Marine Pollution Bulletin
June 2023, Volume 191 Pages 114901 (9p.)
https://doi.org/10.1016/j.marpolbul.2023.114901
https://archimer.ifremer.fr/doc/00833/94503/

The French Mussel Watch: More than two decades of chemical contamination survey in Mediterranean coastal waters

Briand Marine [1], Herlory Olivier [1, *], Briant Nicolas [2], Brach-Papa Christophe [1], Boissery Pierre [3], Bouchoucha Marc [1]

[1] Ifremer, Laboratoire Environnement Ressources Provence Azur Corse, CS 20330, F-83507 La Seyne Sur Mer, France
[2] Ifremer, Unité Contamination Chimique des Ecosystèmes Marins, F-44311 Nantes, France
[3] Agence de l'Eau Rhône Méditerranée Corse – Délégation Paca Corse, F-13001 Marseille, France

* Corresponding author : Olivier Herlory, email address : olivier.herlory@ifremer.fr

Abstract :

Active biomonitoring of chemical contamination (e.g., Cd, Hg, Pb, DDT, PCB, PAH) in French Mediterranean coastal waters has been performed for more than two decades. This study aimed at presenting the current contamination in 2021 and the temporal evolution of concentrations from 2000. Based on a relative spatial comparison, low concentrations were measured in 2021 at most sites (>83 %). Also, several stations with moderate to high levels were highlighted in the vicinity of major urban industrial centers (e.g., Marseille, Toulon) and near river mouths (e.g., Rhône, Var). Over the last 20 years, no major trend was revealed, mostly, especially for the relative high-level sites. This likely constant contamination over time, plus slight increases of metallic elements at a few sites, still raise questions on the efforts that remain to be made. The decreasing trends of organic compounds, in particular PAH, provide evidence of the efficiency of some management actions.

Highlights

► Contamination in mussels from French waters has been monitored over two decades. ► In 2021, most sites showed relative low concentrations. ► Most contaminants revealed constant levels over the last 20 years at most stations. ► Sites near urban industrial centers or river mouths remain contamination hotspots. ► PAH revealed clear decreasing trends for 63 % of stations.

Keywords : Biomonitoring, Caging, Mytilus galloprovincialis, Trace metals, Organic compounds, Temporal trend

Traditionally, marine contamination monitoring has been based on the chemical analysis of a list of contaminants in different environmental matrices. No single matrix provides a holistic view of the chemical contamination of an environment (White 1984; Beyer et al. 2017). Living organisms are relevant means of detecting contaminants since they are able to detect early effects resulting from exposure (Van der Oost et al. 2003). The bioaccumulation phenomenon results from the processes of absorption, excretion and accumulation in a pseudo-equilibrium with levels in the environment (Borchardt 1983; Cossa 1989). It seems to be applicable to all marine organisms and the majority of bivalent metals and lipophilic organic compounds (Harvey et al. 1973), although some are better bioindicators than others depending on the contaminant considered (Furness and Camphuysen 1997; e Silva et al. 2006; Zhou et al. 2008).

Mussels, such as *Mytilus* spp., have been extensively used as sentinel species of contamination in marine coastal systems (*e.g.,* Stephenson et al. 1995; Lauenstein and Daskalakis 1998; Scarpato et al 2010; Briant et al. 2017). As sessile filter feeding, their capability to accumulate contaminants in their tissues at higher concentrations than the surrounding environment (Phillips 1977; Phillips and Rainbow 2013) conducted to the development of local, national and international monitoring programs (Farrington et al. 2016), *e.g.,* in the United States the Mussel Watch Program (NMW, Goldberg 1975; Hunt and Slone 2010; Melwani et al. 2014), in South Africa (Sparks et al. 2014), in Spain (Santos-Echeandia et al. 2021), in France (RNO/ROCCH, Claisse 1989).

Historically, the coast of the Mediterranean Sea has been under very considerable pressure in the form of extensive land-use, decreasing freshwater resources, increasing amounts of sewage, litter and dangerous waste, habitat destruction and the contamination of marine resources (Gabrielides 1995; Danovaro 2003). Moreover, it is particularly threatened due to its half-closed marine configuration, leading to slow water turnover (around one century). The exponential population increases in the last 30 years have led to critical urbanization of the coastline and the intensification of human activities in all the adjacent countries (tourism, agriculture, fisheries and aquaculture, industries and maritime traffic, Jenkins 2013, Holon et al. 2015). Consequently, monitoring protocols have been implemented in different Mediterranean regions in the framework of the Program for the Assessment and Control of Pollution in the Mediterranean region (MEDPOL, https://www.unep.org/unepmap/, and the Land-based sources Protocol of the UNMAP-Barcelona Convention), established in 1975. Since 1979, the French national mussel watch program (RNO/ROCCh) has been carried out each year on native bivalves taken from the French coast (Claisse 1989). However, to face the issue of scarce natural resource stocks in French Mediterranean, a dedicated program called the RINBIO network based on a mussel-transplantation technique was established in 1996 and has been conducted every 3 years since 2000 (Andral et al. 2004, 2011). Thanks to these networks, a large dataset of contaminant concentrations along the French Mediterranean coast is now available (SURVAL:

https://surval.ifremer.fr/Donnees/Cartographie-Donnees-par-parametre#/map). This study is aimed at presenting the main results of the RINBIO network which are: 1) a statement of the current contamination of coastal waters in 2021, and 2) a temporal analysis of the contamination evolution over the last 20 years.

The RINBIO network relies on active biomonitoring (*e.g.,* Taleb et al. 2009; Moschino et al. 2016; Parolini et al. 2020), *i.e.,* caging individuals of the mussel *Mytilus galloprovincialis* (De Kock 1983; Fabris et al. 1994) from a non-contaminated coastal area in the Occitania region (Andral et al. 2004). This transplantation method solves the problem of scarce natural mussel stocks in much of the Mediterranean coastal zone (Andral et al. 2004) and makes it possible to control the source, age, and stage of sexual maturity of the samples. Each batch was made up of adult mussels 18–24 months old, measuring about 50 mm, sorted twice according to the height of the shell through a 19-mm mesh. The 3kg samples were stored in man-made conchyliculture pouches mounted on PVC tubing.

In 2021, mussels were immerged at 66 sites (figure 1 and table 1), of which 57 had been repeatedly sampled every 3 years since 2000. Stations were homogeneously and strategically distributed, all along the 1800 km of the French Mediterranean shoreline, from the Spanish to the Italian borders and all around the island of Corsica. Monitoring sites were localized between few hundred meters to 10 kilometers max from the coast, in the vicinity of river mouths (Aude, Hérault, Rhône, Huveaune, Gapeau, Var, Golu, Tavignano, Fium Urbo), sewage treatment plant discharges (Montpellier, Cortiou, Sicié, Bastia, Marana, Gravone and Ajaccio), surrounding important urbanized and/or industrialized harbors (*e.g.,* Marseille, Toulon, Villefranche, Bonifacio) and at reference locations remote from direct sources of contamination (*e.g.,* Banyuls, Frontignan, Rogliano, Cargèse). For each campaign and at each station, the mussels were immerged between 6 and 8m for 3 months, from March to July (figure 2, details of the method can be found in Andral et al. 2004).

After the recovery of mussel pouches by scuba divers, the mortality rate was determined for each sampling point. Mussels were separated, rinsed in seawater and pre-processed immediately on board according to standardized procedures in line with proposals of OSPAR Commission (2013). Biometric parameters (length, width and shell weight) were recorded on a pool of fifteen specimens. Chemical contaminants (metals and organic compounds) were analyzed on randomly chosen batches of eleven to twenty specimens according to the quantity of biological material retrieved. For each batch, the flesh was scraped out of the opened raw shell with a stainless steel scalpel, stored in acid washed glass vials. Every pooled sample of flesh was weighed and freeze-dried. For biometry, mussel shells were also dried at 60°C in an oven for 48h, then weighed. The ratio of dry flesh weight to dry shell weight (FW/ SW) was used to determine a condition index (hereafter CI) for each sample. This protocol was consistent over the 20 years of monitoring.

For each sample, 9 metallic trace elements (As, Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn, details in SM1) were quantified by inductively coupled plasma mass spectrometry (ICP-MS, iCAP TQ Thermo, quantification limit: 0.001 to 0.83 mg.kg^{-1} dry weight, SM1) after pre-treatment (grinding with a mechanical homogenizer, Kinematica Polytron PT2500E, and lyophilization) and mineralization by an HNO_3 solution at 110°C (Trace Metal Grade, VWR). Quantification of total Hg was determined by atomic fluorescence (AMA-254 Altec) (quantification limit: 0.015 mg/kg dw, SM1). CE 278K, NIST 2976 and NIST-SRM2976 certified mussel reference materials were used to control analytical reliability. 25 organic compounds (DDT and 2 metabolites, 6 PCB, 16 PAH, SM1) were also quantified by isotopic dilution capillary gas chromatography coupled with mass spectrometry (GC/MSMS, LC/MSMS or GC/HRMS) after extraction by hexane/acetone solution and cleaned up with a silica gel /sulfuric acid cartridge (quantification limit: 0.01 to 2 µg.kg^{-1} dw). The results are expressed in mg or µg of contaminants per kilogram of dry mussel flesh and values below the quantification limit were substituted with a half-quantification limit value prior to data treatment.

A relative spatial comparison of concentrations measured at the 66 sites in 2021 was assessed. As the RINBIO network includes a large number of reference stations, the data is dominated by low concentrations, with few stations with high concentrations distributed along the series. Although these stations may be the most important for the monitoring of the chemical contamination, they greatly influence classical statistics. Therefore, to assess contamination class boundaries, we removed for each contaminant 10% of the highest values and then calculated the mean and standard deviation. Then the classes of contamination were estimated according to the mean standard deviation metric (σ) as baseline ($<$ mean + 1*σ), low ($<$ mean + 2*σ), moderate ($<$ mean + 3*σ), high ($<$ mean + 4*σ) and very high ($>$ mean + 4*σ). A temporal trend analysis from 2000 to 2021 was performed at each station using a linear regression analysis (concentrations *versus* years), with a significance threshold set at 0.05. In order to obtain a sufficient statistical weight, only sites sampled for a minimum number of 5 campaigns were taken into account, that is to say 57 stations. Both analyses were performed on raw data which fitted a normal distribution of residuals and equal variances, and applied on each element for metals (except Mn for which the data set was too low) or sums for organic compounds, *i.e.*, ∑3DDT, ∑16PAH and ∑6PCB. All statistical analyses were performed with R Statistical Software (version 4.1.1, R Core Team 2022).

The caging methodology allows cost minimization plus a highly satisfactory retrieval rate, taking into account the shape and diversity of the coasts studied (Andral et al. 2004). Indeed, mussels immersed in 2021 were recovered at 97% of stations, and pouches were lost at only two sites (not shown in the study). The mean mortality rate of the pouches collected was 11.4 ± 3.9 % (from 3.8 to

22.7%), confirming the operational viability and efficiency of the RINBIO approach for monitoring actions along the whole the Mediterranean coast.

This work provides recent values on contamination levels in the north-occidental Mediterranean Sea. In 2021, for the contaminants which have thresholds in application of the European Water Framework Directive (EU WFD 2000/60/EC, SM1), all the concentrations remained below their regulatory levels. Metallic and organic contaminants measured in mussels mainly showed baseline to low concentration levels along the French Mediterranean coastline, *i.e.*, 88 to 100% of sites. However, our study also highlighted several relative hotspots with moderate to very high concentrations. Mainly located in the river mouths and in the vicinity of the major urban and industrial centers, hotspots are linked to the intensity of human activity in the associated watersheds (Andral et al. 2004, 2011). Significant levels of metals and metalloids (As, Cr and/or Mn) likely stemming from natural and anthropic industrial/agricultural activities were detected in coastal sites receiving river inputs, defined as moderate to very high (table 1); stations close to the Aude (*st. 06*, Mn: 9.7 mg.kg^{-1} dw), the Hérault (*st. 08*, Cr: 2.69 mg.kg^{-1} dw), the Rhône (*st. 15*, Mn: 10.4 mg.kg^{-1} dw), the Var (*st. 37* and *39,* As: 113 and 93, Cr: 2.47 and 3.17, Mn: 11.56 mg.kg^{-1} dw), the Tavignano and the Fium Orbu (*st. 47* and *48*, Cr: 3.48 and 2.73, Ni: 4.07 mg.kg^{-1} dw). Two sites near touristic harbors and marinas of Corsica showed considerable Cu concentrations, estimated as high in the surroundings of Bonifacio (*st. 53,* 6.0 mg.kg^{-1} dw) and very high in the surroundings of Porto-Vecchio (*st. 50,* 9.0 mg.kg^{-1} dw, table 1). This contamination may be linked to the use of antifouling paints, as suggested in other harbors, marinas and enclosed bays around the world (Stephenson and Leonard 1994; O'Connor and Lauenstein 2005; Melwani et al. 2014; Briant et al. 2022). Indeed, since the banning of TBT in 1982, Cu became the main active biocide substance employed (Briant et al. 2013). In addition, monitoring performed in the coastal waters surrounding the decommissioned Canari asbestos mine (North East Corsica) confirmed historic Cr and Ni contamination (*st. 63* to *65*, very high mean values of 2.91 and 3.90 mg.kg^{-1} dw respectively, table 1, *e.g.,* Schreier, 1989; Galgani et al. 2006; Lafabrie et al. 2008). Although mining activities ceased 56 years ago, the excess inputs from 15 years of metal mining wastes have led to high concentrations in the environment to date (Marengo et al. 2023). The area from Marseille to the Rhone River was marked by clear PCB contamination (*st. 15* to *21*, table 1). These PCB fluxes might be fueled by several sources along the coast, like the Huveaune river (Kanzari et al. 2014) which has two outlets (*st. 20*, ∑PCB: 26.7 µg.kg^{-1} dw and *st. 21,* 26.4 µg.kg^{-1} dw) and the Rhone river (*st. 15,* 19.6 µg.kg^{-1} dw). These organic contaminants are probably transported west throughout the Ligurian current (Lipiatou and Saliot 1991; Tolosa et al. 1997). Environmental regulations of sources of contaminants enacted in France since 1975 for open systems (*e.g.,* pesticides, coatings) and 1986 for closed systems (*e.g.,* transformers, capacitors, Chevreuil et al. 1988) have clearly reduced the PCB concentrations recorded in sediments along the rivers (Desmet et al. 2012; Mourier et al. 2014; Liber et al. 2019). Also, very high ∑PAH contamination was detected in the bay of Villefranche (*st. 40*, 24.9 µg.kg^{-1} dw, table 1). It is potentially linked to the maritime activities in this bay well known for accommodating yachts and cruise vessels (Baumard et al. 1998), although the nature of the compounds must be investigated to conclude on the source of contamination (petrogenic *vs.* pyrolytic origin). This last point highlights the importance of limiting contaminant inputs in areas at risk with low water turnovers like enclosed bay and harbor sites (Melwani et al. 2014). Moreover, significant levels of DDT by-products (~ 75% of DDE) measured west of the Rhone river (*st. 02* to *15*), in particular at two seaside towns, Port Vendres (*st. 02,* 6.76 µg.kg^{-1} dw) and Valras (*st. 06*, and 6.38 µg.kg^{-1} dw, table 1), are consistent with the high contamination of Occitanian lagoons described in Andral et al. 2004, which may imply common sources of contamination. The persistency of DDT degradation products 50 years after the banning of DDT in France provides evidence of the massive use of this insecticide for sanitary and agricultural purposes in the region in the 1960s. Finally, the results obtained in the bay of Toulon, *i.e.*, *st. 27*, attested to considerable historic contamination by metallic (Pb: 3.15 and Hg: 0.272 mg.kg^{-1} dw) and organic contaminants (PAH: 53.1 and PCB: 76.3 µg.kg^{-1}, table 1) (Andral et al 2004). Indeed, military, industrial and harbor activities have led to considerable inputs in the bay of Toulon over several decades (Andral et al. 2004; Tessier et al. 2011; Tessier 2012; Wafo et al. 2017; Dang et al. 2021; Araújo et al. 2019). The detection of low concentrations in surrounding stations *25*, *26* and *28*, confirmed the presence of compounds nearly strictly restricted to the small bay of Toulon, except for HAP compounds. This localized contamination might be favored by the semi-enclosed configuration of the site.

This study provides data on trends of chemical contamination over time on the scale of the whole French Mediterranean coastline. Except for ∑HAP, the large majority of stations, including many hotspots of contamination detected in 2021, did not show any temporal variation (67 to 96 %, table 2). Contamination levels remained steady for As and Cr in river mouth stations (*st. 06* Valras, *st. 15* Rhône, *st. 38* Antibes N, and *st. 47* Tavignano), Cu in harbor sites (*st. 50* Porto-Vecchio), as well as Cr and Ni in the vicinity of the former asbestos mine at Canari (*st. 65*). The same patterns were found at several sites associated with organic pollution, such as ∑DDT in Occitanian seaside towns (*st. 02* to *08*), and PCB in the area from Marseille Cortiou to the Rhone River (*st. 15* to *21*). Lastly, relatively high concentrations of Hg, Pb, ∑PAH and ∑PCB showed that the historic contamination in the bay of Toulon has lasted for two decades (*st. 27*, table 2). At these locations, management measures taken since 20 years ago to mitigate marine contamination did not seem to significantly reduce the chronic levels of historical contaminants, likely because of their persistent nature (like heavy metals, DDT and PCB) and their potential remobilization from sediment (like trace elements, Campillo et al. 2019; Layglon et al. 2022). Nonetheless, they succeeded in maintaining these concentrations under recommended values characteristic of a good chemical state (EU WFD 2000). These results confirm the need to continue monitoring such historically contaminated sites. For some stations organic and inorganic compounds showed some significant trends during the last two decades. For metals and metalloids, this concerns from 4 to 33% of the sites, depending on the contaminant considered (table 2). Almost all showed increasing concentrations (table 2), except for Cu which presented decreasing concentration patterns at two sites (*st. 02* and *12*, table 2). Most of these increasing metallic concentrations stayed within a range of low values from 2000 to 2021, and only in a few other cases did concentrations increase to significant levels over time. For example, for sites under the influence of the Var and the Fium Orbu rivers (*st. 37, 48* and *49*, table 2), the increase of Cr, As, Ni and/or Pb concentrations by about a factor of 2 to 4 led to moderate to very high contaminations in 2021. Significant increases by a factor ranging from 3 to 5 were seen for Ni and Cr at Ile Rousse (*st. 63*, table 2). These increases in metallic concentrations do not follow most of the temporal trends obtained by biomonitoring programs in some other parts of the world, which present decreasing evolutions (Sturludottir et al. 2013; Melwani et al. 2014; Sparks et al. 2014; Campillo et al. 2019; Santos-Echeandía et al. 2021). However, significant increasing trends have already been described for As and Ni in several sites at Boston and Massachusetts bays (O'Connor and Lauenstein 2006). In this study, the increases may result from different extrinsic factors like an intensification of natural and anthropogenic sources, fluctuations on the hydrological regime of the rivers that could promote a mobilization of contaminants from sediments (Campillo et al. 2019) or else change in speciation and consequently bioavailability of elements. However, levels of contamination in living organisms are also conditioned by intrinsic factors related to the biology of the organisms (Mubiana et al. 2006; Casas et al. 2008; Benedicto et al. 2011). Here, the condition index decreased for almost half of the stations (table 2), which reflects a reduction of trophic resources over the past two decades (*e.g.,* Feuilloley et al. 2022), should lead to an increase in concentration of contaminants in the organisms, especially non-essential metals and metalloids (*e.g.,* Cd, Pb, Hg, Andral et al. 2004). But this reduction of CI could only explain half of the cases (20 stations) for which at least one metal rising trend was emphasized (*i.e.,* 39 sites over 57) and without any particular pattern in contaminant signatures (table 2). Organic compounds revealed exclusively significant decreasing trends at many sites for ∑PCB and ∑PAH (1/3 and 2/3 of sites respectively) but few for ∑DDT (9% of sites, table 2). Many of these decreases have led to improvements over time; for example, all the stations linked to a significant decrease of ∑PAH by factors ~4.5 to 10 (st. *16, 17, 19, 37, 47, 50, 63, 65*) showed high levels in the past whereas they are now at low levels. Even the PAH-contaminated harbor site of Villefranche (*st. 40*) revealed an improvement over two decades. Likewise, ∑PCB levels clearly improved in several sites especially those with harbor activities (*e.g., st. 50* and *56*) or situated near rivers (*e.g., st. 35, 37, 45, 48*) and WTP outlets (*st. 25*). Lastly, the ∑DDT concentrations measured in Palavas (*st. 12*) were divided by a factor of ~3.5 over two decades (11 to 3 $\mu g.kg^{-1}$ dw). DDT is highly environmentally stable with a reported half-life of between 2 and 15 years (Blaylock 2005), so both isomers of DDD and DDE degradation were already the most predominant since 2000, with an increase of DDE (36 to 68%) and a decrease of DDD (50 to 16%) concentrations over time. This result might confirm old contamination in the area of Montpellier, due to the past intensive use of this pesticide (Andral et al. 2011). Therefore, although these compounds are still detected in all marine compartments several decades later due to their high remanence, these results are quite consistent with the effects of regulation and even abolition of many of them in the past (Alonso-Hernández et al. 2015; El Nemr and El-Sadaawy 2016; Buah-Kwofie and Humphries 2017; Dang et al. 2021) and confirm a global decreasing trend for organic contaminants largely reported in the literature (Tolosa et al. 1997; O'Connor and Lauenstein 2006; Kimbrough et al. 2008; Hunt and Slone 2010; Sturludottir et al. 2013; Melwani et al. 2014; Campillo et al. 2019).

In summary, thanks to a long time series of data and to a large spatial spread, *i.e.*, about 60 sampled stations over 20 years, the RINBIO network in Mediterranean Sea is one of the most sustained monitoring programs in the world (*e.g.,* 36 years for the program in the United-States_NOAA NS&T, Farrington et al. 2016). Concentrations detected in 2021 confirmed many of the specific hotspots of metallic and organic contamination and highlighted new ones. It also allowed to update levels for the French Mediterranean coast and Corsica, and allowed comparison to other Mediterranean areas (Santos-Echeandía et al., 2021 and included references); the detected levels of Cd, Cu, Mn and Zn are in the range of common values, whereas Hg and As are respectively relatively lower and higher. Temporal results revealed that contaminants at most sites do not show any trend since 2000, and especially at most of the hotspots showing an historical and constant pollution over two decades. Behind this global observation, there are different situations: no new inputs, a system in equilibrium with old remaining contamination of the sediment or the influence the geological background. That last point, plus the slight increase of few metallic elements over time, signal the scope of the efforts remaining to be made. However, decreasing trends of organic compounds, in particular ∑PAH, also provided crucial evidence of the effectiveness of the management actions taken to reduce certain priority contaminants.

ACKNOWLEDGEMENTS

The authors wish to thank all partners involved in the campaigns since 1996, and especially the SUCHIMED campaign in 2021 (Bouchoucha 2021, SUCHI Med 202 cruise, RV L'Europe, https://doi.org/10.17600/18001619). We are also very grateful to the Rhône-Mediterranean-Corsica Water Agency for its long-term financial and scientific partnership, as well as to all the staff of the Ifremer laboratories (in Toulon, Nantes, Sète and Corsica) and Genavir for their valuable assistance with sample preparation and logistics. We thank the two anonymous reviewers for their insight and thoughts to improve this manuscript.

REFERENCES

Alonso-Hernández, C.M., Tolosa, I., Mesa-Albernas, M., Díaz-Asencio, M., Corcho-Alvarado, J.A., Sánchez-Cabeza, J.A. (2015). Historical trends of organochlorine pesticides in a sediment core from the Gulf of Batabanó, Cuba. Chemosphere, 137, 95-100.

Amouroux, I., Brun, M., (2018). Substances prioritaires DCE : Cohérence et applicabilité des seuils mollusques existants en milieu marin : DCE (NQE, VGE) et OSPAR (EAC, BAC). Ifremer, RBE/BE/ARC : 2018.01-v2, programme AFB, mai 2018, 62 p. https://archimer.ifremer.fr/doc/00441/55242/

Andral, B., Galgani, F., Tomasino, C., Bouchoucha, M., Blottiere, C., Scarpato, A., Benedicto, J., Deudero, S., Calvo, M., Cento, A., Benbrahim, S., Boulahdid, M., Sammari, C. (2011). Chemical contamination baseline in the Western basin of the Mediterranean sea based on transplanted mussels. Archives of Environmental Contamination and Toxicology, 61(2), 261-271.

Andral, B., Stanisiere, J.Y., Sauzade, D., Damier, E., Thebault, H., Galgani, F., Boissery, P. (2004). Monitoring chemical contamination levels in the Mediterranean based on the use of mussel caging. Marine Pollution Bulletin, 49(9-10), 704-712.

Araújo, D.F., Ponzevera, E., Briant, N., Knoery, J., Bruzac, S., Sireau, T., Brach-Papa, C., (2019). Copper, zinc and lead isotope signatures of sediments from a Mediterranean coastal bay impacted by naval activities and urban sources. Applied Geochemistry, 111, 104440.

Baumard, P., Budzinski, H., Michon, Q., Garrigues, P., Burgeot, T., Bellocq, J. (1998). Origin and bioavailability of PAHs in the Mediterranean Sea from mussel and sediment records. Estuarine, Coastal and Shelf Science, 47(1), 77-90.

Benedicto, J., Andral, B., Martínez-Gómez, C., Guitart, C., Deudero, S., Cento, A., Scarpato, A., Caixach, J., Benbrahim, S., Chouba, L., Boulahdid, M., Galgani, F. (2011). A large scale survey of trace metal levels in coastal waters of the Western Mediterranean basin using caged mussels (Mytilus galloprovincialis). Journal of Environmental Monitoring, 13(5), 1495-1505.

Beyer, J., Green, N.W., Brooks, S., Allan, I.J., Ruus, A., Gomes, T., Bråte, I.L.N., Schøyen, M., (2017). Blue mussels (Mytilus edulis spp.) as sentinel organisms in coastal pollution monitoring: a review. Marine Environmental Research 130, 338–365. https:// doi.org/10.1016/j.marenvres.2017.07.024.

Blaylock, B.L., (2005). DDT (Dichlorodiphenyltrichloroethane). In: Anderson, B., Peyster, A.D., Gad, S.C., Hakkinen, P.J., Kamrin, M. Encyclopedia of toxicology (Second Edition). Academic Press. https://doi.org/10.1016/B0-12-369400-0/00291-X

Borchardt, T., (1983). Influence of food quantity on the kinetic of cadmium uptake by Mytilus edulis. Marine Biology 85, 233–244.

Briant, N., Bancon-Montigny, C., Elbaz-Poulichet, F., Freydier, R., Delpoux, S., & Cossa, D. (2013). Trace elements in the sediments of a large Mediterranean marina (Port Camargue, France): levels and contamination history. Marine Pollution Bulletin, 73(1), 78-85.

Briant, N., Chouvelon, T., Martinez, L., Brach-Papa, C., Chiffoleau, J.-F., Savoye, N., Sonke, J., Knoery, J., (2017). Spatial and temporal distribution of mercury and methylmercury in bivalves from the French coastline. Marine Pollution Bulletin, 114(2), 1096-1102. Publisher's official version: https://doi.org/10.1016/j.marpolbul.2016.10.018, Open Access version: https://archimer.ifremer.fr/doc/00353/46465/

Briant, N., Freydier, R., Ferreira Araujo, D., Delpoux, S., Elbaz-Poulichet, F., (2022). Cu isotope records of Cu-based antifouling paints in sediment core profiles from the largest European Marina, The Port Camargue. Science Of The Total Environment, 849, 157885 (10p.). https://doi.org/10.1016/j.scitotenv.2022.157885

Buah-Kwofie, A., Humphries, M.S., (2017). The distribution of organochlorine pesticides in sediments from iSimangaliso Wetland Park: ecological risks and implications for conservation in a biodiversity hotspot. Environmental pollution, 229, 715-723.

Campillo, J.A., Santos-Echeandía, J., Fernández, B., (2019). The hydrological regime of a large Mediterranean river influences the availability of pollutants to mussels at the adjacent marine coastal area: Implications for temporal and spatial trends. Chemosphere 237,124492.

Casas, S., Gonzalez, J. L., Andral, B., Cossa, D., (2008). Relation between metal concentration in water and metal content of marine mussels (Mytilus galloprovincialis): impact of physiology. Environmental Toxicology and Chemistry: An International Journal, 27(7), 1543-1552.

Chevreuil, M., Chesterikoff, A., Létolle, R., (1988). Transport state of PCBs in the river Seine (France). REV SCI EAU, 1(4), 321-337.

Claisse, D., (1989). "Chemical contamination of French coasts. The results of a ten years mussel watch." Marine Pollution Bulletin 20.10: 523-528.

Cossa, D., (1989). A review of the use of Mytilus spp. as quantitative indicators of cadmium and mercury contamination in coastal water. Oceanologica Acta 12, 417–432.

Dang, D.H., Filella, M., Omanović, D., (2021). Technology-Critical Elements: An Emerging and Vital Resource that Requires more In-depth Investigation. Archives of Environmental Contamination and Toxicology, 81(4), 517–520. https://doi.org/10.1007/s00244-021-00892-6

Danovaro, R., (2003). Pollution threats in the Mediterranean Sea: an overview. Chemistry and Ecology, 19(1), 15-32.

De Kock, W.C., (1983). Accumulation of cadmium and PCBs by *Mytilus edulis* transplanted from pristine water into pollution gradients. Can. J. Fish. Aquat. Sci. 40(S2), 282–294.

Desmet, M., Mourier, B., Mahler, B. J., Van Metre, P. C., Roux, G., Persat, H., Lefèvre, I., Peretti, A., Chapron, E., Simonneau, A., Miège, C., Babut, M., (2012). Spatial and temporal trends in PCBs in sediment along the lower Rhône River, France. Science of the Total Environment, 433, 189-197.

E Silva, C.A.R., Smith, B.D., Rainbow, P.S., (2006). Comparative biomonitors of coastal trace metal contamination in tropical South America (N. Brazil). Marine Environmental Research, 61(4), 439-455.

El Nemr, A., El-Sadaawy, M.M., (2016). Polychlorinated biphenyl and organochlorine pesticide residues in surface sediments from the Mediterranean Sea (Egypt). International Journal of Sediment Research, 31(1), 44-52.

European Commission (2006). Commission Regulation (EC) No 1881/2006 of 19 December 2006 setting maximum levels for certain contaminants in foodstuffs. Official Journal of the European Union, 364, 5-24.

European Commission (2013). Directive 2013/39/EU of the European Parliament and of the Council of 12 August 2013 amending Directives 2000/60/EC and 2008/105/EC as regards priority substances in the field of water policy. Official J Eur Union, 226, 1-17.

EU Water Framework Directive (2000/60/EC) of the European Parliament and of the Council of 23 October 2000 establishing a framework for Community action in the field of water policy (OJ L 327, 22.12.2000, pp. 1–73).

Exec. Order No. TREL1819388A (July 27, 2018). Arrêté du 27 juillet 2018 modifiant l'arrêté du 25 janvier 2010 relatif aux méthodes et critères d'évaluation de l'état écologique, de l'état chimique et du potentiel écologique des eaux de surface pris en application des articles R. 212-10, R. 212-11 et R. 212-18 du code de l'environnement. https://www.legifrance.gouv.fr/jorf/id/JORFTEXT000037347756

Fabris, J.G., Richardson, B.J., O'Sullivan, J.E., Brown, F.C., (1994). Estimation of cadmium, lead, and mercury concentration in estuarine waters using the Mussel Mytilus edulis planulatus L. Environmental Toxicology and Water Quality 9, 183–192.

Farrington, J.W., Tripp, B.W., Tanabe, S., Subramanian, A., Sericano, J.L., Wade, T.L., Knap, A.H., (2016). Edward D. Goldberg's proposal of “the mussel watch”: reflections after 40 years. Marine Pollution Bulletin, 110(1), 501-510.

Feuilloley, G., Fromentin, J.M., Saraux, C., Irisson, J.O., Jalabert, L., Stemmann, L., (2022). Temporal fluctuations in zooplankton size, abundance, and taxonomic composition since 1995 in the North Western Mediterranean Sea. ICES Journal of Marine Science, 79(3), 882–900, https://doi.org/10.1093/icesjms/fsab190

Furness, R.W., Camphuysen, K.C.J., (1997). Seabirds as monitors of the marine environment, ICES Journal of Marine Science, 54(4), 726-737, https://doi.org/10.1006/jmsc.1997.0243

Gabrielides, G.P., (1995). Pollution of the Mediterranean sea. Water Science and Technology, 32(9-10), 1-10.

Galgani, F., Chiffoleau, J.F., Orsoni, V., Costantini, L., Boissery, P., Calendini, S., Andral, B., (2006). Chemical contamination and sediment toxicity along the coast of Corsica. Chemistry and Ecology, 22(4), 299-312.

Goldberg, E.D., (1975). The Musssel Watch: a first step in global marine monitoring. Marine Pollution Bulletin 6:111.

Harvey, G.R., Steinhauer, W.G., Teal, J.M., (1973). Chlorinated hydrocarbons in open ocean Atlantic organisms. In: Green, D., Jagner, D. (Eds.), Changing Chemistry of the Oceans. J. Wiley and Sons, NY, pp. 177–186.

Holon, F., Mouquet, N., Boissery, P., Bouchoucha, M., Delaruelle, G., Tribot, A.S., Deter, J., (2015). Fine-scale cartography of human impacts along French Mediterranean coasts: A relevant map for the management of marine ecosystems. PLoS One 10:1–20.

Hunt, C.D., Slone, E., (2010). Long-term monitoring using resident and caged mussels in Boston Harbor yield similar spatial and temporal trends in chemical contamination. Marine Environmental Research, 70(5), 343-357.

Jenkins, S.H., (Ed.), (2013). Mediterranean Coastal Pollution: Proceedings of a Conference Held in Palma, Mallorca, 24-27 September, 1979. Elsevier.

Kanzari, F., Syakti, A. D., Asia, L., Malleret, L., Piram, A., Mille, G., & Doumenq, P. (2014). Distributions and sources of persistent organic pollutants (aliphatic hydrocarbons, PAHs, PCBs and pesticides) in surface sediments of an industrialized urban river (Huveaune), France. Science of the Total Environment, 478, 141-151.

Kimbrough, K.L., Johnson, W.E., Lauenstein, G.G., Christensen, J.D., Apeti, D.A., (2008). An Assessment of Two Decades of Contaminant Monitoring in the Nation’s Coastal Zone. Silver Spring, MD, p. 105.

Lafabrie, C., Pergent-Martini, C., Pergent, G., (2008). First results on the study of metal contamination along the Corsican coastline using Posidonia oceanica. Marine pollution bulletin, 57(1-5), 155-159.

Layglon, N., Lenoble, V., Longo, L., D'Onofrio, S., Mounier, S., Mullot, J.U., Sartori, D., Omanovic, D., Garnier, C., Misson, B., (2022). Cd transfers during marine sediment resuspension over short and long-term period: Associated risk for coastal water quality. Marine Pollution Bulletin, 180, 113771.

Lauenstein, G.G., Daskalakis, K.D., (1998). US long-term coastal contaminant temporal trends determined from mollusk monitoring programs, 1965–1993. Marine Pollution Bulletin, 37(1-2), 6-13.

Liber, Y., Mourier, B., Marchand, P., Bichon, E., Perrodin, Y., Bedell, J.P., (2019). Past and recent state of sediment contamination by persistent organic pollutants (POPs) in the Rhône River: Overview of ecotoxicological implications. Science of the Total Environment, 646, 1037-1046.

Lipiatou, E., Saliot, A., (1991). Fluxes and transport of anthropogenic and natural polycyclic aromatic hydrocarbons in the western Mediterranean Sea. Marine chemistry, 32(1), 51-71.

Marengo, M., Fullgrabe, L., Fontaine, Q., Boissery, P., Cancemi, M., Lejeune, P., Gobert, S. (2023). Ecological and human health risk assessment of potentially toxic element contamination in waters of a former asbestos mine (Canari, Mediterranean Sea): implications for management. Environmental Monitoring and Assessment, 195(1), 1-24.

Melwani, A.R., Gregorio, D., Jin, Y., Stephenson, M., Ichikawa, G., Siegel, E., Crane, D., Lauenstein, G., Davis, J. A., (2014). Mussel watch update: long-term trends in selected contaminants from coastal California, 1977–2010. Marine pollution bulletin, 81(2), 291-302.

Moschino, V., Del Negro, P., De Vittor, C., Da Ros, L. (2016). Biomonitoring of a polluted coastal area (Bay of Muggia, Northern Adriatic Sea): a five-year study using transplanted mussels. Ecotoxicology and Environmental Safety, 128, 1-10.

Mourier, B., Desmet, M., Van Metre, P. C., Mahler, B. J., Perrodin, Y., Roux, G., Bedell, J.-P., Lefèvre, I., Babut, M., (2014). Historical records, sources, and spatial trends of PCBs along the Rhône River (France). Science of the total environment, 476, 568-576.

Mubiana, V. K., Vercauteren, K., & Blust, R., (2006). The influence of body size, condition index and tidal exposure on the variability in metal bioaccumulation in Mytilus edulis. Environmental Pollution, 144(1), 272-279.

O'Connor, T.P., Lauenstein, G.G., (2005). Status and trends of copper concentrations in mussels and oysters in the USA. Mar. Chem. 97, 49–59.

O'Connor, T.P., Lauenstein, G.G., (2006). Trends in chemical concentrations in mussels and oysters collected along the U.S. coast: Update to 2003. Marine Environmental Research 62, 261e285.

OSPAR Commission, (2013). Background document and technical annexes for biological effects monitoring, Monitoring and Assessment Series. Update.

Parolini, M., Panseri, S., Gaeta, F. H., Ceriani, F., De Felice, B., Nobile, M., Rafoss, T., Schnelle, J., Herradae, I., Arioli, F., Chiesa, L. M. (2020). Incidence of persistent contaminants through blue mussels biomonitoring from Flekkefjord fjord and their relevance to food safety. Food Additives & Contaminants: Part A, 37(5), 831-844.

Phillips, D.J.H., (1977). The use of biological indicator organisms to monitor trace metal pollution in marine and estuarine environments—a review. Environmental Pollution (1970) 13.4: 281-317.

Phillips, D.J.H., Rainbow, P.S., (2013). Biomonitoring of trace aquatic contaminants. Vol. 37. Springer Science & Business Media.

R Core Team (2022). R: A language and environment for statistical computing. R Foundation for Statistical Computing, Vienna, Austria.URL https://www.R-project.org/

Santos-Echeandía, J., Campillo, J.A., Egea, J.A., Guitart, C., González, C.J., Martínez-Gómez, C., León, V.M., Rodríguez-Puente, C., Benedicto, J., Benedicto, J., (2021). The influence of natural vs anthropogenic factors on trace metal (loid) levels in the Mussel Watch programme: Two decades of monitoring in the Spanish Mediterranean Sea. Marine Environmental Research, 169, 105382.

Scarpato, A., Romanelli, G., Galgani, F., Andral, B., Amici, M., Giordano, P., Caixach, J., Calvo, M., Campillo, J.A., Albadalejo, J.B., Alessandro Cento, A., BenBrahim, S., Sammari, C., Deudero, S., Boulahdid, M., Giovanardi, F. (2010). Western Mediterranean coastal waters—Monitoring PCBs and pesticides accumulation in Mytilus galloprovincialis by active mussel watching: the Mytilos project. Journal of Environmental Monitoring, 12(4), 924-935.

Schreier, H., (1989). Asbestos in the natural environment. Elsevier.

Sire, A., Amouroux, I., (2016). Determination of thresholds in marine molluscs as an alternative to the Environmental Quality Standards in marine water defined in the Water Framework Directive. SETAC Europe 2016 - 26th annual Meeting. 22-26 May, 2016, Nantes, France. https://archimer.ifremer.fr/doc/00344/45565/

Sparks, C., Odendaal, J., Snyman, R., (2014). An analysis of historical Mussel Watch Programme data from the west coast of the Cape Peninsula, Cape Town. Marine pollution bulletin, 87(1-2), 374-380.

Stephenson, M.D., Leonard, G.H., (1994). Evidence for the decline of silver and lead and the increase of copper from 1977 to 1990 in the coastal marine waters of California. Marine Pollution Bulletin 28, 148–153.

Stephenson, M.D., Martin, M., Tjeerdema, R.S., (1995). Long-term trends in DDT, polychlorinated biphenyls, and chlordane in California mussels. Archives of Environmental Contamination and Toxicology, 28(4), 443-450.

Sturludottir, E., Gunnlaugsdottir, H., Jorundsdottir, H.O., Magnusdottir, E.V., Olafsdottir, K., Stefansson, G., (2013). Spatial and temporal trends of contaminants in mussel sampled around the Icelandic coastline. Science of the total environment, 454, 500-509.

Taleb, Z. M., Benali, I., Gherras, H., Ykhlef-Allal, A., Bachir-Bouiadjra, B., Amiard, J. C., Boutiba, Z. (2009). Biomonitoring of environmental pollution on the Algerian west coast using caged mussels Mytilus galloprovincialis. Oceanologia, 51(1), 63-84

Tessier, E. (2012). Diagnostic de la contamination sédimentaire par les métaux/métalloïdes dans la rade de Toulon et mécanismes contrôlant leur mobilité (Doctoral dissertation, Université de Toulon).

Tessier, E., Garnier, C., Mullot, J.-U., Lenoble, V., Arnaud, M., Raynaud, M., Mounier, S., (2011). Study of the spatial and historical distribution of sediment inorganic contamination in the Toulon bay (France). Marine Pollution Bulletin 62: 2075-2086.

Tolosa, I., Readman, J.W., Fowler, S.W., Villeneuve, J.P., Dachs, J., Bayona, J.M., Albaiges, J., (1997). PCBs in the western Mediterranean. Temporal trends and mass balance assessment. Deep Sea Research Part II: Topical Studies in Oceanography, 44(3-4), 907-928.

Van der Oost, R., Beyer, J., Vermeulen, N.P., (2003). Fish bioaccumulation and biomarkers in environmental risk assessment: a review. Environmental toxicology and pharmacology, 13(2), 57-149.

Wafo, E., Abou, L., Nicolay, A., Boissery, P., Garnier, C., Portugal, H., (2017). Historical Trends of Polycyclic Aromatic Hydrocarbons (PAHs) in the Sediments of Toulon Bay (South of France). International Journal of Environmental Monitoring and Analysis, 5(6), 150-158.

White, H.H., (1984). Mussel madness: Use and misuse of biological monitors of marine pollution. Technical report. Maryland University Sea Grant Program. College Park MD[TECH. REP. MD. UNIV. SEA GRANT PROGRAM.]. 325-338.

Zhou, Q., Zhang, J., Fu, J., Shi, J., Jiang, G., (2008). Biomonitoring: an appealing tool for assessment of metal pollution in the aquatic ecosystem. Analytica chimica acta, 606(2), 135-150.

Table 1. *Concentrations of metallic (As, Cd, Cr, Cu, Hg, Mn, Ni, Pb and Zn, in mg.kg^{-1} dw) and organic (sums of 3 DDT, 6 PCB and 16 PAH, in µg.kg^{-1} dw) contaminants measured in mussels during the 2021 campaign at 66 coastal sites (M: mouth and WTP: Water Treatment Plant). The estimated classes of contamination are indicated by color coding in the legend below, i.e. baseline (< mean + 1*σ), low (< mean + 2*σ), moderate (< mean + 3*σ), high (< mean + 4*σ) and very high (> mean + 4*σ).*

Contamination levels: *baseline* *low* *moderate* *high* *very high*

		As	Cd	Cr	Cu	Hg	Mn	Ni	Pb	Zn	∑DDT	∑PCB	∑PAH
	Stations	*mg.kg^{-1} dw*									*µg.kg^{-1} dw*		
01	*Banyuls*	41	1,07	1,02	3,53	0,075	4,27	1,48	1,70	158	4,35	6,59	7,43
02	*Port Vendres*	47	0,86	0,65	3,37	0,061	4,22	1,01	1,67	127	6,65	9,84	6,66
03	*Argelès*	49	0,96	0,88	3,11	0,062	4,17	1,30	1,80	145	5,86	9,95	6,26
04	*Canet plage*	41	0,90	0,90	3,51	0,063	4,90	1,34	1,56	155	4,88	10,45	6,23
05	*Port la Nouvelle*	43	0,74	1,12	3,23	0,054	4,72	1,40	1,41	118	4,67	7,17	8,37
06	*Valras*	62	1,01	1,66	5,06	0,049	9,71	1,72	2,00	169	6,38	9,74	6,94
07	*Aude M*	38	0,76	1,11	3,78	0,053	7,83	1,35	1,73	123	5,62	8,96	6,87
08	*Hérault M*	39	0,67	2,69	3,52	0,048	7,16	2,29	1,33	131	5,12	6,84	6,38
09	*Agde*	46	0,68	0,96	3,51	0,055	6,41	1,18	1,46	120	3,89	6,91	7,74
10	*Frontignan*	38	0,67	0,97	3,63	0,061	5,66	1,19	0,97	110	3,38	9,51	7,65
11	*Montpellier WTP*	42	0,93	1,10	3,64	0,057	5,13	1,41	1,22	142	3,57	6,85	5,60
12	*Palavas*	39	1,00	0,73	3,58	0,060	4,63	1,30	0,87	200	3,47	10,14	5,97
13	*Grau du Roi*	32	0,75	0,61	3,57	0,050	5,71	0,98	0,85	136	4,91	10,20	5,70
14	*Stes Maries*	39	0,84	0,82	4,24	0,058	6,99	1,24	1,17	155	4,70	10,70	7,00
15	*Rhône M*	37	0,93	1,81	3,98	0,067	10,41	1,91	1,40	167	5,18	19,60	11,38
16	*Ponteau*	43	0,99	0,74	4,23	0,108	5,23	1,32	1,52	176	2,30	14,71	9,56
17	*Carry*	40	1,12	1,23	4,33	0,090	4,06	1,39	1,50	134	2,26	18,74	10,19
18	*Marseille breakwater*	43	1,10	0,73	4,78	0,100	3,94	0,99	1,61	189	3,15	28,01	10,39
19	*Pomègues*	38	1,05	0,94	4,43	0,084	4,13	1,07	1,32	186	2,57	18,58	10,93
20	*Huveaune M*	34	0,90	0,61	3,72	0,101	3,74	0,78	1,40	154	4,84	26,70	12,44
21	*Cortiou WTP*	50	1,43	0,78	4,73	0,093	4,07	1,30	2,60	271	2,21	26,42	12,42
22	*Ile plane*	39	1,26	0,44	4,04	0,087	3,36	0,96	1,58	162	1,47	12,25	9,74
23	*La Ciotat*	52	1,40	0,77	4,15	0,110	4,02	1,39	1,82	225	1,37	6,33	10,43
24	*Ile embiez*	76	1,85	1,01	4,53	0,119	4,46	1,82	2,11	241	1,09	4,29	8,59
25	*Sicié WTP*	57	1,40	1,54	4,62	0,094	6,27	2,00	2,02	245	0,67	3,81	8,61
26	*Toulon BH*	57	1,50	0,82	4,56	0,140	3,78	1,42	2,08	248	1,05	11,84	18,51
27	*Toulon SH*	36	0,65	0,65	5,59	0,272	4,28	0,71	3,15	149	3,10	76,30	53,08
28	*Carqueiranne*	57	1,54	1,01	4,75	0,111	4,62	1,54	1,70	207	0,73	2,36	10,18
29	*Gapeau M*	44	1,37	0,68	4,25	0,119	4,04	1,14	1,45	198	2,40	5,20	10,10
30	*Bregançon*	67	1,50	0,80	4,17	0,110	4,18	1,19	1,70	206	1,35	3,47	8,04
31	*Cavalaire*	64	1,71	1,10	4,21	0,106	3,53	1,85	1,66	208	0,71	1,97	10,23
32	*Pampelone*	67	1,64	0,73	4,65	0,121	4,11	1,27	1,84	225	0,55	1,70	11,81
33	*St Tropez*	62	1,30	1,03	4,82	0,092	5,04	1,33	1,72	201	0,99	3,47	11,33
34	*Argens M*	66	1,46	1,13	4,69	0,090	5,92	1,52	2,04	232	1,32	2,95	9,08

35	*Fréjus*	71	1,45	0,79	4,35	0,114	4,31	1,25	1,80	202	1,12	2,71	13,81
36	*Cannes*	79	1,45	1,00	5,13	0,091	4,21	1,38	1,71	226	1,53	4,28	13,42
37	*Antibes S*	113	1,45	2,47	4,04	0,101	5,70	2,31	2,07	282	1,05	2,66	10,99
38	*Antibes N*	75	1,41	2,06	4,41	0,089	6,90	2,07	1,76	230	1,27	3,66	12,58
39	*Var M*	93	1,52	3,17	4,67	0,085	11,56	3,08	2,21	232	1,57	5,06	13,04
40	*Villefranche*	80	1,61	0,93	4,49	0,139	4,46	1,44	2,52	312	1,28	7,03	24,85
41	*Menton*	77	1,81	1,10	4,41	0,139	5,37	1,86	2,41	277	1,69	3,87	14,87
42	*Rogliano*	36	2,00	1,06	4,46	0,140	3,05	2,56	1,77	289	0,30	2,70	7,90
43	*Bastia WTP*	49	2,08	1,67	4,24	0,138	4,73	3,26	2,12	351	0,29	0,79	14,98
44	*Marana WTP*	45	1,78	1,32	4,53	0,131	4,20	2,34	1,94	286	0,23	0,80	8,74
45	*Golu M*	52	1,77	1,87	4,40	0,140	5,72	2,61	1,98	279	0,28	0,83	8,17
46	*Poggio Mezzana*	58	1,87	1,29	4,75	0,150	4,71	2,34	1,68	237	0,24	0,86	10,16
47	*Tavignano M*	55	1,93	3,48	4,56	0,137	5,80	4,07	2,05	332	0,31	0,67	6,46
48	*Fium Orbu M*	47	1,92	2,73	4,19	0,127	5,21	3,30	1,60	216	0,39	0,70	7,01
49	*Cavu*	57	1,83	1,04	4,27	0,125	4,73	1,94	1,50	218	0,31	0,65	8,55
50	*Porto Vecchio*	46	1,64	1,77	9,05	0,118	5,86	2,26	1,95	257	0,42	1,25	10,23
51	*Sant Amanza*	49	1,81	1,08	4,33	0,117	4,23	1,87	1,46	232	0,37	1,46	7,17
52	*Ile Lavezzi*	47	1,63	0,81	4,23	0,110	3,65	1,38	1,24	228	0,27	1,23	8,03
53	*Bonifacio*	53	1,46	1,16	6,05	0,179	3,85	1,28	2,45	355	4,18	9,63	21,97
54	*Figari Bruzzi*	48	1,97	1,07	4,04	0,132	3,64	2,37	1,64	235	0,14	0,57	8,14
55	*Sartène*	45	1,89	1,01	3,60	0,124	3,20	1,86	1,68	252	0,22	0,92	8,40
56	*Propriano*	46	1,79	0,91	4,19	0,111	4,37	1,87	1,74	289	0,27	0,82	7,76
57	*Ajaccio WTP*	50	1,87	1,56	5,08	0,119	4,25	2,28	2,05	318	0,30	1,40	9,50
58	*Gravone WTP*	52	1,69	0,86	4,12	0,105	3,96	1,58	1,82	219	0,30	1,57	8,34
59	*Cargèse*	51	1,90	1,01	4,15	0,111	3,58	2,02	1,59	269	0,13	0,64	7,32
60	*Porto*	41	1,80	1,69	5,55	0,118	3,34	2,62	1,45	252	0,20	0,99	6,78
61	*Galeria*	58	2,15	0,91	5,81	0,152	3,69	2,32	2,13	335	0,23	0,67	7,39
62	*Revellata*	54	1,89	0,96	4,62	0,124	3,11	1,98	1,66	233	0,23	1,01	7,38
63	*Ile Rousse*	59	1,91	2,74	4,50	0,135	3,71	3,99	1,66	264	0,67	3,74	11,70
64	*St Florent*	51	1,99	1,26	3,54	0,112	3,19	3,84	1,67	332	0,72	0,87	9,13
65	*Canari*	47	1,96	3,08	3,96	0,132	3,67	3,87	1,66	272	0,30	0,79	6,74
66	*Pino*	59	1,84	2,10	3,97	0,134	3,52	3,23	1,94	315	0,14	0,76	7,72
	mean	48	1,37	1,07	4,20	0,097	4,40	1,64	1,66	208	1,49	4,47	8,9
	sd	9	0,41	0,37	0,48	0,027	0,84	0,52	0,31	53	1,46	3,88	2,1

Table 2. *Summary of temporal trend analysis performed on contaminant concentrations and Condition Index values (CI) at 57 stations sampled over 20 years (2000-2021). Results, i.e. no trend (ns), significant "increasing" (↗, red) or "decreasing" (↘, green) trends, are presented for all stations (values in %) and detailed for each station. Also indicated are the number of Rinbio campaigns analyzed (RC) and p-values as p<0.05*, p<0.01 **, p<0.001 ***.*

			As	Cd	Cr	Cu	Hg	Ni	Pb	Zn	∑DDT	∑PCB	∑PAH	CI	
		Increase	*12*	*33*	*16*	*0*	*16*	*23*	*30*	*9*	*0*	*0*	*0*	*0*	
		Decrease	*0*	*0*	*0*	*4*	*0*	*0*	*0*	*0*	*9*	*33*	*63*	*44*	
		No trends	*88*	*67*	*84*	*96*	*84*	*77*	*70*	*91*	*91*	*67*	*37*	*56*	***RC***
Stations	**01**	*Banyuls*	ns	ns	ns	ns	ns	ns	↗ *	ns	ns	↘ *	↘ ***	↘ **	*8*
	02	*Port Vendres*	ns	ns	ns	↘ *	ns	ns	ns	ns	ns	ns	ns	ns	*7*
	03	*Argelès*	↗ *	ns	ns	ns	ns	↗ *	ns	ns	ns	ns	ns	↘ **	*8*
	04	*Canet plage*	↗ *	ns	ns	ns	↗ *	↗ *	ns	ns	ns	↘ *	↘ *	↘ *	*6*
	05	*Port la Nouvelle*	↗ *	ns	ns	ns	ns	ns	ns	ns	ns	ns	↘ **	↘ *	*8*
	06	*Valras*	↗ *	ns	ns	ns	ns	ns	ns	ns	ns	ns	↘ *	ns	*7*
	09	*Agde*	↗ *	ns	ns	ns	ns	↗ *	↗ *	ns	ns	ns	↘ **	ns	*7*
	10	*Frontignan*	ns	ns	↗ *	ns	↗ **	ns	ns	ns	↘ *	ns	↘ *	↘ *	*8*
	11	*Montpellier WTP*	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	*5*
	12	*Palavas*	ns	ns	↗ *	↘ *	↗ *	ns	ns	ns	↘ *	↘ *	↘ **	↘ *	*7*
	13	*Grau du Roi*	ns	ns	ns	ns	ns	ns	ns	ns	↘ *	ns	↘ *	ns	*7*
	14	*Stes Maries*	ns	↗ *	ns	ns	↗ *	ns	↗ **	ns	ns	ns	↘ *	↘ *	*8*
	15	*Rhône M*	ns	ns	ns	ns	ns	↗ *	ns	ns	ns	ns	ns	ns	*6*
	16	*Ponteau*	ns	↗ *	ns	ns	ns	ns	ns	↗ *	ns	ns	↘ **	↘ **	*8*
	17	*Carry*	ns	↗ *	ns	ns	↗ *	ns	ns	↗ *	ns	ns	↘ *	↘ *	*8*
	18	*Marseille breakwater*	ns	↗ **	ns	ns	ns	↗ *	ns	↗ **	ns	ns	ns	ns	*5*
	19	*Pomègues*	ns	↗ **	ns	ns	↗ *	↗ *	ns	↗ **	ns	ns	↘ *	↘ **	*7*
	22	*Ile plane*	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	*6*
	23	*La Ciotat*	ns	↗ **	ns	ns	ns	ns	↗ **	ns	ns	ns	↘ *	↘ *	*8*
	24	*Ile embiez*	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	↘ **	ns	*7*
	25	*Sicié WTP*	ns	ns	↗ **	ns	ns	↗ **	ns	ns	ns	↘ *	↘ ***	↘ *	*8*
	26	*Toulon BH*	ns	↗ *	ns	ns	ns	ns	↗ **	ns	↘ *	ns	ns	↘ *	*8*
	27	*Toulon SH*	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	*8*
	28	*Carqueiranne*	ns	↗ **	ns	ns	ns	ns	↗ **	ns	ns	ns	↘ *	ns	*7*
	29	*Gapeau M*	ns	↗ ***	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	*7*
	30	*Bregançon*	ns	↗ **	ns	ns	ns	ns	↗ **	ns	ns	ns	↘ ***	ns	*8*
	31	*Cavalaire*	ns	↗ **	ns	ns	ns	↗ *	↗ **	ns	ns	ns	↘ **	ns	*8*
	32	*Pampelone*	ns	↗ *	ns	ns	ns	ns	↗ **	ns	ns	ns	↘ *	ns	*8*
	33	*St Tropez*	ns	↗ *	ns	ns	ns	↗ *	ns	ns	ns	ns	↘ ***	ns	*8*
	34	*Argens M*	ns	ns	ns	ns	ns	ns	ns	ns	ns	↘ *	↘ *	↘ **	*8*
	35	*Fréjus*	ns	↗ *	ns	ns	ns	ns	↗ *	ns	ns	↘ *	↘ **	↘ *	*8*
	36	*Cannes*	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	*8*
	37	*Antibes S*	↗ *	↗ *	↗ **	ns	ns	↗ *	↗ *	ns	ns	↘ *	↘ *	↘ *	*8*
	38	*Antibes N*	ns	↗ *	ns	ns	ns	↗ *	↗ *	↗ *	ns	ns	↘ **	ns	*6*
	40	*Villefranche*	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	↘ *	ns	*7*
	41	*Menton*	ns	ns	ns	ns	ns	ns	ns	ns	ns	↘ *	↘ *	ns	*8*
	42	*Rogliano*	ns	ns	ns	ns	↗ *	ns	ns	ns	ns	ns	↘ **	↘ *	*8*
	44	*Marana WTP*	ns	ns	ns	ns	↗ *	ns	ns	ns	ns	ns	ns	ns	*7*
	45	*Golu M*	ns	ns	ns	ns	ns	ns	ns	ns	ns	↘ *	ns	↘ *	*7*
	46	*Poggio Mezzana*	ns	↗ *	ns	ns	ns	ns	ns	ns	ns	↘ *	↘ *	ns	*6*
	47	*Tavignano M*	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	↘ *	↘ *	*8*

	48	*Fium Orbu M*	ns	ns	↗ **	ns	ns	↗ *	↗ *	ns	ns	↘ *	ns	↘ **	*8*
	49	*Cavu*	↗ *	ns	↗ *	ns	ns	ns	ns	ns	ns	↘ *	↘ *	ns	*7*
	50	*Porto Vecchio*	ns	ns	↗ *	ns	ns	ns	ns	ns	ns	↘ ***	↘ **	ns	*8*
	51	*Sant Amanza*	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	↘ *	*5*
	52	*Ile Lavezzi*	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	*5*
	54	*Figari Bruzzi*	ns	ns	ns	ns	↗ *	ns	↗ *	ns	ns	↘ ***	ns	↘ *	*8*
	55	*Sartène*	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	↘ *	ns	*6*
	56	*Propriano*	ns	ns	ns	ns	ns	ns	ns	ns	ns	↘ ***	ns	ns	*8*
	59	*Cargèse*	ns	ns	ns	ns	ns	ns	↗ *	ns	ns	↘ **	ns	↘ *	*8*
	60	*Porto*	ns	↗ *	↗ *	ns	ns	ns	ns	ns	ns	↘ ***	ns	↘ *	*8*
	61	*Galeria*	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	↘ *	*6*
	62	*Revellata*	ns	ns	ns	ns	ns	ns	↗ *	ns	ns	ns	ns	ns	*6*
	63	*Ile Rousse*	ns	ns	↗ **	ns	ns	↗ *	ns	ns	ns	ns	↘ *	ns	*8*
	64	*St Florent*	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	↘ *	ns	*6*
	65	*Canari*	ns	↗ *	ns	ns	ns	ns	ns	ns	↘ *	↘ *	↘ **	ns	*5*
	66	*Pino*	ns	ns	ns	ns	ns	ns	↗ **	ns	ns	↘ *	↘ **	ns	*8*

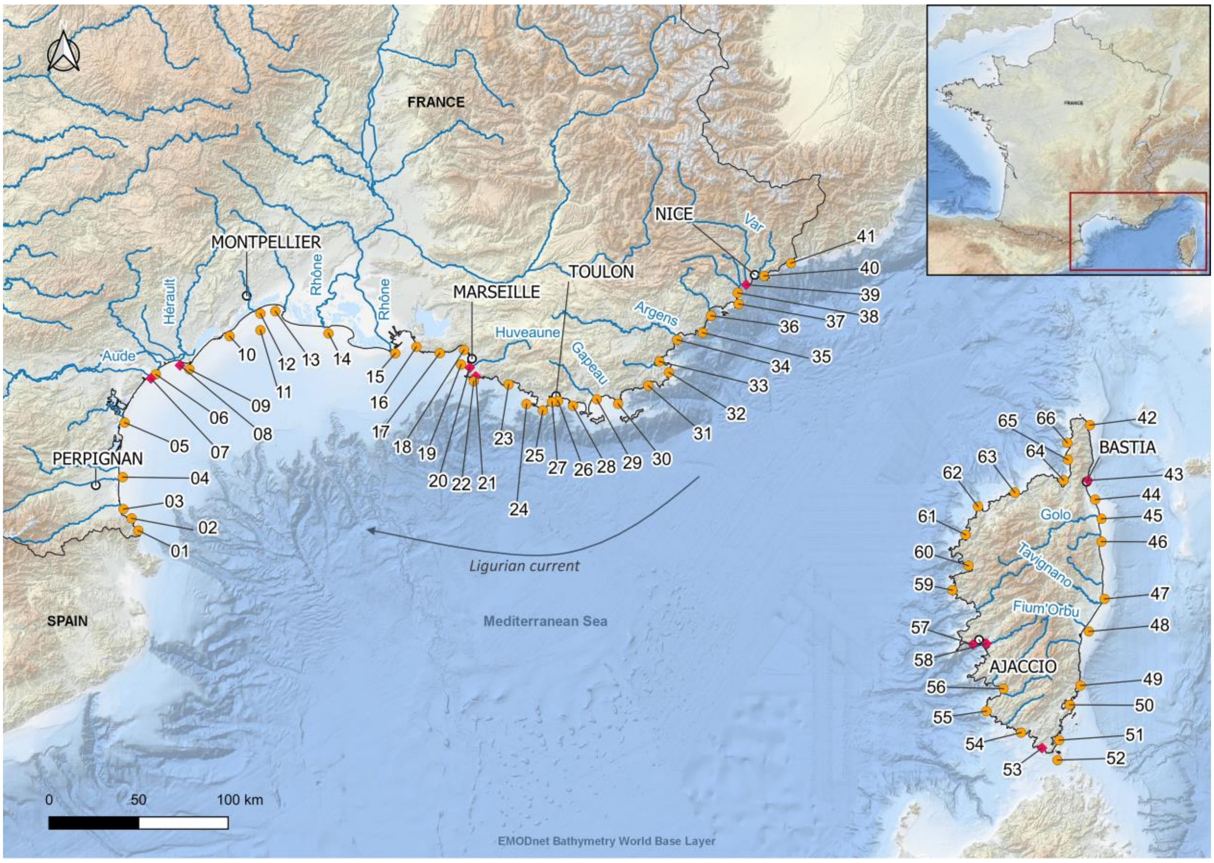

Figure 1. *Location of sampling sites along the French Mediterranean coast: stations included (yellow circles) and excluded (red diamonds) from the temporal analysis, plus main cities, rivers and currents are indicated on the map.*

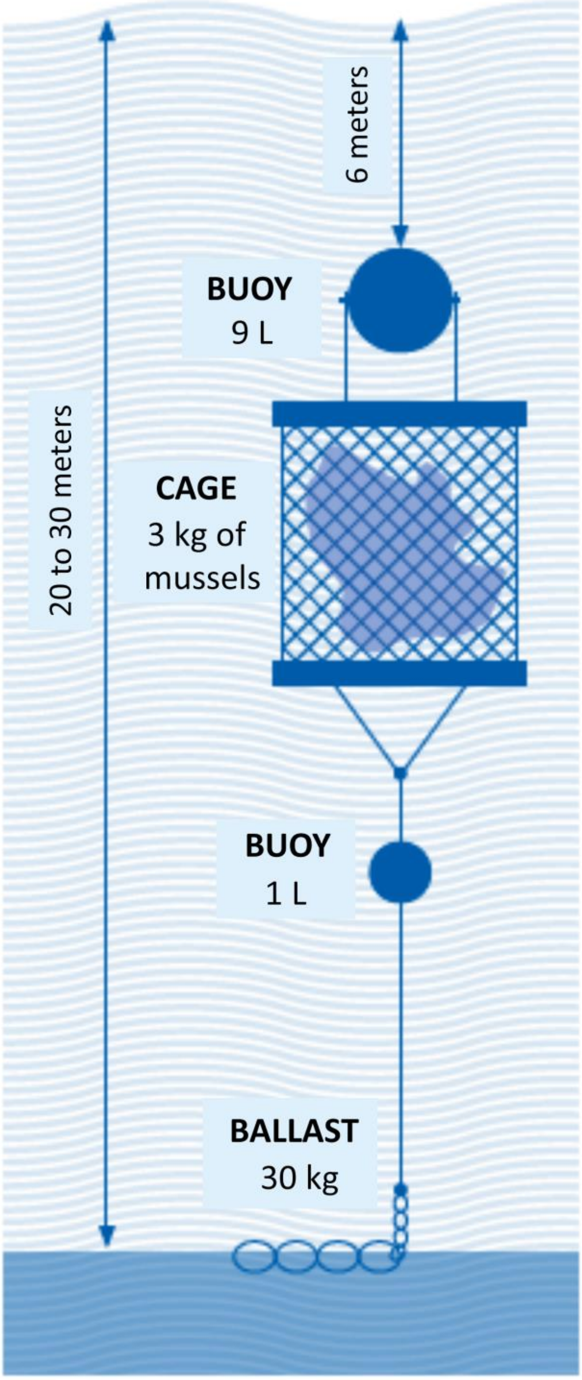

Figure 2. *Description of the RINBIO anchoring system. The 3-kg samples of mussels were stored in conchylicultural pouches mounted on PVC tubing. An 11-l buoy attached to the upper part of the pouch maintained it at a depth of 6 m from the surface, regardless of any condition of biofouling or muddiness. The system was anchored to the bottom by means of a ballast weighing about 30 kg, connected to the pouch by a 7-mm polypropylene line. Moorings were doubled or tripled in zones exposed to trawling.*

Supplementary Material 1. *List of metallic and organic contaminants measured in mussels during campaigns from 2000 to 2021. EC: European sanitary threshold (European Commission 2006), EQS: Environmental quality standards (European Commission 2013 and Exec. Order No. TREL1819388A, 2018), EGV: Environmental Guiding Values proposed for assessment of good chemical status of French coastal waters only (Sire and Amouroux 2016; Amouroux and Brun 2018).*

Contaminant	Analytical method	Limit of Quantification	Threshold type	Threshold value
METALS & METALLOIDS				
As	ICP-MS	0.11 mg.kg^{-1} dw	/	/
Cd		0.001 mg.kg^{-1} dw	EC	1000 μg.kg^{-1} ww
Cr		0.13 mg.kg^{-1} dw	/	/
Cu		0.83 mg.kg^{-1} dw	/	/
Mn		0.07 mg.kg^{-1} dw	/	/
Ni		0.19 mg.kg^{-1} dw	EGV	8677 μg.kg-1 ww
Pb		0.04 mg.kg^{-1} dw	EC	1500 μg.kg-1 ww
Zn		2.2 mg.kg^{-1} dw	/	/
Hg	AMA	0.015 mg.kg^{-1} dw	EC	500 μg.kg-1 ww
DDT				
pp'DDT	isotopic dilution GC/MSMS	0.005 μg.kg^{-1} ww	EQS (∑4isomers) : 1282 μg.kg^{-1} ww	
pp'DDE				
pp'DDD				
PCB				
PCB 28	isotopic dilution GC/HRMS	0.01 μg.kg^{-1} ww	/	/
PCB 52		0.01 μg.kg^{-1} ww	/	/
PCB 101		0.01 μg.kg^{-1} ww	/	/
PCB 138		0.01 μg.kg^{-1} ww	/	/
PCB 153		0.01 μg.kg^{-1} ww	/	/
PCB 180		0.01 μg.kg^{-1} ww	/	/
PAH				
Naphtalene	isotopic dilution GC/MSMS	0.05 μg.kg^{-1} ww	EQS	214 μg.kg^{-1} ww
Acenaphtylene		0.05 μg.kg^{-1} ww	/	/
Acenaphtene		0.05 μg.kg^{-1} ww	/	/
Fluorene		0.1 μg.kg^{-1} ww	/	/
Phenanthrene		1 μg.kg^{-1} ww	/	/
Anthracene		0.1 μg.kg^{-1} ww	EQS	173 μg.kg^{-1} ww
Fluoranthene		0.05 μg.kg^{-1} ww	EQS	30 μg.kg^{-1} ww
Pyrene		0.05 μg.kg^{-1} ww	/	/
Benzo[a]anthracene		1 μg.kg^{-1} ww	/	/
Chrysene		0.05 μg.kg^{-1} ww	/	/
Benzo[b]fluoranthene		0.1 μg.kg^{-1} ww	EQS	5 μg.kg^{-1} ww
Benzo[k]fluoranthene		0.1 μg.kg^{-1} ww	EQS	5 μg.kg^{-1} ww
Benzo[a]pyrene		0.1 μg.kg^{-1} ww	EQS	5 μg.kg^{-1} ww
Dibenz[a;h]anthracene		0.05 μg.kg^{-1} ww	/	/

Benzo[g;h;i]perylene		0.1 µg.kg^{-1} ww	EQS	5 µg.kg^{-1} ww
Indeno[1,2,3,c-d]pyrene		0.1 µg.kg^{-1} ww	EQS	5 µg.kg^{-1} ww