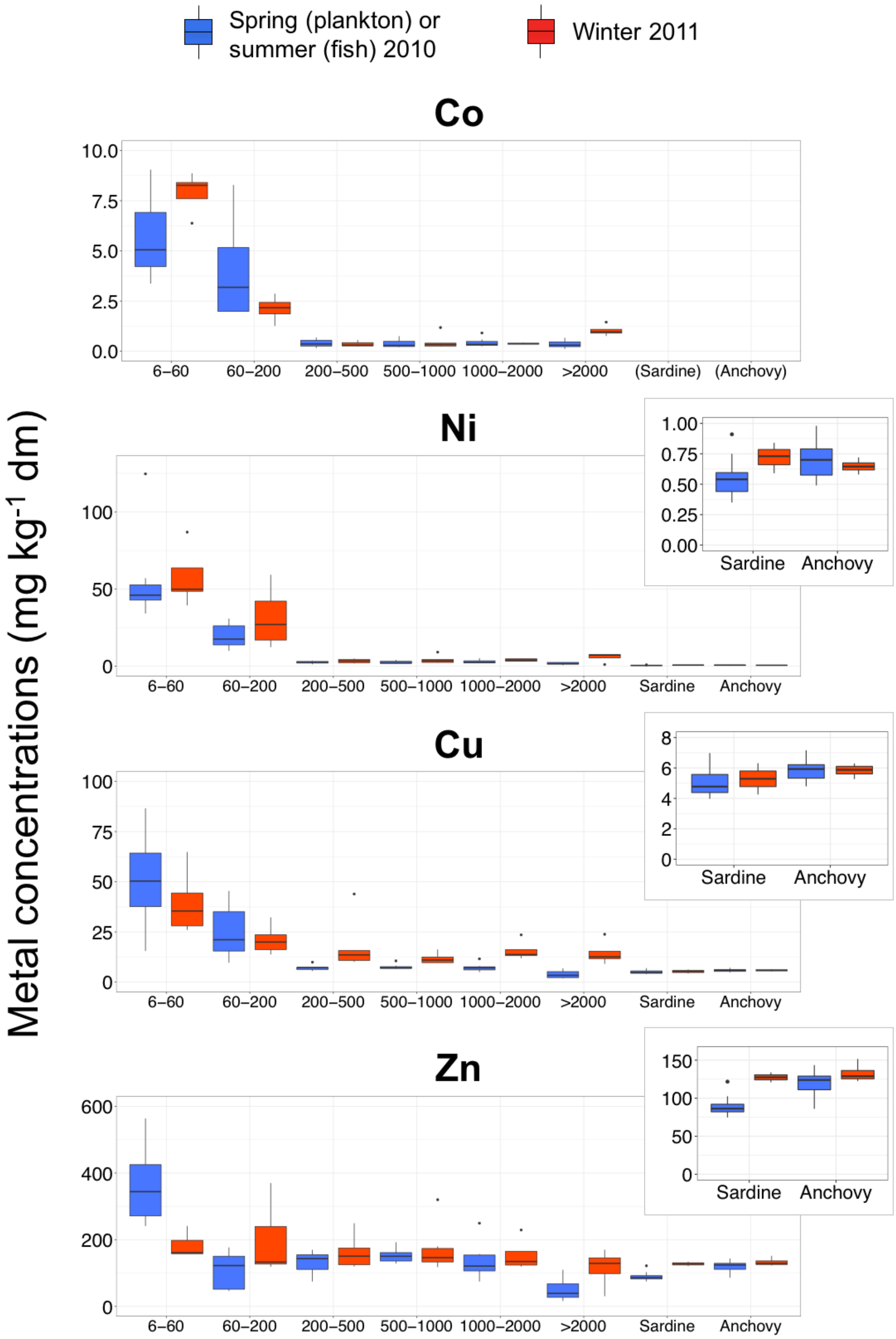

Spring 2010
Winter 2011
Co
Ni
Cu
Zn
Depth (m)
Metal concentrations (ng L-1)
Spring 2010
E–St1
E–St2
E–St3
W–St4
W–St5
W–St5b
W–St7
Winter 2011
E–St10
E–St2
E–St3
W–St4
W–St6
W–St7
Depth range of plankton samples

Fig. 3



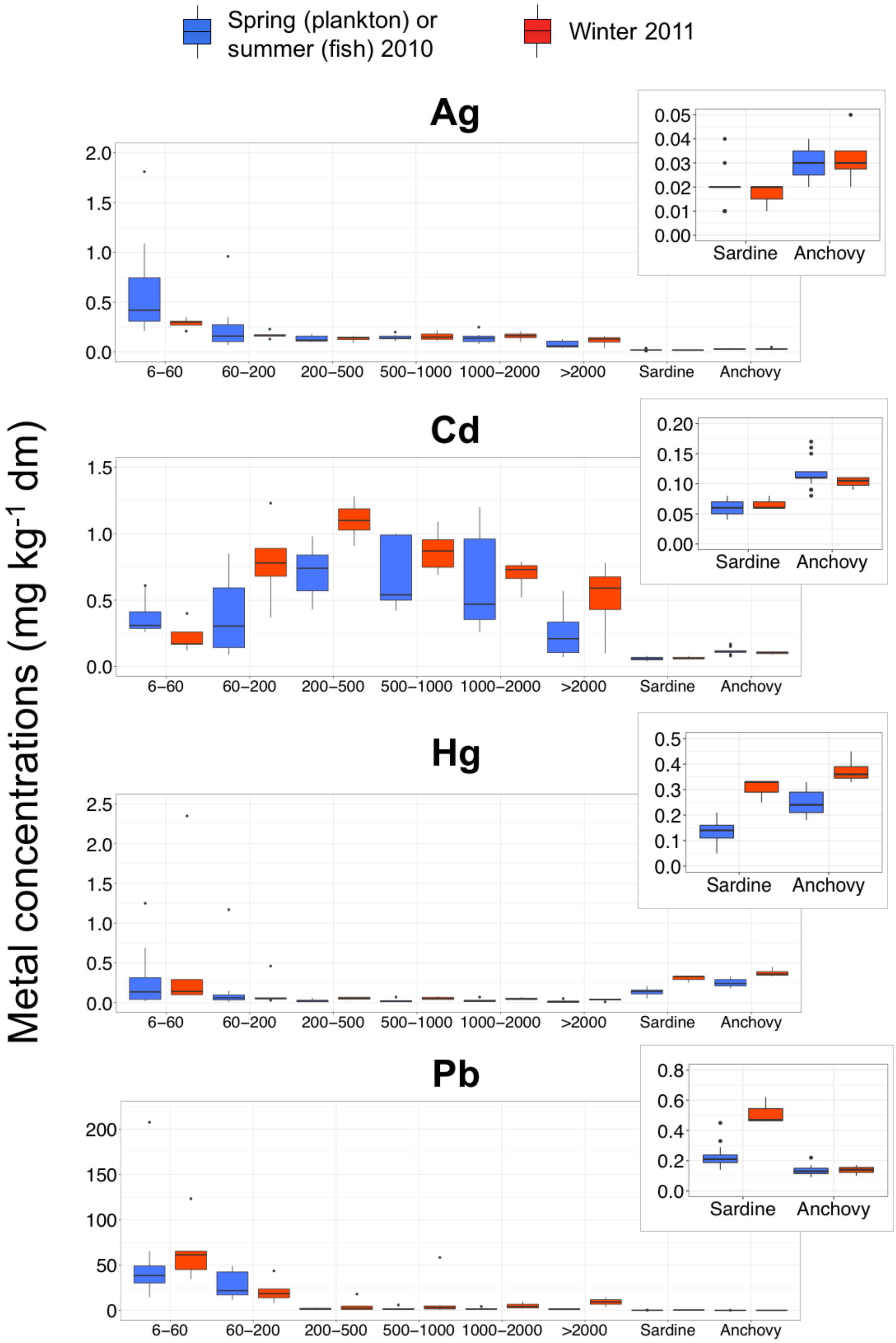
Spring 2010
Winter 2011
0.0 0.5 1.0 1.5 2.0
0 10 20 30 40 50
Hg
(no data)
0 5 10 15 20
Cd
Cd
0 20 40 60 80
Pb
Pb
Depth (m)
Metal concentrations (ng L-1)
Spring 2010
E–St1
E–St2
E–St3
W–St4
W–St5
W–St5b
W–St7
Winter 2011
E–St10
E–St2
E–St3
W–St4
W–St6
W–St7
Depth range of
plankton samples

Fig. 4



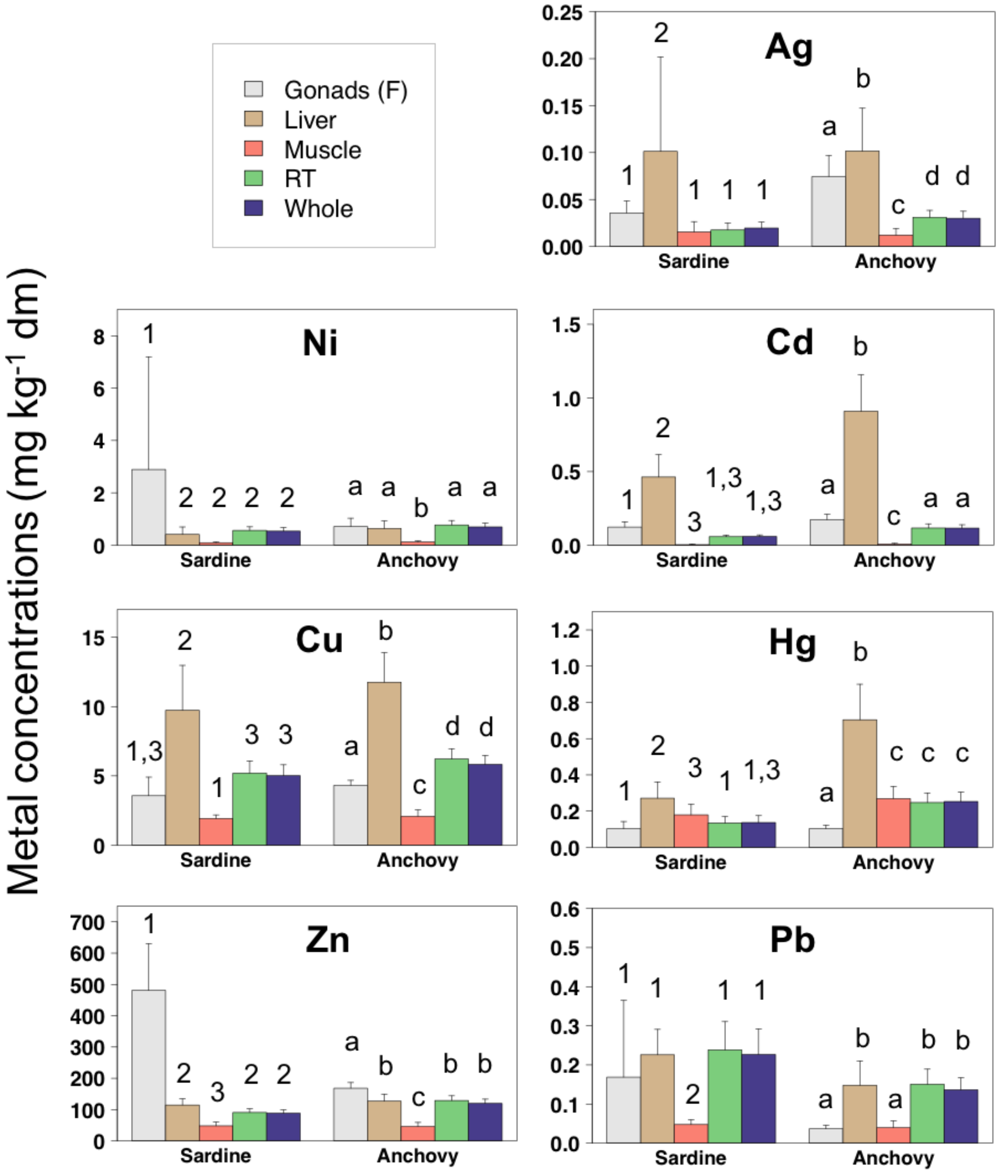
Spring (plankton) or summer (fish) 2010
Winter 2011
Metal concentrations (mg kg-1 dm)
Co
10.0
7.5
5.0
2.5
0.0
6–60
60–200
200–500
500–1000
1000–2000
>2000
(Sardine)
(Anchovy)
Ni
100
50
0
1.00
0.75
0.50
0.25
0.00
Sardine
Anchovy
Cu
100
75
50
25
0
8
6
4
2
0
Zn
600
400
200
0
150
100
50
0

Fig. 5



Spring (plankton) or summer (fish) 2010
Winter 2011
Metal concentrations (mg kg-1 dm)
Ag
Cd
Hg
Pb
6–60
60–200
200–500
500–1000
1000–2000
>2000
Sardine
Anchovy

Fig. 6



Gonads (F)
Liver
Muscle
RT
Whole
Metal concentrations (mg kg-1 dm)
Ag
Ni
Cd
Cu
Hg
Zn
Pb
Sardine
Anchovy

Fig. 7

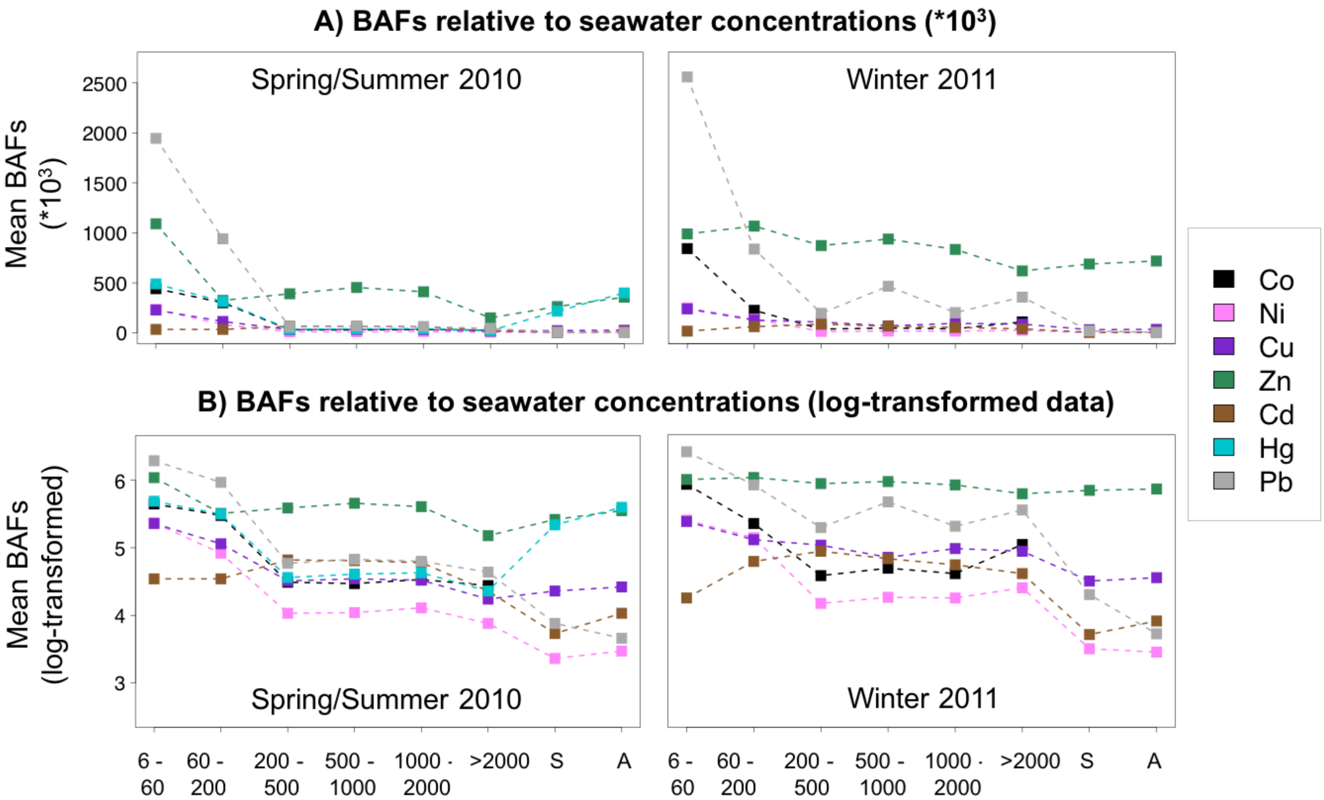

A) BAFs relative to seawater concentrations (*10³)
Spring/Summer 2010
Winter 2011
Mean BAFs
(*10³)
2500
2000
1500
1000
500
0
B) BAFs relative to seawater concentrations (log-transformed data)
Mean BAFs
(log-transformed)
6
5
4
3
Spring/Summer 2010
Winter 2011
6 - 60
60 - 200
200 - 500
500 - 1000
1000 - 2000
>2000
S
A
Co
Ni
Cu
Zn
Cd
Hg
Pb

Fig. 8

Supplemental Material

Table S1: Results obtained for certified reference materials (CRMs) used in trace metal analyses. Values are means ± standard deviation (SD), in ng L^{-1} for seawater and mg kg^{-1} dry mass for biological compartments. The limits of quantification (LOQ, in italics) and the recovery rate (in %, in bold) are also indicated. The symbol "—" appears when the element was not determined nor quantified in the samples. Nc = CRM not certified for the element.

		Essential elements				Non-essential elements			
		Co	**Ni**	**Cu**	**Zn**	**Ag**	**Cd**	**Hg**	**Pb**
Seawater									
LOQ (ng L^{-1}):		*0.2*	*0.001*	*0.01*	*0.01*	—	*0.1*	*0.075*	*0.4*
ORMS-4 (lake water, National Research Council Canada/NRCC)	Measured value	—	—	—	—	—	—	22.68 ± 0.04	—
	Certified value	—	—	—	—	—	—	22.0 ± 1.6	—
	RR (%)	**—**	**—**	**—**	**—**	**—**	**—**	**103**	**—**
CASS-5 (seawater, NRCC)	Measured value	83.1 ± 2.0	0.304 ± 0.005	0.36 ± 0.01	0.67 ± 0.03	—	20.9 ± 1.4	—	9.9 ± 0.9
	Certified value	93	0.32 ± 0.02	0.37 ± 0.03	0.70 ± 0.07	—	21 ± 2	—	11 ± 2
	RR (%)	**89**	**95**	**97**	**96**	**—**	**100**	**—**	**90**
Biological compartments									
LOQ (mg kg^{-1} dm):		*0.25*	*0.13*	*1.25*	*12.5*	*0.03*	*0.03*	*0.015*	*0.03*
BCR-414 (plankton, JRC-European Commission/EC)	Measured value	—	—	—	—	—	—	0.259 ± 0.001	—
	Certified value	—	—	—	—	—	—	0.276 ± 0.018	—
	RR (%)	**—**	**—**	**—**	**—**	**—**	**—**	**94**	**—**
SRM-2976 (mussel tissue, National Institute of Standards and Technology/NIST)	Measured value	—	—	—	—	—	—	0.058 ± 0.006	—
	Certified value	—	—	—	—	—	—	0.061 ± 0.004	—
	RR (%)	**—**	**—**	**—**	**—**	**—**	**—**	**95**	**—**
IAEA-142 (mussel homogenate, International Atomic Energy Agency)	Measured value	—	—	—	—	—	—	0.127 ± 0.006	—
	Certified value	—	—	—	—	—	—	0.126 ± 0.007	—
	RR (%)	**—**	**—**	**—**	**—**	**—**	**—**	**101**	**—**
BCR-422 (cod muscle, JRC-EC)	Measured value	—	—	—	—	—	—	0.543 ± 0.003	—
	Certified value	—	—	—	—	—	—	0.559 ± 0.016	—
	RR (%)	**—**	**—**	**—**	**—**	**—**	**—**	**97**	**—**
BCR-CRM 278 R (mussel tissue, JRC-EC)	Measured value	0.33 ± 0.01	(Nc)	8.98 ± 0.13	79.1 ± 1.7	(Nc)	0.343 ± 0.013	—	1.95 ± 0.04
	Certified value	0.34*	(Nc)	9.45 ± 0.13	83.1 ± 1.7	(Nc)	0.348 ± 0.007	—	2.00 ± 0.04
	RR (%)	**98**	**(Nc)**	**95**	**95**	**(Nc)**	**99**	**—**	**98**
DORM-3 (fish protein, NRCC)	Measured value	(Nc)	1.30 ± 0.10	14.9 ± 0.4	49.2 ± 0.4	0.03 ± 0.01	0.30 ± 0.01	—	0.34 ± 0.08
	Certified value	(Nc)	1.28 ± 0.24	15.5 ± 0.63	51.3 ± 3.1	0.04*	0.29 ± 0.02	—	0.395 ± 0.050
	RR (%)	**(Nc)**	**101**	**96**	**96**	**70**	**104**	**—**	**86**
DOLT-3 (dogfish liver, NRCC)	Measured value	(Nc)	3.17 ± 0.59	31.7 ± 0.0	89.4 ± 0.9	1.21 ± 0.01	19.0 ± 0.0	—	0.32 ± 0.01
	Certified value	(Nc)	2.72 ± 0.35	31.2 ± 1.0	86.6 ± 2.4	1.20 ± 0.07	19.4 ± 0.6	—	0.319 ± 0.045
	RR (%)	**(Nc)**	**116**	**102**	**103**	**101**	**98**	**—**	**100**

* Indicative value on the certificate.

Table S2: Detailed dissolved trace metal concentrations (in ng L^{-1}) determined in seawater, reported per season (spring 2010 vs. winter 2011), per sampling zone (East vs. West), per station and per sampling depth. The salinity is also indicated. Nd = Not determined.

Spring 2010

	Depth (m)	Salinity (psu)	Co	Ni	Cu	Zn	Cd	Hg	Pb
East									
St1	10[#]	38.0	10	225	190	894	11	1.10	60
	20	38.0	9	222	176	217	9	1.07	52
	30	38.1	9	216	165	196	9	1.79	40
	40	38.1	9	222	150	152	9	0.41	23
	50	38.1	7	204	168	149	10	0.82	21
	Mean*		**9**	**218**	**170**	**322**	**9**	**1.04**	**39**
St2	2	20.9	30	374	488	565	9	0.60	29
	10[#]	37.7	11	222	174	191	9	0.45	27
	20	37.9	9	213	205	171	9	0.78	31
	30	38.0	12	218	157	182	10	1.07	36
	40	38.0	13	225	167	242	10	0.46	34
	Mean*		**15**	**250**	**238**	**270**	**10**	**0.67**	**32**
St3	10[#]	37.9	10	225	369	245	10	0.48	26
	20	38.1	9	218	152	162	10	0.50	25
	30	37.9	8	221	148	144	10	(Nd)	25
	40	38.2	7	211	137	172	10	0.51	23
	50	38.1	8	224	154	167	11	0.49	27
	Mean*		**8**	**220**	**192**	**178**	**10**	**0.50**	**25**
West									
St4	10[#]	37.7	12	225	166	417	9	0.48	29
	20	37.9	14	237	179	191	10	0.42	24
	30	37.9	13	223	195	211	10	0.46	23
	Mean*		**13**	**228**	**180**	**273**	**10**	**0.46**	**25**
St5	8	37.3	14	250	225	228	18	(Nd)	26
	15[#]	37.3	14	249	239	219	13	0.76	23
	25	37.3	14	252	216	448	13	0.43	26
	40	37.7	12	246	217	241	12	0.46	28
	Mean*		14	249	224	284	14	0.55	26
St5b	5	37.0	15	247	212	287	12	0.48	23
	12[#]	37.1	14	238	197	172	11	0.71	22
	25	37.0	14	244	203	204	12	0.53	22
	Mean*		**14**	**243**	**204**	**221**	**11**	**0.58**	**22**

Winter 2011

	Depth (m)	Salinity (psu)	Co	Ni	Cu	Zn	Cd	Hg	Pb
East									
St10	1	38.0	8	223	175	297	14	(Nd)	37
	10[#]	38.0	8	211	162	189	14	(Nd)	33
	20	38.0	10	223	323	220	15	(Nd)	75
	30	38.0	8	227	172	186	14	(Nd)	38
	40	38.0	8	217	163	216	14	(Nd)	37
	Mean*		**8**	**220**	**199**	**222**	**14**	**(Nd)**	**44**
St2	10[#]	38.0	9	232	158	185	16	(Nd)	31
	20	38.0	10	233	160	184	15	(Nd)	35
	30	38.0	8	224	157	184	14	(Nd)	35
	40	38.0	9	228	158	175	15	(Nd)	40
	Mean*		**9**	**229**	**158**	**182**	**15**	**(Nd)**	**35**
St3	1	37.6	9	213	158	195	10	(Nd)	25
	10[#]	37.9	7	205	126	140	11	(Nd)	21
	20	37.9	8	227	146	152	10	(Nd)	24
	30	37.9	8	217	137	144	11	(Nd)	23
	40	37.9	7	216	147	221	11	(Nd)	23
	Mean*		**8**	**216**	**143**	**171**	**11**	**(Nd)**	**23**
West									
St4	1	35.5	10	242	160	237	11	(Nd)	21
	10[#]	37.6	9	240	163	167	11	(Nd)	21
	20	37.7	9	239	161	168	11	(Nd)	23
	30	37.8	9	228	156	176	11	(Nd)	23
	Mean*		**9**	**237**	**160**	**187**	**11**	**(Nd)**	**22**
St6	1	37.6	9	220	179	293	12	(Nd)	20
	7	37.6	10	245	177	216	12	(Nd)	22
	15[#]	37.6	9	245	175	202	12	(Nd)	23
	25	37.6	9	245	157	202	12	(Nd)	21
	35	37.6	9	251	163	194	11	(Nd)	24
	45	37.8	9	223	144	180	14	(Nd)	24
	Mean*		**9**	**238**	**166**	**215**	**12**	**(Nd)**	**23**

St7	9	37.5	21	265	214	241	13	0.46	23	**St7**	1	37.6	12	251	217	290	13	(Nd)	23
	15[#]	37.7	21	264	207	209	12	0.42	22		7	37.6	12	249	201	203	12	(Nd)	25
	Mean*		**21**	**264**	**210**	**225**	**12**	**0.44**	**22**		15[#]	37.5	13	253	211	226	13	(Nd)	25
											20	37.6	12	252	205	213	13	(Nd)	24
											Mean*		**12**	**251**	**208**	**233**	**13**	**(Nd)**	**24**

* Data used for the calculations of mean BAFs including dissolved metal concentrations measured throughout the sampled water column (Fig. 6, Tables S4 and S5).

[#] Data used for the calculations of mean BAFs including dissolved concentrations measured at 10-15 m depth only (i.e. plankton sampling depth; results not shown).

Table S3: Detailed trace metal concentrations (in mg kg^{-1} dry mass) determined in size-classed plankton, reported per season (spring 2010 vs. winter 2011), per sampling zone (East vs. West) and per station. Ns/d = Not sampled/not determined.

Spring 2010

	Size class (µm)	Co	Ni	Cu	Zn	Ag	Cd	Hg	Pb
East									
St1	6-60	3.4	45.6	73.3	256	1.09	0.60	1.248	65.5
	60-200	2.0	13.9	45.4	142	0.96	0.60	1.165	49.3
	200-500	0.15	1.3	7.3	142	0.18	0.82	0.056	0.88
	500-1000	0.21	1.8	10.6	192	0.20	0.99	0.072	1.5
	1000-2000	0.28	5.3	11.6	249	0.25	1.2	0.069	1.9
	> 2000	0.24	1.6	6.9	110	0.13	0.57	0.045	1.6
St2	6-60	9.0, 9.1*	41.5, 51.2*	47.8, 52.9*	241, 277*	0.31, 0.32*	0.26, 0.28*	0.156, 0.188*	32.0, 35.0*
	60-200	5.0, 8.3 *	29.0, 30.9 *	21.5, 32.1*	103, 148*	0.16, 0.35*	0.42, 0.59*	0.060, 0.151*	18.2, 40.9*
	200-500	0.25	2.0	7.3	150	0.14	0.86	0.030	0.87
	500-1000	0.21	1.9	6.6	160	0.13	1.0	0.031	0.59
	1000-2000	0.25	2.2	7.3	157	0.17	0.98	0.030	0.91
	> 2000	0.56	2.5	4.4	46	0.11	0.32	0.018	1.7
St3	6-60	5.5	43.3	61.3	399	0.52	0.35	0.687	43.6
	60-200	2.0	15.1	20.7	177	0.25	0.85	0.083	47.4
	200-500	0.27	2.3	6.4	170	0.11	0.98	0.024	1.0
	500-1000	0.29	2.6	7.2	162	0.15	0.99	0.024	0.81
	1000-2000	0.38	2.7	7.1	151	0.15	0.94	0.029	1.1
	> 2000	0.22	1.3	2.5	34	0.06	0.21	0.008	0.61
West									
St4	6-60	6.2	34.1	23.2	372	0.21	0.29	0.105	42.0
	60-200	5.7	25.2	43.8	156	0.16	0.19	0.057	25.2
	200-500	0.69	3.6	7.3	143	0.11	0.64	0.025	2.2
	500-1000	0.68	4.1	7.1	128	0.17	0.53	0.024	2.2
	1000-2000	0.59	2.5	5.0	74	0.08	0.26	0.020	1.5
	> 2000	0.67	2.6	3.4	39	0.05	0.08	0.019	1.9
St5	6-60	3.7	57.1	42.4	316	0.63	0.31	0.037	25.3
	60-200	2.0	10.0	17.1	46	0.11	0.15	0.015	11.8
	200-500	0.36	2.2	5.5	75	0.10	0.50	0.007	1.7
	500-1000	0.30	2.0	8.2	135	0.14	0.54	0.010	1.3
	1000-2000	0.34	2.1	5.8	101	0.12	0.47	0.008	1.5
	> 2000	0.11	0.63	1.9	16	0.05	0.13	0.002	0.28
St5b	6-60	4.6	125	86.6	564	1.81	0.61	0.017	208
	60-200	2.1	13.5	10.7	47	0.07	0.09	0.014	14.1
	200-500	0.47	3.3	6.2	80	0.12	0.43	0.006	2.6

Winter 2011

	Size class (µm)	Co	Ni	Cu	Zn	Ag	Cd	Hg	Pb
East									
St10	6-60	8.9	48.6	44.4	158	0.30	0.12	2.350	61.4
	60-200	2.4	17.0	23.6	240	0.17	0.78	0.464	43.7
	200-500	0.28	3.6	43.9	167	0.12	1.2	0.074	5.2
	500-1000	0.29	4.5	11.9	180	0.12	1.1	0.071	5.5
	1000-2000	0.36	5.1	23.5	229	0.10	0.79	0.073	10. 2
	> 2000	(Ns/d)	(Ns/d)	(Ns/d)	(Ns/d)	(Ns/d)	(Ns/d)	(Ns/d)	(Ns/d)
St2	6-60	(Ns/d)	(Ns/d)	(Ns/d)	(Ns/d)	(Ns/d)	(Ns/d)	(Ns/d)	(Ns/d)
	60-200	(Ns/d)	(Ns/d)	(Ns/d)	(Ns/d)	(Ns/d)	(Ns/d)	(Ns/d)	(Ns/d)
	200-500	0.56	5.1	15.8	249	0.15	1.3	0.068	18.1
	500-1000	1.2	9.1	16.3	320	0.19	0.94	0.076	58.5
	1000-2000	(Ns/d)	(Ns/d)	(Ns/d)	(Ns/d)	(Ns/d)	(Ns/d)	(Ns/d)	(Ns/d)
	> 2000	(Ns/d)	(Ns/d)	(Ns/d)	(Ns/d)	(Ns/d)	(Ns/d)	(Ns/d)	(Ns/d)
St3	6-60	8.3	39.4	26.0	158	0.21	0.17	0.286	45.1
	60-200	1.3	59.3	32.3	370	0.23	1.2	0.064	23.6
	200-500	0.46	4.7	15.2	178	0.16	1.1	0.061	4.1
	500-1000	0.38	4.0	12.7	154	0.22	0.73	0.055	3.5
	1000-2000	0.42	3.3	13.7	144	0.21	0.75	0.052	5.8
	> 2000	0.95	6.9	9.1	137	0.14	0.78	0.042	11.0
West									
St4	6-60	8.4	63.7	64.9	241	0.35	0.26	0.144	123
	60-200	2.9	27.0	16.2	133	0.17	0.68	0.052	18.5
	200-500	0.26	2.0	10.5	134	0.14	1.0	0.046	1.0
	500-1000	0.25	2.3	10.0	132	0.15	0.80	0.041	1.1
	1000-2000	0.35	4.7	11.8	125	0.16	0.71	0.042	2.1
	> 2000	1.4	7.9	12.6	170	0.16	0.64	0.049	14.3
St6	6-60	6.4	86.9	35.4	198	0.27	0.17	0.098	34.6
	60-200	1.9	42.2	20.0	127	0.16	0.89	0.052	8.2
	200-500	0.24	3.2	10.1	122	0.09	0.91	0.038	0.62
	500-1000	0.23	2.9	9.6	118	0.11	0.69	0.036	0.72
	1000-2000	0.39	3.2	13.8	119	0.17	0.52	0.038	2.9
	> 2000	0.97	7.5	23.8	121	0.12	0.54	0.037	8.0
St7	6-60	7.6	49.9	28.0	161	0.31	0.40	0.103	65.4
	60-200	2.2	12.4	13.8	119	0.13	0.37	0.033	14.1
	200-500	0.33	1.8	11.9	119	0.15	1.1	0.037	1.2

	500-1000	0.26	2.1	6.7	137	0.14	0.47	0.010	1.7	500-1000	0.41	2.3	9.6	138	0.15	0.96	0.036	2.8
	1000-2000	0.31	2.1	6.4	112	0.14	0.42	0.021	1.9	1000-2000	(Ns/d)	(Ns/d)	(Ns/d)	(Ns/d)	(Ns/d)	(Ns/d)	(Ns/d)	(Ns/d)
	> 2000	0.37	2.4	6.0	89	0.11	0.35	0.008	2.0	> 2000	0.76	1.1	12.6	30	0.04	0.10	0.008	3.5
St7	6-60	4.4	46.5	15.5	504	0.31	0.31	0.036	14.6									
	60-200	4.2	20.0	9.7	53	0.09	0.12	0.038	18.6									
	200-500	0.61	2.9	9.9	159	0.18	0.74	0.013	3.1									
	500-1000	0.75	3.7	6.9	150	0.11	0.42	0.010	6.1									
	1000-2000	0.91	4.3	8.0	121	0.09	0.29	0.010	4.4									
	> 2000	0.30	1.6	2.0	20	0.04	0.07	0.002	1.1									

* Exceptionally, two samples were collected and analyzed for this fraction at this station.

Table S4: Mean bioaccumulation factors (BAFs) calculated per element type (essential vs. non-essential), reported per season (spring (plankton) or summer (fish) 2010 vs. winter 2011) and per biological compartment. Concentrations used for calculations were in mg L^{-1} for seawater (including dissolved concentrations of all the sampled water column), and mg kg^{-1} dry mass for biological compartments (plankton and fish). Nd = Not determined.

BAFs relative to *dry mass*	**Essential elements**				**Non-essential elements**		
	Co	**Ni**	**Cu**	**Zn**	**Cd**	**Hg**	**Pb**
Spring or Summer 2010							
6-60 µm	428 761	232 173	248 653	1 444 604	34 243	511 885	2 129 335
60-200 µm	292 479	82 453	124 069	430 540	34 118	327 532	1 030 065
200-500 µm	29 901	10 552	35 195	517 921	64 622	38 071	64 557
500-1000 µm	28 892	10 911	37 593	600 977	64 151	42 800	73 827
1000-2000 µm	32 648	12 621	36 062	544 352	59 165	44 219	69 376
> 2000 µm	26 415	7 537	19 057	199 331	22 456	24 119	48 085
Sardine (Wh*)	(Nd)	2 232	24 748	348 582	5 324	227 948	8 259
Anchovy (Wh*)	(Nd)	2 913	28 766	472 684	10 495	417 278	4 996
Winter 2011							
6-60 µm	842 765	248 766	230 584	909 054	17 824	(Nd)	2 307 768
60-200 µm	225 671	136 122	122 738	980 543	62 693	(Nd)	755 432
200-500 µm	37 841	14 651	103 889	801 698	87 353	(Nd)	176 687
500-1000 µm	48 845	18 175	67 760	861 720	68 785	(Nd)	420 605
1000-2000 µm	40 434	17 562	91 127	766 240	54 884	(Nd)	183 254
> 2000 µm	109 873	25 221	84 257	569 031	40 781	(Nd)	322 338
Sardine (Wh*)	(Nd)	3 117	30 693	632 459	5 211	(Nd)	18 040
Anchovy (Wh*)	(Nd)	2 788	33 841	660 097	8 238	(Nd)	4 785

*Wh = whole individuals (reconstructed data).

Table S5: Spearman rank order correlation coefficients (r) between mean metal BAFs calculated for the pelagic food web analyzed in the Gulf of Lions. Significant correlations at $p<0.05$ are indicated in bold characters.

	Co	**Ni**	**Cu**	**Zn**	**Cd**	**Hg**	**Pb**
Co	—						
Ni	**0.986**	—					
Cu	**0.909**	**0.926**	—				
Zn	**0.608**	**0.621**	**0.700**	—			
Cd	-0.406	0.403	0.394	0.318	—		
Hg	**0.943**	0.310	0.524	0.333	-0.357	—	
Pb	**0.972**	**0.982**	**0.918**	**0.579**	0.403	0.238	—