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Long-term trends (2002–2016) reveal an increase of mercury levels along with the decline of several metal elements in striped dolphins (*Stenella coeruleoalba*) stranded in the North-West Mediterranean

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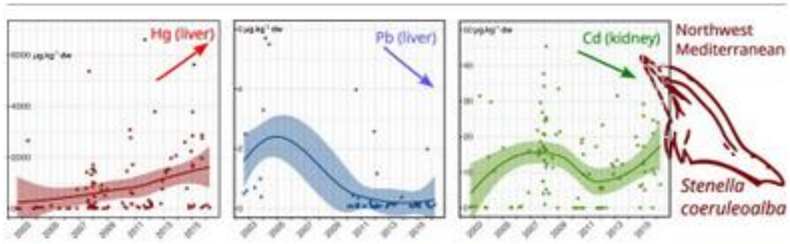
Abstract :

The determination of 18 metals and metalloids was realized in 4 tissues of 62 Stenella coeruleoalba specimens stranded along the French Mediterranean coastline from 2010 to 2016. While most concentrations were comparable to mean worldwide levels, Hg levels were alarming (1190 μg g−1 dw, in average). The results were discussed together with previous measurements in the same area, from 2002 to 2009. The elements Ni and Pb (−81 % and −88 % in liver in 2010–16 compared to 2002–09), and Cd (−40 % in kidney in 2010–16 compared to 2002–09) as well as V (−79 % in liver in 2013–16 compared to 2010–12), showed promising decreasing trends, and the decrease of Zn and Cu levels below baseline values could indicate a global decreasing burden of metal contaminants. In contrary, Hg dramatically increased in dolphins since 2007 (+135 % in liver in 2010–16 compared to 2002–09), regardless of total length. On the other hand, Se levels increased only slightly since 2012, potentially not offering anymore an efficient protection against Hg, with mean Se-to-Hg molar ratios below unity in most tissues (0.26, 0.56, 1.81, and 0.57 in liver, kidney, lung and muscle, respectively).

Graphical abstract



Highlights

► Hg increased by a factor 2.35 in *S. coeruleoalba* livers between 2002–09 and 2010–16 ► Hg increase and Se-to-Hg ratio decrease coincide with a morbillivirus mortality event ► Pb, V, Ni, Zn decreased during past decades in NW Mediterranean *S. coeruleoalba* ► Cd declined continuously since 1989 (−58 %) in stranded *S. coeruleoalba* kidneys

Keywords : Cetacean, Metals, Mercury, Bioaccumulation, Temporal trends, Organic contaminants

1. Introduction

Chemical contaminants originating from human activities have been massively rejected in marine waters since decades. The Mediterranean Sea is particularly concerned by chemical pollution, including metal and metalloid elements, but also organic pollutants such as polychlorobiphenyls (PCBs), polyaromatic hydrocarbons (PAHs), dioxins and furans (PCDD/F), pesticides (…) [UNEP/MAP, 2017]. Many of them remain at alarming levels in marine biota, for instance high PCB levels were still recorded in > 80 % of stranded *Stenella coeruleoalba* tissues between 2002 and 2016 in the French Mediterranean waters [Dron et al., 2022]. Metallic contaminants are also a matter of concern in this region. As an example, the French mussel watch program revealed positive trends in 33 %, 16 % and 30 % of the monitoring stations for Cd, Hg, and Pb, respectively [Briand et al., 2023]. While being an obvious and persistent matter of concern for marine environment and living organisms, high contaminant levels may be also threatening human health through seafood consumption [Stankovic and Jovic, 2012; Traina et al., 2019; Jeanjean et al., 2022]. Regarding environmental and health issues, monitoring chemical contaminants through long-term perspectives is critical to understand the mechanisms and pathways of contaminants through the environmental compartments. Thus, studies concerned by source identification, reactivity, toxicity, impact on immune response to diseases (…) may prevent marine organisms conservation issues and human health risks, and provide stakeholders the keys to reduce efficiently the contamination sources.

Odontocetes are long-lived top predators in the marine environment, and therefore, the evaluation of chemical contaminants in their tissues constitutes a valuable indicator reflecting the global state of contamination of the marine environment through bioaccumulation and biomagnification processes [Delgado-Suarez et al., 2023]. The levels of metals and metalloids are regularly reported in tissues of cetaceans worldwide, and their concentrations in the Mediterranean basin remain critical, with the case of mercury (10 to 100 $\mu g.g^{-1}$ dry weight, in dolphin muscle) being the most studied but also the most alarming [López-Berenguer et al., 2020; Delgado-Suarez et al., 2023]. Also, long-term monitoring of contaminants levels in cetaceans is essential for establishing baseline levels, evaluating toxicological effects thresholds, and verifying the impacts of changing regulations, among other purposes [Borrell et al., 2014; Brown et al., 2015; Capanni et al., 2020; Dron et al., 2022].

The present paper focuses primarily on the levels of 18 metal and metalloid elements measured in 4 tissues (liver, kidney, muscle and lung) of *Stenella coeruleoalba* stranded along the French Mediterranean coast between 2010 and 2016. The analysis of several tissues provides valuable information on accumulation mechanisms and may steer future monitoring strategies [Dron et al., 2022, 2023]. Furthermore, the study includes poorly studied metal elements, for instance molybdenum (Mo) which is, to our knowledge, first reported here in Mediterranean dolphins. The data from previous metal measurements (2002–2009) in stranded *S. coeruleoalba* from the same area [Wafo et al., 2014] were integrated with the more recent results presented here to evaluate and discuss the trends of metals and metalloids over a 15-year time span. Moreover, the same *S. coeruleoalba* specimens, collected in both 2002–09 and 2010–16 periods, also benefited from PCB, pesticides and PAH measurements in the same tissues [Dron et al., 2022, 2023]. These results were also integrated into the interpretations to provide a broader overview of the state of chemical contamination for the *S. coeruleoalba* from the French Mediterranean coasts. Such an overview of several organic contaminants families, together with metal elements, was rarely reported in the literature, but may be very valuable by highlighting potential cocktail effects, clarifying future conservation issues related to a wide panel of pollutants, and potentially improving the understanding of mechanisms involved in the bioaccumulation of contaminants in *S. coeruleoalba*.

2. Materials and methods

2.1. Sampling of stranded dolphins

Metal elements were determined in 4 tissues (liver, kidney, lung, muscle) of 62 *Stenella coeruleoalba* (striped dolphin), stranded along the French Mediterranean coast from February 2010 to April 2016. As described previously for the determination of PCBs, pesticides [Dron et al., 2022], and PAHs [Dron et al., 2023] in the same individuals, the sampling was performed by the National Marine Mammals Stranding Network (RNE) and coordinated by the Mediterranean Cetaceans Study Group (GECEM, now named MIRACETI) [Dhermain et al., 2015; Dhermain, 2016]. The area hosts several major urban centers (Montpellier, Marseille, Toulon, Nice), industrial and military harbors (Fos and Toulon, respectively), and was well described in these previous works [Dron et al., 2022, 2023]. It is divided on the West-East axis in its center by the Rhône river (annual average flow of 1700 m^3 s^{-1}) delta. The stranding sites are reported on a map in Supplementary Information S1. Most of them are located east of the Rhône delta, due to greater dolphin populations associated to deeper waters and coastal steep slopes within the Pelagos Sanctuary, and to facilitated logistics [Dron et al., 2022, , 2023].

The length of the stranded dolphins studied here ranged from 90 cm to 214 cm, mean length was 166.5 cm. At approximately 1.5 years old, corresponding to 120 cm total length [Calzada et al., 1997; Marsili et al., 2004], *S. coeruleoalba* change in feeding habits, switching from lactation to solid food, potentially impacting contaminants bioaccumulation [Calzada et al., 1996]. Therefore, individuals with total length below 120 cm were considered separately as “calves” ($N = 11$ out of 62).

The decomposition states, as defined by IJsseldijk et al. (2019), of the carcasses analyzed here, were mainly fresh (decomposition condition category DCC 1, $N = 30$) and intermediate (DCC 2 and 3, $N = 28$). Only 4 individuals in more degraded decomposition states DCC 4 and 5 were sampled. However, as for organic contaminants [Dron et al., 2022, 2023], Tukey-HSD post-hoc tests showed that DCC had no incidence on metal elements concentrations in any of the studied tissues.

2.2. Metal elements determination and statistical analyses

18 metal elements (Al, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Mo, Cd, Sn, Sb, Pb, Se, Ag, and Hg) were measured in the dolphin tissue samples. A sample aliquot of 0.3 g was dissolved in a mixture of HNO_3 (7.5 mL, 65 % *v*/v) and HCl (2.5 mL, 37 % v/v). Microwave-assisted mineralization (Ethos LabStation) was carried out at 180 °C for 20 min, before ICP-MS analysis (ICAP Q, ThermoElectron). Mercury (Hg) was determined separately, by adding 100 μL of a 10 $\mu g\ L^{-1}$ gold solution prior to mineralization, to form a Hg—Au complex avoiding Hg volatilization. Duplicates were prepared and analyzed for all samples. All the results were expressed as $\mu g\ g^{-1}$ dry weight (dw), detection limits ranged from 0.05 to 0.10 $\mu g\ g^{-1}$ dw and relative standard deviations (RSD) from 5 % to 10 %. Further details on the analytical procedure, its performances and quality check can be found in previous works [Ratier et al., 2018; Dron et al., 2019].

Data handling and statistical analyses were performed with the R software version 4.1 [R Core Team, 2021]. Analyses of variance followed by the Tukey post-hoc tests were realized to evaluate the significance ($p < 0.05$) of differences between variables mean values (aov and TukeyHSD functions, “stats” package). Hierarchical clustering was performed using the hclust function (“stats” package), and its circle representation using “circlize” and “dendextend” packages [Gu et al., 2014; Galili, 2015]. The trend or level stationarity of the temporal variations were evaluated by the Kwiatkowski–Phillips–Schmidt–Shin KPSS test (kpss.test function, “tseries” package) [Kwiatkowski et al., 1992; Dron et al., 2022, 2023]. The visualization of trends was supported by moving regression (geom_smooth function, “ggplot2” package) used with LOESS (locally estimated scatterplot smoothing) at a 0.9 span value and a 0.95 confidence interval.

3. Results

3.1. Distribution of metal elements among tissues and gender

The mean concentrations and ranges measured in the 4 sampled tissues of stranded *S. coeruleoalba* are provided in Table 1, and full data are provided as Supplementary Data. Among the 18 elements investigated here, all were detected in at least 95 % of the different tissue samples except Sb (51 % in liver and kidney, 38 % in lung and 34 % in muscle), Al (82 % in liver and kidney, 88 % in muscle) and V (88 % in muscle).

The concentrations of metal elements were significantly higher in liver than in other tissues for 10 elements, in particular Hg, Ag, Se, Mo, Cu and Mn (Table 1). Hierarchical clustering (Fig. 1) shows that these elements were divided into two groups, whether metal elements were at much higher levels in adults than calves (Ag, Se, Hg) or evenly distributed (Mn, Fe, Cu, Zn, Mo, As). Other elements did not have significantly higher levels in liver, but their concentrations were the most elevated in kidney (Cd), lung (Co, Sn), muscle (Cr) or were homogeneously distributed (Al, V, Ni, Pb).

Table 1

Metal elements concentrations (μg g^{-1} dw) mean values ± standard deviations (min - max) in the analyzed tissues of *Stenella coeruleoalba* stranded along the French Mediterranean coast in the 2010–2016 period (nd = not detected). Bracketed letters correspond to significant differences according to Tukey post-hoc tests, and significantly highest levels are highlighted in bold.

	Liver (*N* = 53)	Kidney (*N* = 49)	Lung (*N* = 16)	Muscle (*N* = 58)
Al	29.4 ± 110 [a] (0.4–707)	5.27 ± 5.86 [a] (0.6–33.3)	11.2 ± 4.39 [a] (1.60–18.4)	13.4 ± 44.9 [a] (1.20–318)
V	0.56 ± 1.19 [a] (0.01–5.58)	0.82 ± 1.71 [a] (0.01–5.36)	0.03 ± 0.02 [a] (nd – 0.08)	0.72 ± 1.60 [a] (nd – 6.64)
Cr	0.23 ± 0.29 [b] (0.02–1.25)	**0.38 ± 0.36 [ab]** **(0.01–1.35)**	**0.25 ± 0.15 [ab]** **(0.05–0.62)**	**0.42 ± 0.44 [a]** **(0.05–1.88)**
Mn	**10.6 ± 5.7 [a]** **(0.54–28.5)**	2.26 ± 1.10 [b] (0.45–5.23)	0.75 ± 0.40 [b] (0.40–2.01)	1.25 ± 1.51 [b] (0.27–10.4)
Fe	**799 ± 448 [a]** **(130–2590)**	490 ± 194 [b] (142–1180)	**906 ± 239 [a]** **(520–1230)**	562 ± 245 [b] (190–1060)
Co	0.04 ± 0.03 [bc] (nd – 0.22)	0.07 ± 0.06 [b] (nd – 0.29)	**0.36 ± 0.19 [a]** **(0.01–0.63)**	0.03 ± 0.02 [c] (nd – 0.10)
Ni	0.25 ± 0.40 [a] (0.02–2.64)	0.23 ± 0.29 [a] (0.03–1.38)	0.16 ± 0.13 [a] (0.03–0.50)	0.37 ± 0.78 [a] (0.01–5.63)
Cu	**26.4 ± 24.0 [a]** **(3.70–163)**	10.3 ± 4.9 [b] (2.70–28.5)	2.66 ± 0.32 [b] (2.20–3.60)	6.31 ± 4.49 [b] (1.50–24.7)
Zn	**82.6 ± 40.8 [a]** **(17.3–221)**	46.0 ± 19.2 [bc] (11.8–97.4)	59.4 ± 13.2 [b] (39.5–90.6)	32.2 ± 21.1 [c] (9.40–111)
As	**4.64 ± 3.65 [a]** **(0.39–15.6)**	2.71 ± 1.68 [b] (0.12–8.61)	1.34 ± 0.49 [b] (0.40–2.13)	1.57 ± 1.55 [b] (0.22–8.51)
Mo	**2.85 ± 2.20 [a]** **(0.06–13.9)**	0.29 ± 0.33 [b] (0.02–2.31)	0.08 ± 0.05 [b] (0.02–0.23)	0.21 ± 0.32 [b] (0.01–1.65)
Cd	3.19 ± 3.79 [b] (0.01–23.1)	**10.0 ± 9.0 [a]** **(0.01–31.6)**	0.29 ± 0.16 [b] (0.04–0.69)	1.82 ± 4.80 [b] (nd – 21.9)
Sn	1.16 ± 0.72 [b] (0.10–3.35)	1.14 ± 0.75 [b] (0.07–4.11)	**2.33 ± 1.88 [a]** **(0.17–8.18)**	**1.59 ± 2.25 [ab]** **(0.05–14.8)**
Sb	0.02 ± 0.05 [a] (nd – 0.20)	0.01 ± 0.02 [a] (nd – 0.08)	0.01 ± 0.00 [a] (0.01–0.02)	0.01 ± 0.02 [a] (nd – 0.11)
Pb	**0.30 ± 0.68 [a]** **(0.04–3.98)**	**0.14 ± 0.16 [ab]** **(0.03–0.96)**	**0.14 ± 0.15 [ab]** **(0.01–0.56)**	0.09 ± 0.11 [b] (0.01–0.59)
Se	**98.3 ± 146 [a]** **(0.90–699)**	11.7 ± 12.7 [b] (0.21–52.0)	**49.9 ± 48.1 [ab]** **(1.45–155)**	9.62 ± 14.6 [b] (0.42–98.0)
Ag	**3.17 ± 2.72 [a]** **(0.02–11.1)**	0.93 ± 1.85 [b] (nd – 8.65)	0.02 ± 0.02 [b] (0.01–0.06)	0.53 ± 1.33 [b] (nd – 6.64)
Hg	**1190 ± 1488 [a]** **(16.0–6610)**	54.6 ± 43.2 [b] (5.00–182)	63.8 ± 31.0 [b] (10.0–111)	45.6 ± 62.3 [b] (10.0–458)

3.1.1. Essential elements (Mn, Fe, Cu, Zn, Mo) and As

The concentrations of essential elements Mn, Fe, Cu and Zn (Table 1) were in the range of recent studies from different regions worldwide [Delgado-Suarez et al., 2023]. These values also correspond to recently defined baselines [Chen et al., 2020]. No correlation with dolphins body length (R^2 < 0.3 in all tissues), neither significant differences towards gender, were observed. Molybdenum (Mo) may be considered as an essential element [Smedley and Kinniburgh, 2017] and had a distribution pattern very similar to Mn. Besides, both metals were strongly correlated in all tissues ($p < 0.01$) except liver. It was here in the midrange of the very few works reporting Mo levels in cetaceans, and to our knowledge limited to the 1975–99 period in northern Pacific, southwestern Atlantic and Baltic waters [Parsons, 1999; Agusa et al., 2008]. The role of As as an essential element remains unclear in cetaceans [Shoham-Frider et al., 2016], however, it appears slightly higher than in other studies [Martínez-López et al., 2019; Delgado-Suarez et al., 2023], but follows the accumulation patterns of Mn, Fe, Cu, and Zn (Fig. 1).

3.1.2. Hg, Ag, and Se

Compared to studies covering the same period (2010–2016), the total Hg levels in *S. coeruleoalba* tissues were in the upper end (muscle) or much higher (liver) than other records from the Mediterranean Sea [Esposito et al., 2020; Sedak et al., 2022; Delgado-Suarez et al., 2023] and higher compared to data from Atlantic and Pacific coasts in muscle [Delgado-Suarez et al., 2023] and other tissues [Gui et al., 2017; Méndez-Fernandez et al., 2022]. On the other hand, while Se is commonly suggested to be involved in Hg detoxification processes [López-Berenguer et al., 2020; Paton et al., 2024; von Hellfeld et al., 2024], its concentrations were comparable or even lower in the present liver samples than in other Mediterranean *S. coeruleoalba* [Sedak et al., 2022; Delgado-Suarez et al., 2023], but higher than in other species from China Sea and New-Zealand [Gui et al., 2017; Stockin et al., 2021]. Even though, significant correlations were observed between Hg and Se in all tissues. In liver and lung tissues, the best correlations, R^2 = 0.66 ($p < 10^{-7}$) and 0.52 ($p = 0.0016$), respectively, were obtained on a log10 basis (compared to 0.58 and 0.47 on linear basis, respectively), and in kidney and muscle on a linear basis with R^2 = 0.50 and 0.73 ($p < 10^{-7}$), respectively (0.47 and 0.39 on log10 basis, respectively). The distribution of Ag towards gender and tissues was very similar to Hg (Fig. 1), and a significant correlation with Se was found in liver (R^2 = 0.52, $p < 0.001$), but not in the other tissues (Table 1). The concentrations of Ag measured in the present work were much higher than recently defined baselines [Chen et al., 2017; Cagnazzi et al., 2020], but in the midrange of former levels recorded in the 1975–99 period [Ikemoto et al., 2004; Agusa et al., 2008]. The elements Hg, Ag and Se were also well correlated to body length in liver tissues, with best results obtained on log10 basis (R^2 = 0.78, 0.40 and 0.50, $p < 10^{-6}$, respectively), but no strong correlation was found in other tissues (R^2 < 0.3). Thus, calves had much lower Hg, Ag and Se levels than adults (Fig. 1).

3.1.3. Cd and other metals

Other metal elements were globally in the same range as observed in Mediterranean *S. coeruleoalba* [Esposito et al., 2020; Sedak et al., 2022; Delgado-Suarez et al., 2023] and from other regions [Gui et al., 2017; Stockin et al., 2021; Méndez-Fernandez et al., 2022; Delgado-Suarez et al., 2023].

The elements Co and Sn had significantly higher concentrations in lung tissues, while Cd was preferentially accumulated in kidney (Table 1), as observed previously in the Mediterranean region [Wafo et al., 2014; Martínez-López et al., 2019]. The median Cd concentration in kidney was also two orders of magnitude higher in adults (9.56 μg g^{-1}) than calves (0.03 μg g^{-1}), and comparable disparities were observed in other tissues with levels higher in adults by a factor of 2.5, 3.6, and 38

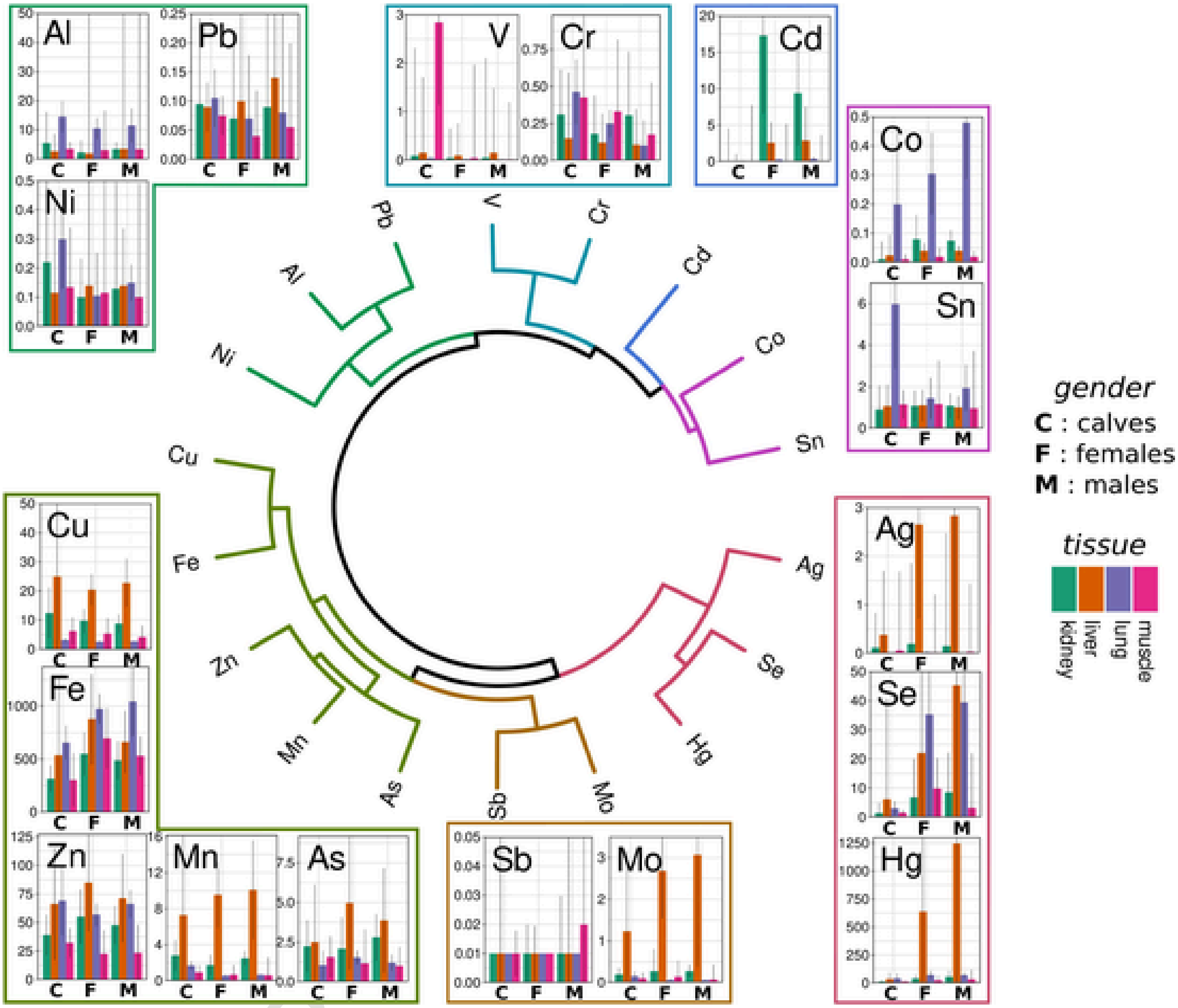


Fig. 1. Circle dendrogram obtained from hierarchical clustering realized with scaled metal concentrations in all tissues (height = 20, k = 7, see also Supplementary Information S2 for classical screeplot representation), and barplots representing median levels (with standard deviations whiskers, μg g^{-1} dw) according to tissues and gender.

in muscle, lung and liver, respectively. Other metal elements (Ni, Al, Pb, V, Cr, Co and Sn) had comparable concentrations in calves, female and male adults (Table 1, Fig. 1).

3.2. Spatial patterns

As indicated in previous works [Dron et al., 2022, 2023], studying the concentrations of contaminants in stranded *S. coeruleoalba* tissues on a geographic basis is submitted to several potential sources of uncertainties and must be taken with caution. The living perimeter of *S. coeruleoalba* is still to be precised, but appears to be composed of relatively sedentary coastal populations, particularly in deep coastal waters such as in the east half of the study area [Gannier, 1999; Gannier et al., 2022], as well as free-ranging dolphin groups possibly more often encountered in offshore waters [Gaspari et al., 2007; Gaspari et al., 2019]. As carcasses and impaired dolphins can drift over long distances with currents, stranding sites may not necessarily reflect living areas [Peltier et al., 2020; Deslias, 2021]. In the present case, it can also be noticed that sampling locations were not homogeneous with time, mainly in the most anthropized areas of Nice, Marseille and Fos before 2013, and in less urbanized areas after 2014 (Supplementary Information S1).

Globally, the highest concentrations of most metals were found in tissues from the most anthropized areas of Fos-Marseille, Toulon and Nice. The Fig. 2 and Supplementary Information S3 show that the elements Hg, Cr, Cu and in a lower extent Mn were higher in liver samples collected from individuals stranded in the Fos industrial area. This observation can be extended to the groups of elements identified by hierarchical clustering (Fig. 1). It is consistent with the industrial activities and the high Hg, As, Cu, Mn, Mo levels measured in fish in these areas [Dron et al., 2019]. Liver tissues also showed high Pb and Cu levels in the Toulon area, where Pb and Cu pollution from the military harbor has been well documented [Tessier et al., 2011]. On the other hand, muscle tissues were more contaminated by most metal elements in Nice and Marseille (Fig. 2, Supplementary Information S3). This discrepancy between liver and muscle tissues was similar for PAHs, and could derive from different integration time and accumulation mechanisms between

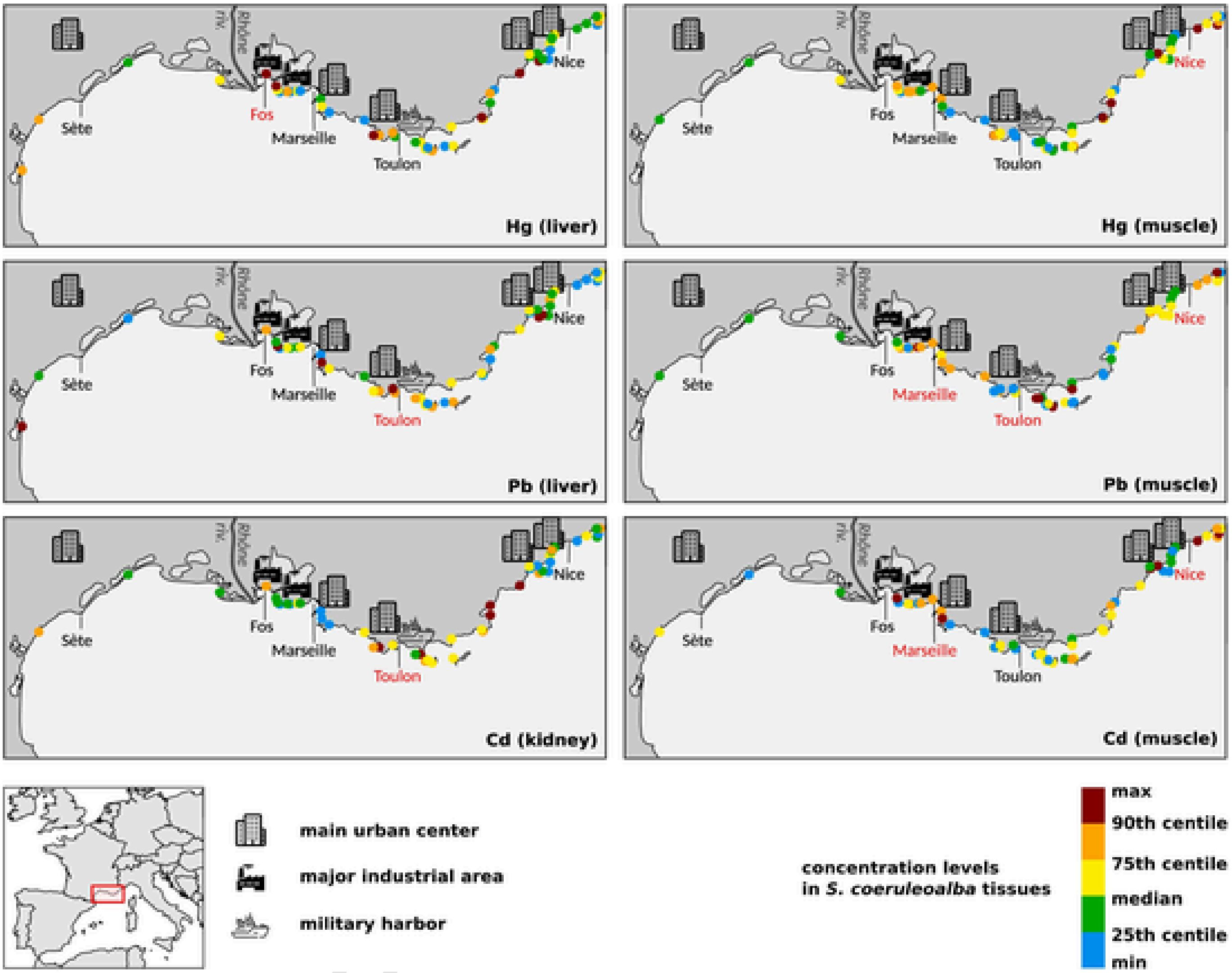


Fig. 2. Geographical distribution of Hg, Pb and Cd concentrations in liver (Hg, Pb) or kidney (Cd) and muscle tissues of *S. coeruleoalba* along the French Mediterranean coastline in 2010–16. Red text underlines the areas where the highest metal levels were observed.

these two tissues [Dron et al., 2023]. Finally, Cd levels in kidney were higher between Toulon and Nice, where urbanization is more limited. Consistently, Cd was reported at moderate levels in environmental studies caried out in the Fos industrial area [Dron et al., 2019; Jeanjean et al., 2022].

4. Discussion

4.1. Long term trends of metal concentrations in S. coeruleoalba tissues

Among the 18 metal elements monitored in 2010–2016 in liver, kidney, muscle and lung tissues, 10 had been previously analyzed in the same conditions over a former set of samples from tissues of *S. coeruleoalba* stranded in 2002–2009 in the same geographical area [Wafo et al., 2014]. These data were available for at least 50 % of the 55 individuals studied for 8 elements in all of the same 4 tissues (Cr, Mn, Fe, Cu, Zn, Cd, Se and Hg) and 15–22 % for Ni and Pb, in all tissues except muscle (no data). The detailed results of the KPSS stationarity/trend tests for the whole 2002–2016 period are listed in Supplementary Information S4. Early metal elements analyses in various tissues of *S. coeruleoalba* stranded along the French Mediterranean coast by Augier et al. [2001] in 1989–1991 and 1997–1998 were also partially available (detailed data available only for the 1997–1998 period), but they did not totally cover the study area (limited to the east in 1989–1991 and to the west shores in 1997–1998). They were thus considered for discussion but not integrated in the KPSS tests, neither in graphical representations.

The concentration levels in the tissues of *S. coeruleoalba* of 10 elements out of 18 were stationary over the 2002–2016 period, including Al, Cr, Co, As, Sn, Sb, and Ag, as well as most of the essential elements (Mn, Fe, and Cu) which remained in the range of former studies worldwide [Honda et al., 1983; Muir et al., 1988; Monaci et al., 1998; Augier et al., 2001; Wafo et al., 2014]. These essential elements were also homogeneous among size and gender (Fig. 1), corroborating the hypothesis that physiological needs (growth, oxygen transport, hormones regulation, bone development…) and homeostasis are their main regulation factors [Honda et al., 1983; Chen et al., 2020]. There was no specific contamination or, at least, these elements were well regulated by *S. coeruleoalba* in the range of concentrations encountered along the French Mediterranean coasts.

Only 4 metal elements, V, Ni, Zn and Pb showed clear decreasing trends. In particular, Pb levels dropped by −88 % to −96 % (among the tissue), between the 2002–2005 and the 2010–2016 periods (Fig. 3). This strong fall-off may be related to the ban of lead in fuels, progres-

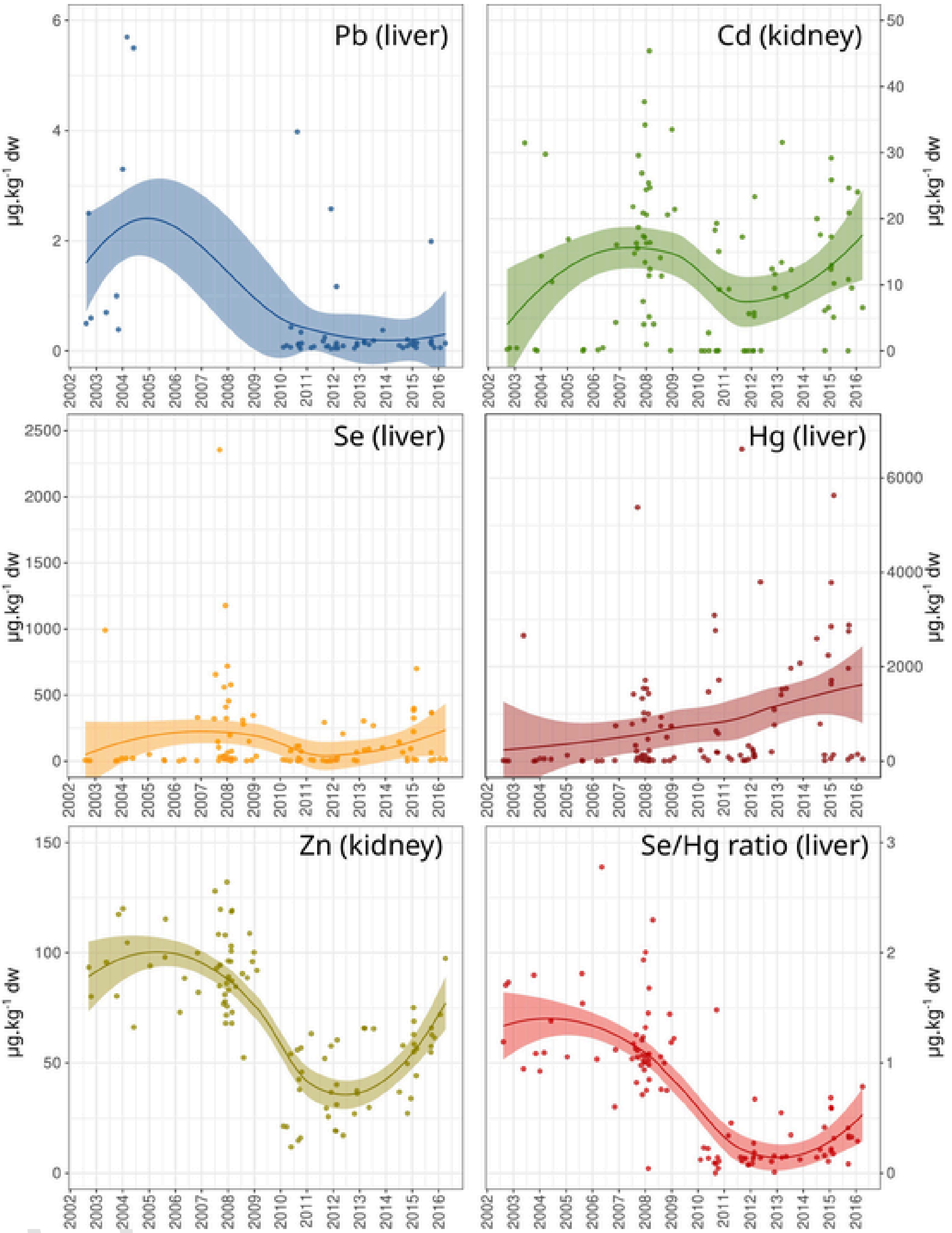


Fig. 3. Metal concentrations in liver (Pb, Se, Hg) and kidney (Cd, Zn) tissues from *S. coeruleoalba* stranded along the French Mediterranean coast over the 2002–2016 period, and Se/Hg ratio in liver. Curves are issued from LOESS smoothing (95 % confidence interval and span = 0.9).

sively implemented between 1990 and 2010 in Mediterranean countries [Ritchie and Roser, 2022]. The Pb levels decreased in the late 90's in the western Mediterranean atmosphere [Migon et al., 2008; Austruy et al., 2019], but the results of mussel watch programs were discordant with a concomitant decline of Pb in mussel from Spanish waters [Santos-Echeandía et al., 2021], while an increase was observed in mussel from the French Mediterranean [Briand et al., 2023]. However, a strongly decreasing trend was measured in the dolphins from the French Mediterranean coastline during the 2000s, as observed in *Delphinus delphis* (common dolphin) stranded along French Atlantic coasts for instance [Méndez-Fernandez et al., 2022]. Former levels measured in 1989–1991 (mean 0.49 μg.g^{-1} dw) were comparable to 2002–2005 results, and those of 1997–1998 (mean 0.05 μg.g^{-1} dw) much lower [Augier et al., 2001]. The individuals from the 1997–1998 all originated from the west part of the studied area, possibly less subjected to Pb contamination due to moderate urbanization or to a higher trophic dilution associated to higher primary production in this area. A very similar trend was observed here for Ni (decrease by 61 to 93 % among tissue), and a continuous decline of Ni in *S. coeurleoalba* tissues is revealed since the former 1989–1991 and 1997–1998 (mean 3.50 and 0.58 μg.g^{-1} dw, respectively) [Augier et al., 2001]. The analyses of V were limited to 2010–2016, but showed singularly high concentrations above 1 µg.g^{-1} dw for several dolphins in 2010–2012, followed by lowered and homogeneous levels, in liver, muscle and kidney (all <0.5 µg.g^{-1} dw) (Supplementary Data). Finally, the essential element Zn decreased by −29 % to −47 % in 2010–2016 compared to 2000–2009, which levels were comparable with 1997–1998 and 1989–1991 analyses [Augier et al., 2001]. This decrease appeared as a sudden drop in 2009–2010, particularly intense in kidney and muscle tissues (Fig. 3) and further discussed in the following sections 4.2.1 and 4.2.2.

The trends of Cd differed among tissue. In liver tissues, Cd levels marked a slight peak in 2007–2008. The mean concentration measured in 2010–2016 is comparable to the former 1997–1998 measurement though, and lower to the earlier 1989–1991 analyses [Augier et al., 2001]. This peak was followed by a strong decrease in 2010–2013, but since, Cd concentrations seem to increase back again (Fig. 3). The results in kidney, which is the most accumulating tissue for Cd in cetaceans, indicate a continuous decline of Cd (−58 %) since 1989–1991 (mean 24.0 µg.g^{-1} dw) and 1997–1998 (mean 17.2 µg.g^{-1} dw) [Augier et al., 2001], down to 14.9 µg.g^{-1} dw in 2002–2010 and finally 10.0 µg.g^{-1} dw in 2010–2016 (Table 1). The 2010–2016 levels are significantly lower than the 2002–2010 (Tukey post-hoc test $p = 0.02$). Comparisons with older values from Augier et al. [2001] were limited to *t*-test with 2002–2010 levels because only mean and ranges were available. Then, the 2002–2010 levels were significantly lower than the 1989–1991 mean (t-test $p = 1.4 \times 10^{-6}$) but not compared to the 1997–1998 mean value (t-test $p > 0.1$).

On the other hand, a strongly increasing trend was observed for Hg since 2007, with mean concentrations in liver rising from 235 ± 672 µg.g^{-1} dw in 2002–2007 to 1189 ± 1488 µg.g^{-1} dw in 2010–2016 (liver medians increasing from 14.2 to 323 µg.g^{-1} dw, respectively, Fig. 3). Similar increasing trends were measured in other tissues. While the 2002–2007 level confirmed a decreasing trend formerly observed by Augier et al. [2001] (1657 µg.g^{-1} dw in 1989–1991 and 332 µg.g^{-1} dw in 1997–1998), the strong increase measured in 2010–2016 almost restored the former 1989–1991 Hg contamination state. Interestingly, a similar, relatively sudden and severe, increase in Hg liver concentrations was also observed in *D. delphis* stranded along the French Atlantic coasts, occurring during the same period, between 2005 and 2008 [Méndez-Fernandez et al., 2022]. The Hg levels measured in 2002–2007 were lower than what measured in stranded *S. coeruleoalba* from the nearby Spanish Mediterranean at this period (571 ± 607 µg.g^{-1} dw), but the most recent 2010–2016 levels were close to the former 1990–1993 concentrations in dolphins from this Spanish area (989 ± 804 µg.g^{-1} dw) [Borrell et al., 2014]. Recent results from more southern Spanish Mediterranean waters (2009–2016) suggest that Hg remained stable since 2007 in *S. coeruleoalba* from this area (approx. 558 ± 649 µg.g^{-1} dw) [Martínez-López et al., 2019], in contrary to those from French Mediterranean as revealed in the present study. However, post-2010 results from the northern Spanish Mediterranean coast would be interesting to confirm this recent discrepancy between French and Spanish Mediterranean *S. coeruleoalba* populations. It is noteworthy that, in the present study, the Hg levels were higher in the 2010–2016 period regardless of dolphins total length (*i.e.* age), as shown in Fig. 4. The strong elevation of Hg levels is all the more of concern, since the concentrations of Se implied in the MeHg detoxification process in cetaceans did not increase proportionately [Kershaw and Hall, 2019; López-Berenguer et al., 2020]. After a peak in 2007–2008, Se levels dropped back to their initial 2002–2006 levels and only slowly increased since 2013 (Fig. 3). Finally, Mo also showed slightly but significantly increasing trends in liver and kidney since its first measurements in 2010 (Supplementary Information S4).

4.2. Comparison with other high trophic level marine species

Odontocetes are long-living top predators, and thus particularly subjected to contaminant bioaccumulation. The concentrations of Hg measured in the muscle of the stranded *S. coeruleoalba* sampled in this study were in the range of other odontocetes, globally 10–100 µg g^{-1}. This is slightly above what measured in top predator sharks, tuna and swordfish (5–15 µg g^{-1}), and greater than other carnivorous species such as whales, conger and hake (0.5–5 µg g^{-1}), as reported in Table 2. Thus, the higher accumulation of Hg in odontocetes may not be strictly related to diet and age. It should also involve other processes, possibly related to the storage of Hg as HgSe nanoparticles in their tissues, for in-

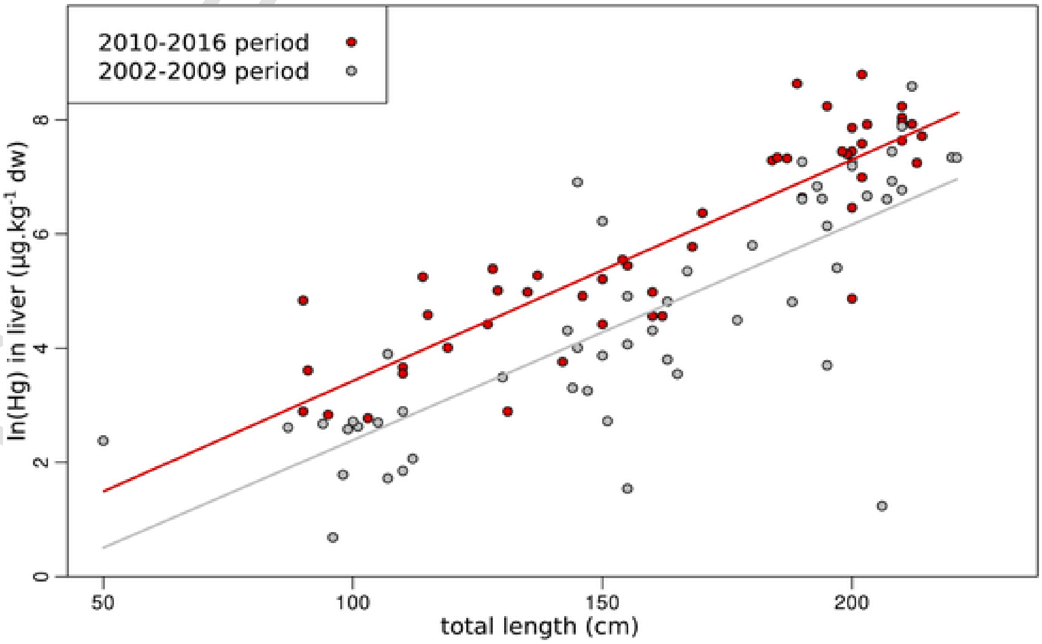


Fig. 4. Hg concentration levels (ln scale) among total length for dolphins stranded in 2002–2009 (grey circles) and 2010–2016 (red circles), and corresponding linear fit lines ($R^2 = 0.61$ and 0.78, respectively, $p < 0.001$).

Table 2
Hg, Cd, and Pb concentrations (mean and standard deviation, or range, μg g^{-1} dw) measured in muscle tissues of high trophic level (> 4) cetaceans and fishes.

Specie	Region	Year	Hg	Cd	Pb	Ref
Top predators – cetaceans						
S. coeruleoalba	Med. NW	2010–16	45.6 ± 62.3	1.82 ± 4.80	0.09 ± 0.11	this study
S. coeruleoalba	Med. NW	2002–09	26.4 ± 36.0	0.07 ± 0.05	1.53 ± 1.25	1
T. truncatus⁎	Med. Adriatic	2000–02	114 ± 122	0.04 ± 0.04	0.06 ± 0.04	5
G. griseus⁎	Med. Adriatic	2000–02	171 ± 98	0.35 ± 0.18	0.02 ± 0.04	5
G. melas⁎	Atlantic NE	2012	10.9 ± 6.9	0.42 ± 0.27	NA	6
P. macrocephalus⁎	Med. Adriatic	2014	13.3 ± 4.6	0.08 ± 0.04	< 0.04	8
P. macrocephalus⁎	Atl. E	2000–17	NA	0.80 ± 0.27	0.11 ± 0.04	7
Top predators – sharks						
H. griseus⁎	Med. Aegean	2015–16	4.80 ± 4.00	0.04 ± 0.04	2.76 ± 5.60	10
H. perlo⁎	Med. Aegean	2015–16	10.4 ± 6.4	0.12 ± 0.04	0.20 ± 0.08	10
P. glauca⁎	Med. Aegean	2015–16	9.60 ± 4.80	0.01 ± 0.01	1.40 ± 2.48	10
C. carcharias	Pacific	2016–18	13.1 ± 5.8	NA	NA	13
Top predators – other fish						
T. thynnus (farm)⁎	Med. Ionian	2010–11	3.48 ± 1.20	NA	NA	9
T. thynnus	Atlantic Cadiz	2017	NA	<0.10–0.32	<2.9–3.3	11
T. thynnus⁎	Med.	2005–18	0.80–3.36	NA	NA	12
X. gladius⁎	Med.	2005–18	0.28–2.48	NA	NA	12
Carnivorous – whales						
B. physalus	Med.	2000–15	0.32–0.65	0.01–0.04	0.05–0.20	14
Carnivorous – small sharks						
G. melastomus	Med. NW	2012	NA	0.01 ± 0.01	0.06 ± 0.07	2
S. canicula	Med. NW	2012	NA	0.01 ± 0.01	0.03 ± 0.02	2
Carnivorous – other fish						
C. conger	Med. NW	2012	1.46 ± 1.13	< 0.05	< 0.05	3
M. merluccius	Med. NW	2013	0.70 ± 0.91	NA	NA	4
S. sarda	Atlantic Cadiz	2017	NA	<0.10–0.30	<2.9–9.8	11
K. pelamis	Atlantic Cadiz	2017	NA	<0.10–0.30	<2.9–4.6	11

1. Wafo et al., 2014; 2. Mille et al., 2018; 3. Dron et al., 2019; 4. Cresson et al., 2015; 5. Bilandžić et al., 2012; 6. Gajdosechova et al., 2016; 7. Lozano-Bilbao et al., 2021; 8. Squadrone et al., 2015a, 2015b; 9. Milatou et al., 2023; 10. Roubie et al., 2024; 11. Pintado-Herrera et al., 2024; 12. Goyanna et al., 2023; 13. Le Croizier et al., 2022; 14. Delgado-Suarez et al., 2023

⁎ concentrations converted to dw assuming 75 % water content, NA = Not analyzed.

stance, which is very specific to cetaceans and for which insights have been very recently developed [Paton et al., 2024; von Hellfeld et al., 2024]. The levels of Cd in muscle tissues of *S. coeruleoalba* and other odontocetes follows what observed for Hg (Table 2). It should be noted that the great Cd mean level recorded in the present study is due to extreme concentrations measured in 9 individuals stranded between 2010 and mid-2012 (2.77 to 21.9 μg g^{-1}). Since 2013, the mean level came back to 0.05 ± 0.03 μg g^{-1}, *i.e.* below 2002–09 mean level. This is in the range of top predator cetaceans and sharks, but above other high trophic level carnivorous species (Table 2), thus similar conclusions to what indicated for Hg can be drawn. Lately, some works suggested that the metabolic pathways of Cd and Hg may have similarities, such as the formation of Cd—Se complexes [Gajdosechova et al., 2016]. On the other hand, biomagnification of Pb through trophic levels is not systematically established, in particular when it comes to top predators [Sun et al., 2020; Madgett et al., 2021], and may depend on foraging depth, or environmental parameters, among other [Le Croizier et al., 2016 and Le Croizier et al. 2022]. Comparing the Pb levels in the *S. coeruleoalba* stranded in the NW Mediterranean with other high trophic level species, it shows that Pb is very variable but globally at low levels in all of the studies reported here (Table 2). It is still noteworthy that high Pb levels mainly occur in top predators while it is often not detected in lower trophic level carnivores.

4.3. *Implications for toxicological aspects and cocktail effects*

4.3.1. *Mercury and other metal elements*

The concentration levels of mercury (Hg) were pretty alarming in the tissues of the stranded *S. coeruleoalba* investigated here, and the situation is worsening since they are continuously increasing since 2007–08 (Fig. 3). With minimal concentrations of 16, 5, 10 and 10 μg g^{-1} dw in liver, kidney, lung, and muscle, respectively, Hg levels could have induced potential risks of neurological damage or hepatic toxicity among other, for a large proportion of individuals [Rawson et al., 1993; Gajdosechova et al., 2016]. Since Se is suggested as a demethylation pathway for MeHg through the formation of SeHg particles in cetaceans tissues, the Se-to-Hg molar ratio is often used as an indicator of the toxicological potential of Hg in marine mammals [Kershaw and Hall, 2019; El Hanafi et al., 2023; Paton et al., 2024; von Hellfeld et al., 2024]. The toxicological risk is considered to raise when the Se-to-Hg molar ratio comes below unity [Ikemoto et al., 2004; Burger and Gochfeld, 2013; Gajdosechova et al., 2016; Paton et al., 2024]. Also, the Se-to-Hg ratio showed a sudden decrease in 2009–2010, and nearly all (98 %) of the liver samples had Se/Hg < 1, indicating that Se levels could not be sufficient to produce an efficient protection against Hg. The mean ratios were 0.26, 0.56, 1.81, and 0.57 in liver, kidney, lung and muscle, respectively, with only 1 out of 53 above unity in liver samples, 4 out of 49 in kidney, 12 out of 16 in lung, and 12 out of 51 in muscle. This sudden decrease of the Se/Hg ratio in 2009–2010 was concomitant to the quick decrease of Zn levels, and also followed the triggering of increasing Hg levels (Fig. 3). The concomitance of the decrease in Zn, measured by ICP/MS chemical analysis, and

Se/Hg molar ratio, which is the result of a calculation involving species with opposite trends (Hg, Se), discards the possibility of an analytical bias.

The Hg levels in the stranded dolphins began to raise in 2007–2008, simultaneously to a documented morbillivirus epizootic event [Keck et al., 2010], and 1–2 years before the sudden decreasing of the Se-to-Hg ratio. The tight chronology of these different observations questions about possible causal connections. As it has been suggested for PCBs and other organochlorine pollutants [Dron et al., 2022], Hg may also impair the cetaceans immune capacities thus favoring their susceptibility to infections such as the morbillivirus [Kershaw and Hall, 2019], or may even have directly caused the stranding of the most contaminated specimens. On the other hand, no particular contamination event implying a significant discharge of Hg has been recorded to our knowledge in northwestern Mediterranean before or during this period. However, the contaminants data available here is not sufficient to evaluate potential hypotheses on the Hg increase. The peculiar and sudden aspect of the drop in the Se/Hg molar ratio also remained unexplained. Further research is clearly needed to better understand and consider variations in Hg contamination and its toxicological impacts, especially since Hg remains at high levels in cetaceans worldwide, particularly in the Mediterranean Sea [Shoham-Frider et al., 2016; Tian et al., 2021; Delgado-Suarez et al., 2023] and that the significance of estimating health risks from Se-to-Hg ratio is still under discussion [Burger and Gochfeld, 2013; Gajdosechova et al., 2016]. The further study free-ranging individuals could also improve our understanding of the toxicological implications of high Hg levels in stranded animals.

Among other metal elements, Cd remained at relatively moderate levels since 2002, except for 9 individuals in 2010–12. Considering the few works mentioning Cd risk levels in odontocetes [Chen et al., 2017], it suggest that Cd does not seem to be a major threat to the stranded dolphins investigated here. However this should be reviewed in the future, in light of more precise toxicological studies. As indicated previously, Pb levels decreased considerably since the early 2000's, and this can be considered a true gain from international policies regarding Pb in fuel. The detoxification processes for many toxic trace elements imply metallothioneins (MTs), which are also regulated by essential elements such as Zn and Cu [Das et al., 2000; Rahman et al., 2019]. Also, the decline of Zn and Cu levels could be a sign of decreasing MTs, and thus a possible consequence of a global decreasing burden of other metal contaminants including, for instance, the aforementioned declining, V and Ni [Méndez-Fernandez et al., 2014].

4.3.2. Global contaminants burden

Concerning metal elements, the present study showed particularly alarming results towards Hg and the evolution of its concentrations in the stranded dolphins tissues, but in the same time, promising trends concerning several other elements, among which Pb, Cd, V, Ni, Zn, and Cu. Also, this determination of metal elements in stranded *S. coeruleoalba* was realized in dolphin tissue samples that were also investigated for organochlorine contaminants (PCBs, pesticides), and PAHs, which results were previously published [Dron et al., 2022, 2023]. Since two decades, substantial efforts were realized to maintain and even improve the monitoring of analyzed chemical contaminants in cetaceans, both in qualitative and quantitative terms, with an average of chemical pollutants increasing from 8.9 per individual before 2009 to 11.0 since 2010. It appears that among Hg, the bioaccumulation of PCBs was a strong matter of concern in the *S. coeruleoalba* stranded along the French Mediterranean shoreline [Dron et al., 2022]. Even though, organic contaminants and metals were nearly never correlated in any tissue (Supplementary Information S5), this situation still points out the question of the cocktail effects of chemical contaminants towards cetacean health, and their potential incidence on toxicity. It also suggests that most individuals come under a high exposure to at least one contaminant. While some pesticides showed significantly decreasing trends in these dolphins, PCBs and DDT metabolites decreased only very little since their ban in the late 70's, and PAH remained stable since the past years [Dron et al., 2022, 2023]. Consequently, and in addition to conclusions regarding Hg, it goes without saying that cetacean populations undergo a continuous, persistent, and significant threat due to their exposure to chemical contaminants released in the marine environment by human activities. Unfortunately, in the light of the temporal trends observed for Hg, PCBs, and some pesticides, it should still remain for decades if more effective actions to reduce their concentrations are not considered. While recent significant progress have been made towards the understanding of Hg detoxification pathways [El Hanafi et al., 2023; Paton et al., 2024; von Hellfeld et al., 2024] and towards the toxicological impacts of PAHs [Godard et al., 2006; López-Berenguer et al., 2023] and PCBs [Williams et al., 2020], research should still be supported and pursued concerning the routes for bioaccumulation, the definition of concentration baselines, and the toxicological risks or potential thresholds towards the contaminants burden of cetaceans. These are essential for the further application of effective and concerted policies.

5. Conclusions

An improving effort is being deployed along the French Mediterranean coastline for the monitoring of chemical contaminants in tissues of stranded cetaceans. The determination of 18 metal and metalloid elements is reported here in 62 specimens of *S. coeruleoalba* stranded between 2010 and 2016. These results were further discussed with organic contaminant levels (PCBs, pesticides, PAHs) analyzed in the same samples and previous measurements from the 2002–2009 period, providing a 15-year trend.

While several metal elements show encouraging decreasing trends (Pb, V, Ni, Cd), the concentrations of Hg increased to alarming levels since 2007, coinciding with a morbillivirus epizootic event. At the same time, Zn and Se-to-Hg molar ratio went through a severe and persistent falloff.

The large resources devoted to the analysis of a large panel of contaminants in French Mediterranean stranded dolphins have proven to be particularly essential and meaningful, highlighting the persistent nature of chemical contamination through the marine environment. It also pointed out questions upon the living habits and the state of conservation of *S. coeruleoalba* in this area, requiring supplementary knowledge through genetic and biological analyses to better understand the mechanisms involved and leading to the contaminants variations observed.

CRediT authorship contribution statement

Julien Dron: Writing – original draft, Visualization, Methodology, Investigation, Data curation, Conceptualization. **Emmanuel Wafo:** Writing – review & editing, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Conceptualization. **Florence Chaspoul:** Investigation, Formal analysis, Data curation. **Pierre Boissery:** Writing – review & editing, Validation, Supervision, Resources, Project administration. **Frank Dhermain:** Writing – review & editing, Methodology, Investigation, Funding acquisition, Data curation, Conceptualization. **Marc Bouchoucha:** Writing – review & editing, Resources, Project administration, Funding acquisition. **Philippe Chamaret:** Supervision, Resources. **Daniel Lafitte:** Supervision, Resources.

Uncited reference

Calzada et al., 1997

Declaration of competing interest

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

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Data availability

All data are available in a separate CSV file.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2024.177741.

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Long-term trends (2002-2016) reveal an increase of mercury levels along with the decline of several metal elements in striped dolphins (*Stenella coeruleoalba*) stranded in the North-West Mediterranean

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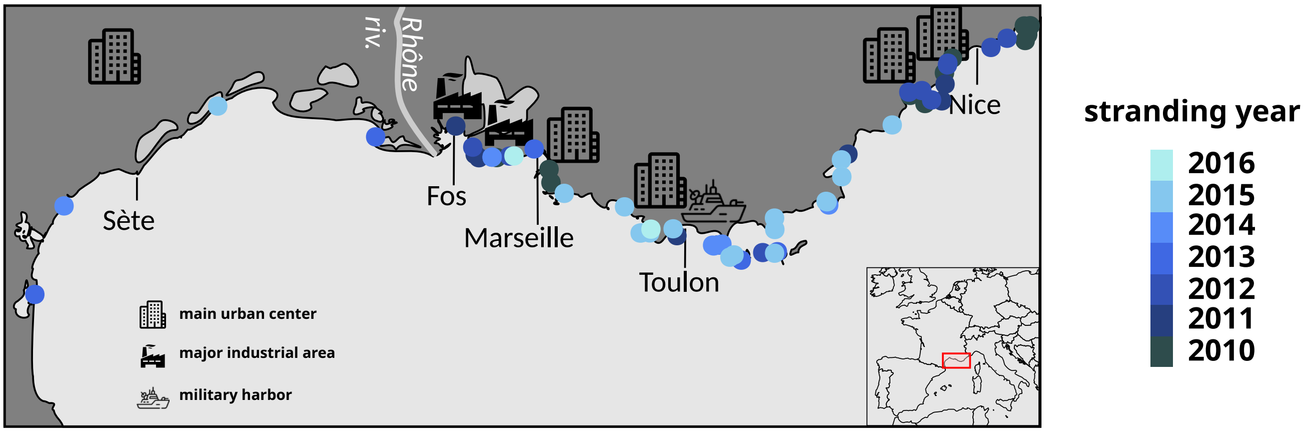
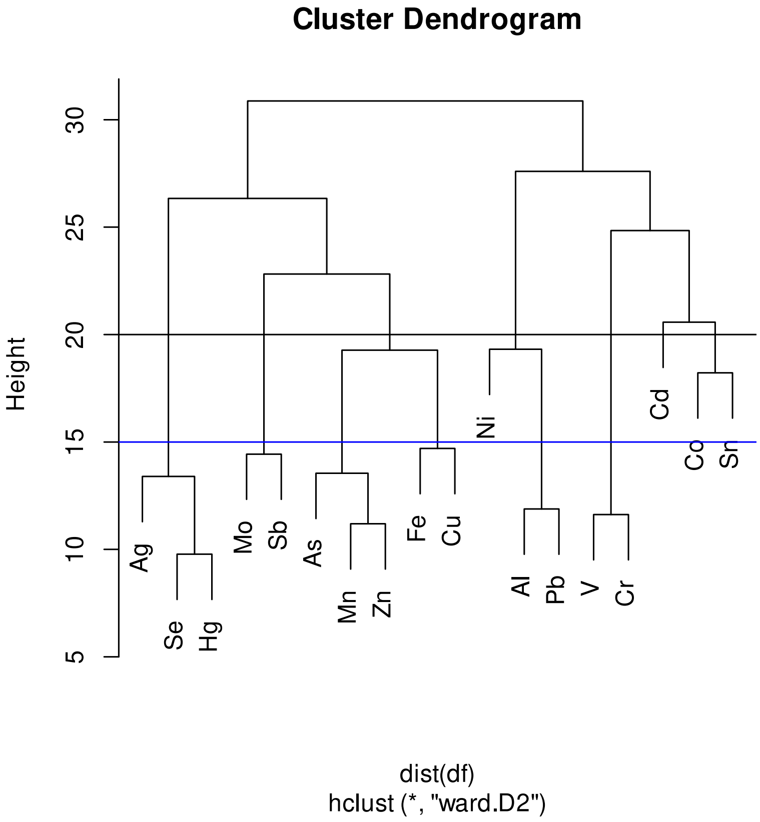
SUPPLEMENTARY INFORMATION

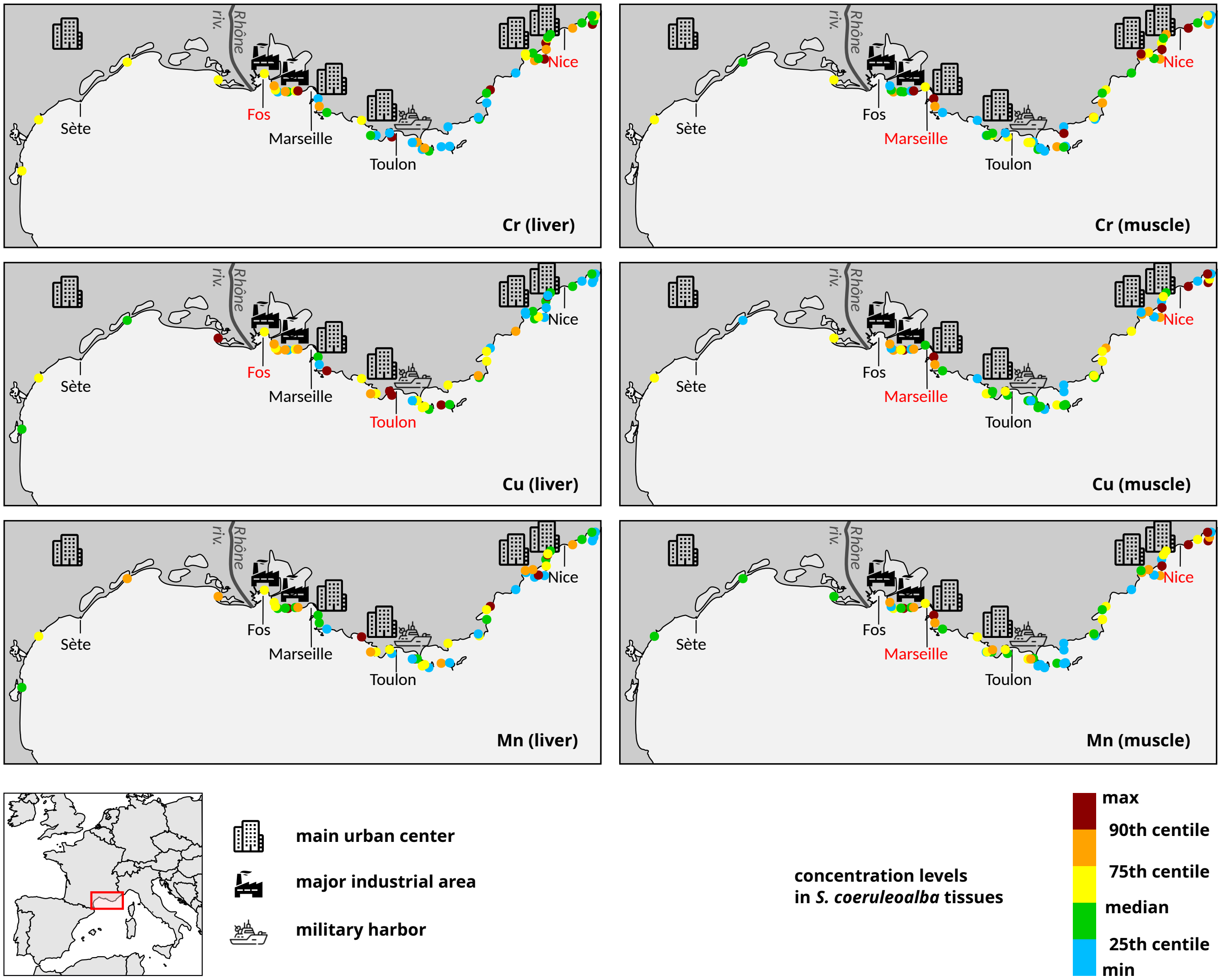
Supplementary Information S1. Stranding timeline from 2010 to 2016, represented geographically.



Supplementary Information S2. Straight classical screeplot representation of the dendrogram obtained from hierarchical clustering realized with scaled metal concentrations in all tissues (height = 20, k = 7).



Supplementary Information S3. Geographical distribution of Cr, Cu and Mn concentrations in liver and muscle tissues of *S. coeruleoalba* along the French Mediterranean coastline in 2010-16. Red text underlines the areas where the highest metal levels were observed.



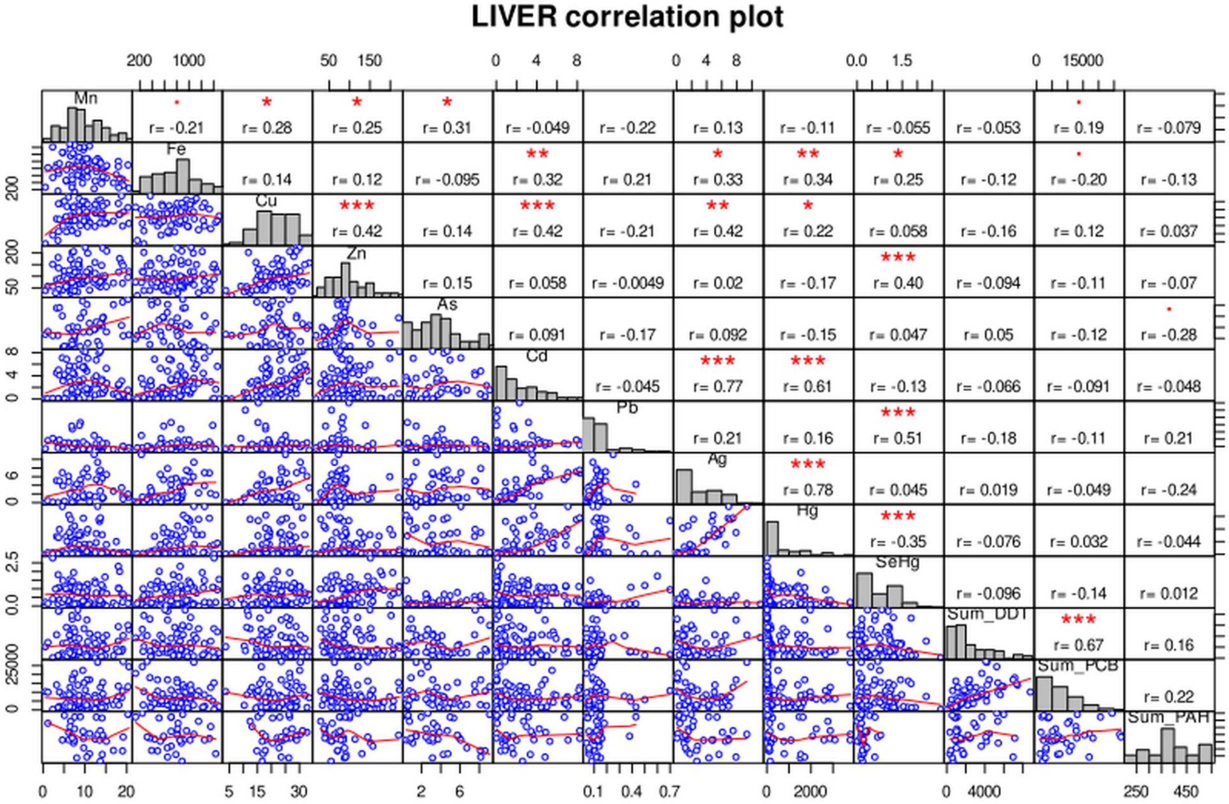
Supplementary Information S4. Results of KPSS-test, p-values obtained for level and trend stationarity tests and conclusion of the test. To precise the results, the evolution of median values over the 2002-2009 and 2010-2016 are compared (results indicated as % of evolution). NA indicates that no data was available in the 2002-2006 period to compare with. Then, and when needed, graphical observations complete the KPSS conclusions and median comparisons.

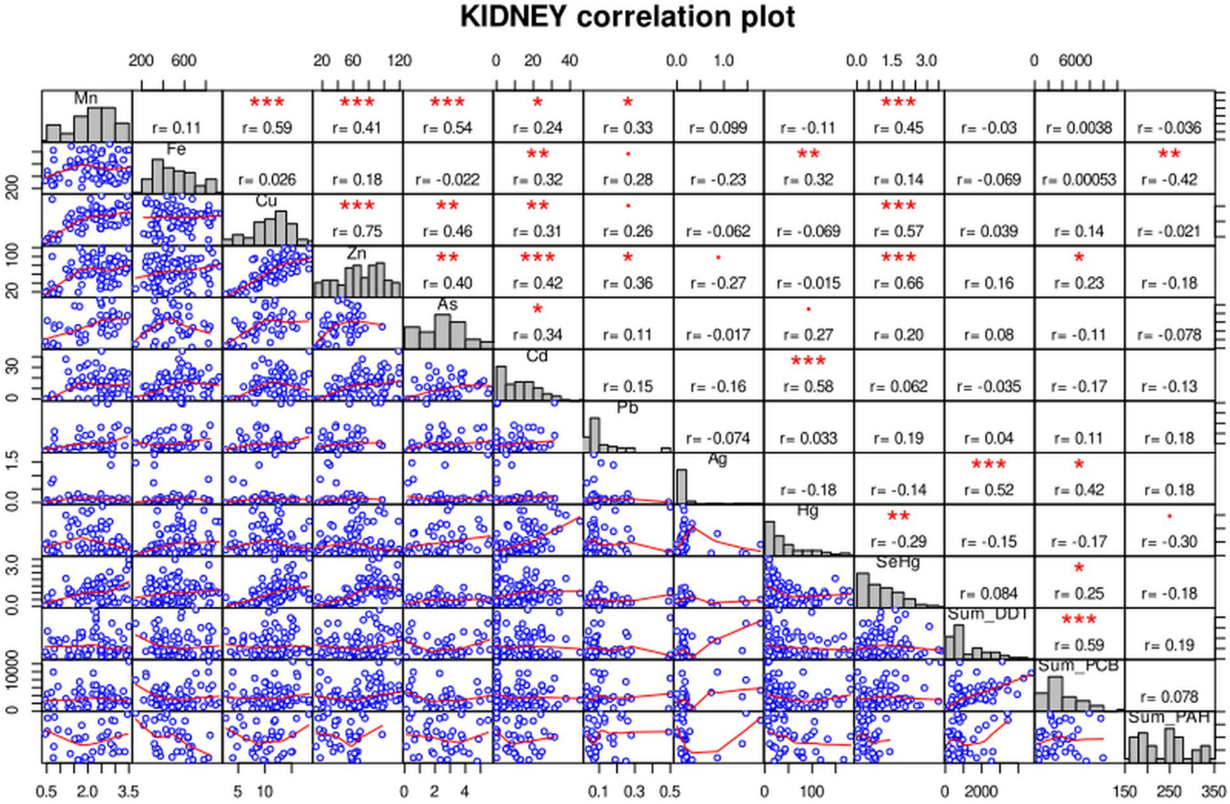
	tissue	**N**	**level**	**trend**	**conclusion**	**2002-09 vs 2010-16**	**Graphical observations**
Al	liver	47	>0.1	>0.1	stationary	NA	no variation
	kidney	41	>0.1	0.07	stationary	NA	no variation
	lung	17	>0.1	0.09	stationary	NA	no variation
	muscle	51	>0.1	>0.1	stationary	NA	positive
V	liver	53	**0.05**	>0.1	**trend**	NA	negative
	kidney	49	**0.02**	>0.1	**trend**	NA	negative
	lung	15	>0.1	>0.1	stationary	NA	no variation
	muscle	51	**0.01**	>0.1	**trend**	NA	negative
Cr	liver	87	>0.1	>0.1	stationary	-10 %	
	kidney	86	>0.1	>0.1	stationary	53 %	
	lung	48	>0.1	>0.1	stationary	-9 %	
	muscle	70	>0.1	>0.1	stationary	18 %	
Mn	liver	98	>0.1	>0.1	stationary	12 %	
	kidney	93	**0.01**	>0.1	**trend**	-1 %	
	lung	45	>0.1	>0.1	stationary	-32 %	
	muscle	88	**0.01**	**0.02**	**Irregular trend**	21 %	
Fe	liver	98	**0.03**	>0.1	**trend**	-11 %	
	kidney	93	>0.1	>0.1	stationary	-16 %	
	lung	45	>0.1	0.06	stationary	-7 %	
	muscle	89	>0.1	0.01	variable	18 %	
Co	liver	51	0.08	>0.1	stationary	NA	no variation
	kidney	47	**0.02**	>0.1	**trend**	NA	positive
	lung	16	>0.1	>0.1	stationary	NA	no variation
	muscle	56	**0.03**	0.07	**trend**	NA	negative
Ni	liver	61	>0.1	**0.05**	variable	-81 %	negative
	kidney	60	>0.1	>0.1	stationary	-61 %	negative
	lung	24	>0.1	>0.1	stationary	-64 %	negative
	muscle	61	>0.1	>0.1	stationary	-93 %	negative

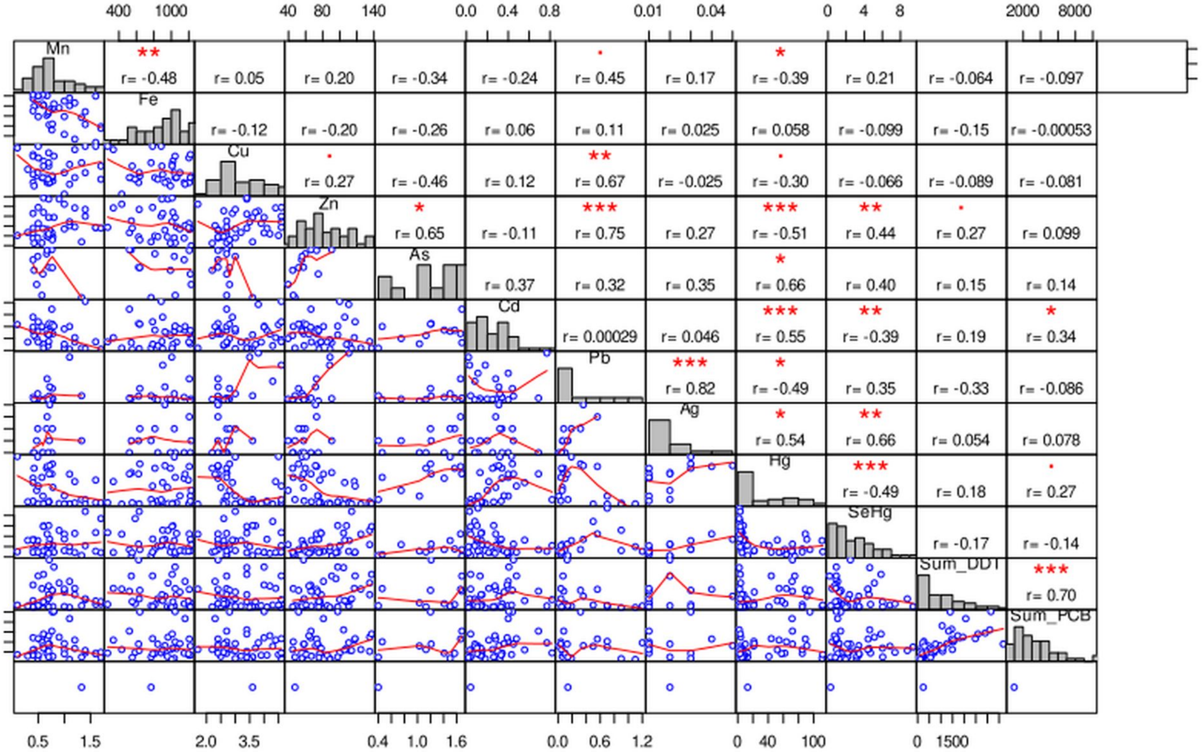
Cu	liver	105	0.08	>0.1	stationary	-16 %	
	kidney	101	>0.1	>0.1	stationary	-26 %	
	lung	51	>0.1	>0.1	stationary	-31 %	
	muscle	92	**0.03**	**0.01**	**Irregular trend**	-4 %	
Zn	liver	106	**0.01**	>0.1	**trend**	-37 %	
	kidney	101	**0.01**	0.07	**trend**	-47 %	2009 drop
	lung	52	**0.04**	>0.1	**trend**	-44 %	2009 drop
	muscle	92	>0.1	0.06	stationary	-29 %	
As	liver	53	>0.1	>0.1	stationary	NA	no variation
	kidney	49	>0.1	>0.1	stationary	NA	no variation
	lung	16	>0.1	>0.1	stationary	NA	no variation
	muscle	58	**0.01**	>0.1	**trend**	NA	negative
Mo	liver	53	**0.04**	>0.1	**trend**	NA	positive
	kidney	49	**0.04**	0.08	**trend**	NA	positive
	lung	16	>0.1	>0.1	stationary	NA	no variation
	muscle	58	>0.1	>0.1	stationary	NA	no variation
Cd	liver	102	>0.1	**0.03**	variable	-13 %	positive since 2007
	kidney	98	**0.01**	**0.03**	**Irregular trend**	-40 %	
	lung	49	>0.1	>0.1	stationary	64 %	
	muscle	86	**0.01**	**0.02**	**Irregular trend**	-27 %	peak in 2010-12
Sn	liver	53	**0.01**	>0.1	**trend**	NA	positive
	kidney	49	>0.1	>0.1	stationary	NA	no variation
	lung	16	>0.1	>0.1	stationary	NA	no variation
	muscle	58	>0.1	>0.1	stationary	NA	no variation
Sb	liver	27	>0.1	>0.1	stationary	NA	no variation
	kidney	25	>0.1	>0.1	stationary	NA	no variation
	lung	6	>0.1	0.06	stationary	NA	no variation
	muscle	20	>0.1	>0.1	stationary	NA	no variation
Pb	liver	62	**0.04**	**0.01**	**Irregular trend**	-88 %	
	kidney	61	**0.02**	**0.04**	**Irregular trend**	-94 %	
	lung	25	**0.02**	**0.03**	**Irregular trend**	-94 %	
	muscle	62	0.09	0.06	stationary	-96 %	
Se	liver	106	0.10	**0.01**	variable	-27 %	peak in 2007-08, positive since
	kidney	101	**0.02**	**0.01**	**Irregular trend**	-57 %	peak in 2007-08

	lung	52	>0.1	>0.1	stationary	232 %	positive since 2007
	muscle	93	>0.1	>0.1	stationary	-22 %	positive since 2007
Ag	liver	53	>0.1	>0.1	stationary	NA	no variation
	kidney	47	>0.1	>0.1	stationary	NA	no variation
	lung	16	>0.1	>0.1	stationary	NA	no variation
	muscle	55	**0.03**	>0.1	**trend**	NA	negative
Hg	liver	106	**0.03**	**0.04**	**Irregular trend**	335 %	
	kidney	100	>0.1	**0.04**	variable	53 %	positive since 2007
	lung	52	>0.1	>0.1	stationary	914 %	positive since 2007
	muscle	93	0.07	>0.1	stationary	209 %	positive since 2007
Se/ (Hg+0.5Ag) molar ratio	liver	106	**0.01**	>0.1	**trend**	-86 %	negative (2009 drop)
	kidney	100	**0.01**	>0.1	**trend**	-70 %	negative (2009 drop)
	lung	52	>0.1	>0.1	stationary	-54 %	negative
	muscle	93	**0.01**	>0.1	**trend**	-63 %	negative

Supplementary Information S5. Correlation plots including metals and organics for which thresholds have been defined in Table 2, in liver tissues (1), kidney (2), lung (3), and muscles (4). In order to avoid an excessive incidence of outliers, values exceeding 3*IQR (interquartile range) were discarded. “r” is the correlation coefficient, and *p*-values are given as *** $p < 0.001$; ** $p < 0.01$; * $p < 0.05$; · $p < 0.1$.







MUSCLE correlation plot