

**Hazardous substances in the
Mediterranean:
a spatial and temporal assessment**

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PREAMBLE

The Report presents a spatial and temporal assessment on hazardous substances in sediment and biota from the Mediterranean marine and coastal environment, based on the MED POL database and available international literature. The Report has been prepared by Prof. Joan Albaigés, Ms. Carla Murciano and Mr. Jordi Pon (Department of Environmental Chemistry, CID-CSIC, Spain), under the supervision of UNEP/MAP - MED POL.

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Executive summary

Marine pollution monitoring is one of the main components of the Barcelona Convention that is implemented through the Mediterranean Pollution Monitoring and Research Programme (MED POL). The Programme started in 1975 with the main aim of establishing a network of Institutions undertaking marine pollution work and the collection of information regarding the level of pollution in the Mediterranean Sea. The monitoring activities basically covered heavy metals (mainly mercury and cadmium) and halogenated hydrocarbons (mainly PCBs and DDTs) in sediments and marine biota.

The development and maintenance of these national monitoring programmes was the aim of the second phase, adopted in 1981, whereas later (1996), in Phase III, the emphasis shifted from pollution assessment to pollution control. A major outcome of this Phase was the setting up of a Database (MED POL.mdb) and the inter-linked web version ([http://195.97.36.231/MED POL/](http://195.97.36.231/MED_POL/)).

The critical analysis of the results, achievements, deficiencies and experience of the MED POL Phase III, particularly those related directly to the practical aspects of pollution control, were used in designing the operational details of a realistic and well focused Phase IV (2006-2010) (UNEP, 2005). In this respect, monitoring was better integrated into the scope of MAP and used as a tool for: i) assessing the state and trends of the marine and coastal environment and the effects of pollution; and ii) assisting the countries' implementation of the LBS, Dumping and Hazardous Wastes Protocols. The incorporation of Spain, France and Italy to the monitoring network during Phase IV represented a significant step forward in the implementation of MED POL. Data collection and handling, reporting and data management policies and procedures were enforced.

The scope of the present work is to prepare a Thematic Assessment Report on Hazardous Substances in the Mediterranean coastal environment, including sediment and biota contamination, based on the MED POL database and available international literature, and comment on possible pollution trends (not necessarily statistically supported).

The analysis will cover the full period of MED POL Phase III and Phase IV, until 2010, and will incorporate additional scientific information included in the four sub-regional assessment reports, which were prepared by MAP in the framework of the gradual application of the Ecosystem Approach for the management of human activities in the Mediterranean (UNEP/MAP, 2010b).

The MED POL monitoring database is at the moment hosting monitoring data of 14 Mediterranean countries, of particular relevance for the present assessment (Table 1), and although the content is highly variable and the portion of data for each component and country is uneven it constitutes a relevant source of information.

In principle, only representative data was used, basically considering the existence of sufficiently large datasets. In this respect, only trace metals were found suitable for assessment at the regional level in sediments and trace metals and organochlorinated compounds in marine biota. In the case of biota, the Database includes a large number of marine species, but the present report is focused on the bivalve *Mytilus galloprovincialis* and the benthic fish *Mullus barbatus*, as they are the more common and widely analyzed species in the region.

Table 1. Available data of hazardous substances in the MED POL Database

Country	Sediments		Biota	
	Trace Metals	Organics	Trace Metals	Organics
Albania			2001 - 2007	2001 - 2007
Croatia	2002 – 2005, 2009	2009	2000 - 2005, 2009	1999 – 2005, 2009
Cyprus			1999, 2001, 2004, 2005	1999 - 2007
Egypt	2006, 2009, 2010	2006, 2009, 2010	2006, 2009, 2010	2009, 2010
France	2006-2009	2006-2009	1997 - 2009	1999 - 2009
Greece	1999, 2004, 2005, 2007	2004, 2005	1999, 2000, 2004 - 2007	2004, 2005
Israel	1999 - 2009		1999 - 2009	
Italy	2001 - 2005	2001-2005	2001 - 2005	2001 - 2005
Morocco	2007		1999 - 2007	2007
Slovenia		1999-2006, 2010	1999 - 2010	
Spain			2004 - 2007	2004 - 2007
Syria	2007		2007	
Tunisia	2006 - 2009		2001 - 2009	
Turkey	1999, 2003 - 2009		1999 2003 - 2009	2003 - 2009

As far as the specific parameters are concerned, they were selected on the basis of those considered of priority concern and taking into account the total number of samples and the geographical and temporal coverages. Thus, the selection included the trace metals Cd, Hg, Pb, Zn and Cu, and the families of DDTs and PCBs, as the more representative persistent organic pollutants (POPs). The Drin's (aldrin, endrin and dieldrin), HCB and lindane were also considered.

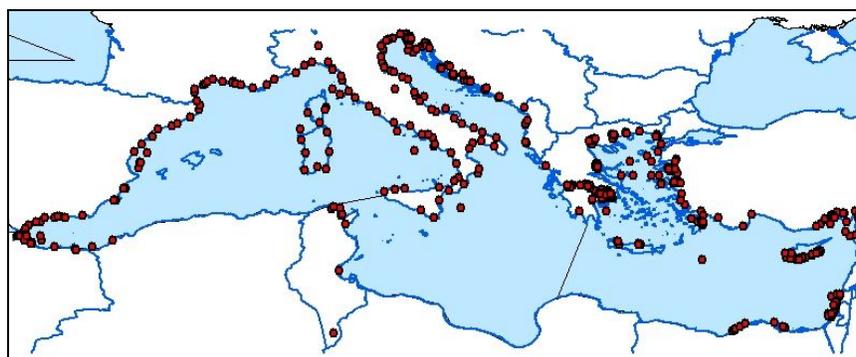
In order to harmonize the database, all concentrations were recorded in $\mu\text{g g}^{-1}$ or ng g^{-1} dry weight. In cases where they were reported in other units or in wet weight basis, they were converted accordingly. In summary, after selection and harmonization of data, a total of **34738 observations**, corresponding to more than 400 stations monitored during the MED POL Phases III and IV (1999-2010) have been included in the present assessment.

The assessment is divided in two sections: i) occurrence and geographical distribution of trace metals (in sediments and biota) and chlorinated pesticides and PCBs (in biota); and ii) temporal trends. Special reference is made to the assessment of the four eco-regions in which the Mediterranean has been divided.

i) Occurrence and geographical distribution of trace metals and chlorinated compounds

A comparison of the levels of trace metals found in sediments and marine biota, and chlorinated pesticides, notably DDTs and lindane, and PCBs, in marine biota of the different Mediterranean eco-regions is summarized in the following sections.

In general, good data coverage is found for the Northern part of the Mediterranean basin while only a small number of sampling stations are defined in the Southern riparian countries, as shown in the Figure below. Therefore, data has been complemented with available information from the literature as well as other MAP reports. In comparing data, special use is made of the median concentration values because they constitute a better representation of the left-skewed populations studied.



Map of MED POL stations.

Trace metals in sediments

An overview of the concentration ranges of Cd, total Hg, Pb, Zn and Cu in coastal sediments of the region is summarized in the following Table. As it is shown, Cd and Hg exhibit values several orders of magnitude lower than Zn, being Pb and Cu in an intermediate and similar position. Although the number of samples is far from being representative of the basin (the southern Central and Aegean-Levantine eco-regions are underrepresented) and the lithology of the continental shelf may influence the occurrence of trace metals in sediments, the values can be used as reference for comparison purposes.

Trace metals in sediments. Median (range) concentrations ($\mu\text{g g}^{-1}$ dw)

Eco-region	Cd	HgT	Pb	Zn	Cu
WESTERN MEDITERRANEAN	1.60 (0.23-7.61)	0.16 (0.02-12.6)	19.40 (0.24-256)	50.10 (1.0-731)	13.90 (0.68-107)
ADRIATIC	0.21 (0.01-18.5)	0.10 (0.01-166.9)	9.83 (0.39-1033)	67.00 (5.0-980)	16.31 (1.39-465)
CENTRAL MEDITERRANEAN	0.09 (0.01-2.80)	0.05 (0.00-6.00)	4.39 (0.01-103.4)	32.66 (0.02-205)	5.35 (0.01-67.5)
AEGEAN- LEVANTINE	0.10 (0.01-8.47)	0.15 (0.00-6.70)	16.89 (0.01-404)	59.34 (0.02-1505)	18.52 (0.01-962)

In general, the Western Mediterranean is the region where concentrations are higher as a result of the intense human activities that can cause chemical contamination of coastal areas, followed by the Adriatic and the Aegean-Levantine basin. However, these values are in the lower range than those reported in previous assessments derived from MEDPOL I and II (UNEP, 1996).

The analysis of levels by countries reveals the occurrence of some stations with high values of Hg, Pb, Cu and Zn in Croatia. A wide range of values for all metals was found in Italy, particularly for Pb. Average values of Cd, Zn and Cu in the higher range were found in Morocco and Syria whereas in Egypt and Israel were in the lower end. Values of Pb and Zn were also high in Greece, and of Zn and Cu in Turkey.

Besides urban and industrial inputs, rivers and streams are major contributors of metals of anthropogenic or natural origin to coastal areas. Moreover, land-based sources of specific trace metals are not only set down at coastal sediments but may end up deep in the marine canyons. Atmospheric deposition may also contribute to the contamination of deep sea sediments.

In the **Western Mediterranean**, major hotspots for metals, particularly Cd, Pb, Hg and Cu, are located in the areas of Marseilles-Fos and Toulon, in France, and Cartagena-Valle Escombreras-Portman, in Spain, holding important urban-industrial and mining areas, respectively. Sensitive areas are represented by the coastal lagoons (e.g. Berre, Thau and Mar Menor). The area influenced by the Rhone River discharges exhibited increased levels of all metals except Hg.

The levels of Pb are important along the Italian coast, especially in areas around the Gulf of Genova (e.g. Savona, La Spezia, Genova, Livorno) that also show evidence of Zn accumulation, while high levels of Pb are found in Naples. These pollution sources can be related to industrial and domestic wastes and harbour activities. Mercury levels are also high in the area of Messina and Palermo and Regio Calabria. The contributions related to tectonic sources are important in volcanic and geothermal sources near as southern Tyrrhenian Sea. These contributions could explain natural important levels of mercury in some islands of the basin.

Significant concentrations of Zn, Cu and particularly Cd have been reported on the coast of Morocco (Tangier-Martil and Nador), whereas higher levels of Hg were found in Algeria (Algiers).

Trace metals have been extensively monitored in the **Adriatic Sea**. The Po River, draining the most industrialized part of northern Italy, is an important pollution vector in the area. High levels of Cd, Hg, Pb and Zn are found in the northern Adriatic, including the Venice and Grado Lagoons. Mercury and Pb are also critical contaminants in the Gulf of Trieste and Rijeka Bay. The occurrence of some discrete stations with high levels of Hg, Pb, Cu and Zn are found in Croatia, such as at Kastela and Martinska Bays. In Albania, Hg contamination from a former inland chlor-alkali plant is evidenced in sediments of Vlora Bay.

The **Central Mediterranean** exhibits elevated levels of Hg in the Gulf of Taranto and in both the Tunisian and Italian coasts of the Strait of Sicily. However, an extensive study in the Ionian Sea revealed that mercury levels were generally comparable to those from other Mediterranean areas. The Tunis and Bizerta Lakes (Tunisia) also also exhibits high contents of Pb. In the Hellenic coastal zone, the most elevated levels of such contaminants in sediments (e.g. Amvrakikos and Patraikos Gulfs) can be associated with the main sewage (domestic and industrial) outfalls. Very high values of Cd have been found close to Patras.

The analysis of representative trace metals in sediments of the **Aegean – Levantine basin** revealed the occurrence of high values of Cd and Zn at Iskenderun Bay (Turkey). Very high levels of Cd, Pb, Zn and Cu have been found in Izmit Bay. Lead was also high in Ismir Bay. Most of the stations, particularly Ismir, Edremit and Candarli Bays, exhibited moderate levels of Cu and Zn. Low to moderate levels were found in Israel, with high values of Hg and Zn in some stations (e.g. Haifa Bay), whereas Cd was high in the northern coast of Syria. Conversely, and with the exception of Hg, relatively low values were found in Egypt, around the mouth of the Nile River. The distribution patterns reflect anthropogenic sources originating from point and diffuse land-based sources, providing useful information on the identification of hotspots in the area although not fully comprehensive.

In Greece, monitoring of metals in sediments revealed a pollution gradient across the coastal areas, indicating different pollution fingerprints, with higher concentrations of Pb, Cu and Zn in the areas of influence of Athens, Thessaloniki and Kavala (Saronikos, Strymonikos and Kavala Gulfs).

Trace metals in bivalves and fish

The mussel *Mytilus galloprovincialis* has been the most widely used sentinel organism in the region. In the case of fish, *Mullus barbatus* (red mullet) has been extensively used for monitoring but only in the Aegean-Levantine region.

The summary of data corresponding to trace metals in mussels, shown in the following Table, provides the range of values found in each sub-region. The accumulation follows a similar trend than for sediments Cd < HgT < Pb < Cu << Zn. Here, the reported values are, excluding the hotspots, of the same order of magnitude than those obtained during the MED POL I and II. Like in the case of sediments, the median values can be used as a reference for a preliminary assessment of spatial trends and identification of hot spots.

Trace metals in *Mytilus galloprovincialis*. Median (range) concentrations ($\mu\text{g g}^{-1}$ dw)

Eco-region	Cd	HgT	Pb	Zn	Cu
WESTERN MEDITERRANEAN	0.66 (0.01-10.0)	0.13 (0.01-7.4)	2.00 (0.01-79.1)	130 (0.01-5337)	6.30 (0.05-114)
ADRIATIC	0.80 (0.03-2.73)	0.14 (0.01-8.45)	1.53 (0.07-67.5)	122 (5.7-467)	8.01 (0.51-81.6)
CENTRAL MEDITERRANEAN	0.43 (0.09-3.40)	0.16 (0.00-7.00)	0.81 (0.07-5.36)	87 (11.6-565)	9.32 (1.36-70.5)
AEGEAN- LEVANTINE	0.75 (0.05-5.27)	0.08 (0.01-0.63)	2.28 (0.84-5.97)	131 (0.00-325)	7.84 (1.01-112)

The concentrations of trace metals in sediments and mussels exhibit a fair correlation except in the case of Zn, which can be attributed to a major influence of the sediment lithology. As it was observed for sediments, concentrations of trace metals in mussels are higher for the Western Mediterranean and the Adriatic Sea, although the data set is lacking information from several eastern and southern countries

In most cases, low or moderate levels were found, in particular for Cd, Hg and Zn, and to a lesser degree, for Pb and Cu. In this respect, Cd, Hg and Zn display a rather even distribution along the coasts of the different eco-regions, with a few hotspots in the

coasts of Spain, in the areas of Cartagena (Cd) and Alboran (Zn), Italy, in the coasts of Sardinia (Cd) and Naples (Cd, Hg and Zn), and Turkey (Ismir Bay) (Cd). Mercury hotspots are also found in Croatia (Rijeka Bay) and Greece (Patras). A large number of stations exhibit high values of Pb, usually associated to urban areas (e.g. Barcelona, Marseille, Genova, Fiumicino and Naples) and mining spots (Portman, Spain). High levels of Cu have been found in the Ligurian (e.g. Genova) and Thyrenian Seas (e.g. Fiumicino and Naples) as well as in the coast of Sardinia.

Besides the MEDPOL monitoring, a large number of studies has been carried out in coastal enclosures and open areas of the Western Mediterranean, Adriatic and Aegean Sea. In the **Western Mediterranean**, some stations like those close to Genova, at the Ligurian Sea, and Naples are chronically polluted by all metals. In addition to these, certain sites can be considered as particular hot spots. High levels of Cd and Pb are found in the Sardinia coast (Portoscuso), Palermo and Cartagena-Portman. Higher levels of Hg and Pb were also found in coastal waters of the Tyrrhenian Sea (e.g. Fiumicino and Messina). All these stations are close to mining, industrial and/or urban areas. High levels of Cu have been found in the Ligurian (e.g. Genova) and Thyrenian Seas (e.g. Fiumicino and Naples) as well as in the coasts of Calabria and Sardinia.

High levels of Pb are usually associated to direct river/urban sewage inputs. This explains the levels found in mussels collected in the Marseille Gulf and Hyeres Bay (France), and the mouth of the Llobregat River (NE Spain), also influenced by the Barcelona metropolis, Malaga, Fiumicino, etc.

Very high Hg concentrations were measured at five scattered sites along the Western Mediterranean basin (Portoscuso, Palermo and Maddalena-Sardinia Island, in Italy; Skikda in Algeria, and El Portus in Spain). There was also a predominance of high Hg values at Algiers Bay, along the north-west Italian coast (Piombino, Portoferraio) and in the French coast at Hyeres ouest.

Finally, the levels of Zn are also high in the Alboran coast of Spain as well as in the southern coast of Italy (Calabria and Sicilia)

Trace metals are extensively monitored throughout the **Adriatic Sea** using mussels as bioindicators, mostly confined in urban and industrial areas. Concentrations are moderate in Northern, NE and NW part of the basin. High concentrations of Pb and Cu were found in the Venice Lagoon and in the areas affected by the Po River discharges, consistent with urban/industrial discharges. The Central Eastern and Western Adriatic has also been extensively monitored. Concentrations of Pb and Hg are relatively high in the Mali Ston Bay (Croatia). High levels of Cd, Hg and Cu are found in the Kastela and Rijeka Bays, due to the discharge of untreated urban and industrial wastewaters. In Albania, Pb and Cu contamination above the mean levels were found in mussels collected in Vlora Bay and close to Durres, a known dumping site of industrial residues.

The **Central Mediterranean** is not well represented in the Database. A few samples from south Italy and Greece are available. High levels of Hg were found in the Gulf of Patras (Greece) and moderately high levels of Cu were also found at the Sicily coast.

Trace metal analysis in biota in the **Aegean-Levantine region** exhibited low values in the case of *Mytilus galloprovincialis*, with the exception of Cd, Pb and Cu in Turkey (Akçay and Ismir Bays) and Cd and Zn in Greece (Piraeus). In Israel, *Mactra corallina* was used as indicator organism which exhibited relatively high values of Cd and Cu in the northern coast (Haifa). In Egypt, the clam *Donax trunculus* was the preferred indicator, exhibiting low concentrations that could just reflect a different accumulation rate of metals. Information is missing for the southern and eastern countries

The red mullet (*Mullus barbatus*) showed rather uniform metals bioaccumulation through the region but certain stations exhibited high levels of Cd, Hg, Pb and Cu. The most significant feature is the uneven distribution of Cd, with significant hotspots in certain stations of Greek Islands (Crete, Mytilini and Chios). High values of Hg and Pb were also found in Crete (Chania Bay) and Cyprus (Larnaca and Limassol Bays). The levels of Zn are also high in the Avramikos, Saronikos and Thermaikos Gulfs (Greece). The area of Mersin (Turkey) exhibit high values of Cu and the mullets collected in the northern coast of Syria exhibit high concentrations of Zn and Cu. However, the limited number of stations precludes any further consideration.

Chlorinated compounds in bivalves and fish

Chlorinated pesticides have been extensively analyzed in Mediterranean biota since the inception of MED POL (UNEP, 1990). Mussels and mullets have been the most widely studied organisms in the whole basin as part of many case studies published in the literature and recently assessed on the occasion of the implementation of the Stockholm Convention (UNEP, 2002). However, it has been only since the last decade that they have been continually monitored, and data gathered in the MED POL Database, although data on mullets are limited to Cyprus and Turkey.

An additional number of individual studies is found in the literature. As it can be seen, the levels reported for samples collected during the 90s exhibit much higher values than the more recent ones. This may clearly reflect a decreasing trend resulting from the ban of this pesticide in the region but also that the assessment of the spatial distributions of chlorinated compounds in the Mediterranean should be based on published data around the last 5 years.

As shown in the following Table, concentrations of aldrin, dieldrin, endrin, lindane and hexachlorobenzene in *Mytilus galloprovincialis* are in the low ng g⁻¹ range. Concentrations of DDTs were one order of magnitude higher, with p,p'-DDE being, in general, the predominant component, although recent inputs of DDT in some areas cannot be ruled out.

Chlorinated pesticides in *Mytilus galloprovincialis*. Median (range) concentrations (ng g⁻¹ dw)

Eco-region	Aldrin	Dieldrin	HCB	Lindane	Σ DDTs
WESTERN MEDITERRANEAN	0.34 (0.01-11.10)	0.76 (0.01-4.54)	0.54 (0.07-8.60)	0.60 (0.01-20.95)	10.80 (0.24-322)
ADRIATIC	0.45 (0.001-8.00)	0.37 (0.001-7.70)	0.10 (0.005-3.00)	0.24 (0.001-88.4)	11.00 (0.01-9779)
CENTRAL MEDITERRANEAN	1.63 (0.27-3.00)	1.0 (0.4-26.0)	0.15 (0.12-0.15)	0.11 (0.1-1.40)	10.24 (0.40-26.0)
AEGEAN-LEVANTINE	3.96 (0.44-22.11)	3.61 (0.04-38.45)	0.25 (0.01-26.52)	--	9.85 (1.01-173)

Concentrations are higher in the Aegean-Levantine region for aldrin and dieldrin, and in the Western Mediterranean for HCB and lindane. However, the highest values of HCB and lindane are found in Turkey and Albania, with a number of important hot spots. In the case of DDTs, their median values are similar for the four eco-regions, around 10

ng g⁻¹ dw, but again extremely high values are found in the Adriatic, corresponding to Albania.

Despite the similarity of the mean DDT levels for the four eco-regions, available data indicate that contaminants are not uniformly distributed throughout the sub-regions. In the **Western Mediterranean**, areas of particular concern include estuaries (Rhône, Ebro), ports, bays and gulfs (Barcelona, Marseille-Fos, Bays of Algiers and Tunis, Naples, etc.). There is evidence that river inputs represent the most important source of pesticides entering the Western Mediterranean Sea.

In the Southern part of the W. Mediterranean, significant accumulation values of DDT were recorded at the Nador Lagoon (Morocco) and in Algiers Bay.

Coastal waters of the **Adriatic Sea** belong to unpolluted areas of chlorinated pesticides, with the exception of some stations from Albania. Concentrations up to 9779 ng g⁻¹ dw of total DDTs were found at the Durres and Vlora Bay, most probably due to the presence of stockpiles of obsolete pesticides in the country. Moderate concentrations of Lindane and DDTs were found in the Gulf of Trieste and the Marches region (Ancona), respectively.

Information on the **Central Mediterranean** is almost lacking, although the region seems relatively free of hotspots of chlorinated hydrocarbons in marine bivalves, at least according to the limited availability of data.

In the **Aegean-Levantine** eco-region, organochlorine compounds, mainly DDTs, were determined in mussels and red mullets across the Greek coastal waters and in some stations of Turkey and Cyprus. In all cases the concentrations of DDTs were quite low, although moderate concentrations of DDTs were present in a few stations of Turkey (Ismir Bay) and Greece (Amvrakikos Gulf), probably attributed to the intense agricultural activities in the area.

Mullus barbatus was also the indicator species used in the Aegean-Levantine Sea. The spatial distribution of DDTs revealed a homogeneous pattern (av. 12.4 ng g⁻¹), indicating no point sources of pollution, in accordance with the banning of these compounds since the 70s. The values for Cyprus and Turkey can also be considered in the low range.

PCBs occur in the vicinity of industrial and urban sites, as well as in major river mouths. The geographical distribution of concentrations (7 ICES PCB congeners) in the indicator organism *Mytilus galloprovincialis* is shown in the following Table. The median values show the higher levels in the Adriatic where the Albania samples are well above the average, with values up to 1500 ng g⁻¹ dw in one station of France.

PCBs in Mytilus galloprovincialis. Median (range) concentrations (ng g⁻¹ dw)

Eco-region	CB138	CB153	Σ7CBs
WESTERN MEDITERRANEAN	4.20 (0.07-566)	6.18 (0.16-603)	12.6 (0.30-1500)
ADRIATIC	6.10 (0.1-350)	4.80 (0.05-85.5)	90.84 (2.23-875)
CENTRAL MEDITERRANEAN	5.00 (0.30-23.0)	4.27 (0.70-38.0)	--
AEGEAN-LEVANTINE	0.49 (0.03-25.3)	0.77 (0.05-39.8)	2.61 (0.18-135)

In the **Western Mediterranean**, the baseline levels are high and the sites most affected are the areas of Barcelona, Marseille and the Ligurian Sea, from Livorno to Nice, including the Genova harbour, and at the mouths of Rhone and Ebro Rivers. It was clear that in western Mediterranean coastal areas, rivers and wastewater discharges are the major sources of PCBs.

Information on the **Central Mediterranean** is almost lacking but, apparently, it is relatively free of hotspots.

The **Adriatic Sea** has been extensively monitored for PCBs. It includes from unpolluted to moderately polluted coastal areas. Concentrations are low along the western coast, with areas with high concentrations in the eastern bank, along the coasts of Croatia and Albania.

In the case of **Aegean-Levantine** spatial analysis is limited to Cyprus (fish) and Turkey (mussels and fish). The concentrations in mussels (Turkey) were rather low. The values on *Mullus barbatus* in the MED POL database can also be considered in the medium-low range, taking into account the higher accumulation capacity of fish with respect to mussels.

PCBs in marine biota across the Greek coastal environment, determined within the Greek monitoring system, were also low, the highest contamination being observed in mussels collected from the Saronikos Gulf (industrial and urban effluents). On the other hand, bioaccumulation in fish revealed a homogeneous pattern indicating no point sources of pollution.

ii) Temporal trends

The detection of temporal trends in concentrations of contaminants in the marine environment has been one of the aims of the MED POL Programme. However, in a previous evaluation of the generated database (UNEP, 2005) it was concluded that the objectives preliminarily set, were not enough to achieve the temporal trend of any selected contaminant for a selected site. The major reason for this was the difficulty in analysing data, especially when normalization was not intended for reducing the intrinsic variance by taking into account the differences in morphology (e.g. sediment grain size) or composition (e.g. tissue fat content) of the samples.

Moreover, there are very few countries with more than five years ongoing programmes to fulfil the requirements of a temporal trend assessment, and the situation is even less satisfactory in the case of sediments that require at least a 10yrs period for evidencing and assessing significant variations.

Taking into account the limited data available, trends are just preliminarily assessed. The analysis has been mainly focused on *Mytilus galloprovincialis* (MG) and *Mullus barbatus* (MB), with the exception of Israel, where the clam *Mactra corallina* (MC) has been considered. Moreover, a few available time series for sediments from Israel (Haifa Bay) and France (Gulf of Lions) have also been considered.

A general downward trend of concentrations of **trace metals** is observed in sediment samples from the Haifa Bay, except for Cu. In the Gulf of Lions, the French RNO monitoring system has shown an apparent decrease of metals, particularly for Pb. Unfortunately, there is no information about the most polluted area of Marseille-Fos. On

the contrary, mercury, a critical contaminant in the Gulf of Trieste, do not show a decline of concentration even 10 years after the closure of the Idrija mercury mine.

In the case of mussels, the country median values do not exhibit clear trends for metals, with the exception of particular situations like Cd in Slovenia and in Morocco. However, in several cases there is an apparent decline of outlier values, such as in the case of Italy, which may reflect a general improvement on the hotspots.

Individual trends for representative stations of the different sub-regions do not exhibit specific tendencies but it can be concluded that, in general, concentrations are relatively constant or declining. In the Western Mediterranean few stations, usually corresponding to those exhibiting the higher values (e.g. Marseille, Fos and Pombino), are slightly increasing, although those of Genova and Naples show evidence of a decline. Data on trace metals for samples of *Mytilus galloprovincialis* collected in 21 stations along the period 1979-2006 within the French monitoring system (RNO) clearly show a general decline of concentrations during this time span.

In the Adriatic, the levels are not showing significant changes along the time. However, general upward trends are observed in the stations holding high values, particularly for Cd (e.g. Rijeka and Kastela Bays in Croatia and Durres and Vlora Bay in Albania). A long survey (1999-2010) performed by Slovenia in the Gulf of Trieste has shown that the concentrations of Hg in mussels have not declined during this period.

The coverage of Central Mediterranean and Aegean-Levantine regions is very limited. Levels of Cd, Hg and Pb in mussels decreased during the period 2001-2008 in the Bizerta Lake (Tunisia). In Israel, the levels found in clams during the last 10 years in Haifa Bay show a significant increasing trend, despite the concentrations of trace metals in sediments revealed a consistent decline. On the other hand, in Turkey the red mullet collected in three stations in the south of the country (Mersin, Goksu and Tirtan) revealed upward trends of metals.

The levels of **chlorinated pesticides** in mussels clearly evidence a decline over time which is consistent with the banning of production and use of these compounds. The median values in mussels from Croatia and France exhibit clear decreasing trends, as well as the outlier values in the latter. The only exception seems to be Albania (e.g. Durres and Vlora Bay) which is recognized to keep stockpiles of obsolete chlorinated pesticides. The concentrations in mussels of the Ismir Bay, identified as a hot spot in the region, also exhibit a significant increase, probably due to a less efficient management of the existing regulations concerning the stocking of these obsolete pesticides. In general, the decrease is faster for lindane and the other chlorinated pesticides than for DDT that appears to be consistent with the higher long-life of the latter.

The assessment of **PCBs** is more difficult because the lack of long-term consistent data mainly due to the change in concentration units (from Aroclor to individual congeners). The profiles obtained do not allow deriving any conclusion. However, the temporal trends for some individual stations generally reveal conservative or even increasing trends, probably reflecting an inefficient management of the existing regulations concerning their use and stocking.

In the Western Mediterranean, the stations evidencing relatively high levels of PCBs are still exhibiting increasing trends whereas those more pristine seem to be stable or decrease. For the other regions there is limited availability of data to allow drawing precise conclusions. Temporal trends from the Greek MED POL monitoring programme

also indicate no reduction of pollutant levels despite the ban, indicating continuous inputs into the coastal environment.

Similar trends were observed in the French monitoring network of coastal pollution using bivalves as sentinel organisms (IFREMER, 2001). In general, it was found that during the period 1979-1998 the decreasing trends were in the order: Σ DDT > HCHs > PCBs, which may reflect that the regulation of the use of these chemicals and, consequently, of the contaminant inputs to the sea was more efficient for DDT and lindane than for PCBs.

A recent assessment of the Mediterranean sediments contamination by persistent organic pollutants (Gomez *et al.*, 2007) has also concluded that a decreasing trend is more evident for DDTs than for PCBs, indicating a steady input of the latter in the Mediterranean Sea and the need for an improved management of their potential sources.

Conclusion

The MED POL monitoring database (MED POL.mdb) and the inter-linked web version (http://195.97.36.231/MED_POL/) constitutes a relevant source of information for assessing the state of the Mediterranean Sea. The efforts made during the MED POL Phase III and Phase IV have been successful in building up and improving this essential instrument of environmental policy. Although at the moment is hosting monitoring data of only 14 Mediterranean countries, and the portion of data for each component and country is uneven, it constitutes the most comprehensive record of monitoring data for the whole basin and, therefore, it should be consolidated.

However, from the above analysis, it must be stressed that there is a need to establish monitoring programs in many countries to fill the geographical data gaps, and ensure the continuation of existing temporal trend data. These programs must be able to generate comparable and accurate data, taking into account the intrinsic variability of the environmental matrices considered. For example, the adoption of normalization procedures which could account for the differences in sediment characteristics (organic carbon or Al contents) as well as the implementation of quality assurance/quality control procedures are considered essential.

A useful outcome of the database can be the establishment of background concentrations for the target compounds in Mediterranean biota and sediments, which is necessary in order to have reference values for comparison with field data.

Moreover, with the improvement and development of analytical techniques, the identification and quantification of emerging substances of potential concern for the marine environment because of their persistence, toxicity and bioaccumulation properties, is continuously increasing. They are believed to be ubiquitous but information on their occurrence in the Mediterranean is limited and should be improved.

Finally, the conceptual approach of the MED POL Program, updated with the recent knowledge and experience generated by the scientific community needs to incorporate relevant assessment tools for hazardous substances in marine sediments and biota. Specifically, there is a need to establish environmental assessment criteria (EAC) for the hazardous substances included in the MED POL database: trace metals, chlorinated pesticides and PCBs.

1. Introduction

1.1 Background

The implementation of the Barcelona Convention aiming at the reduction of anthropogenic impact to the marine/coastal environment down to a level that can be assimilated by the own system, requires continued efforts on the development of monitoring programmes and a step-by-step adoption of knowledge-based management decisions at the global and local scales.

When the Mediterranean Pollution Monitoring and Research Programme (MED POL) started in 1975 its main aim was the establishment of a network of Institutions undertaking marine pollution work and the collection of information regarding the level of pollution in the Mediterranean Sea. The monitoring activities covered heavy metals in marine biota (mainly mercury and cadmium), halogenated hydrocarbons in marine biota (mainly PCBs and DDTs), and petroleum hydrocarbons in seawater. The development and maintenance of these national monitoring programmes was the aim of the second phase of MED POL, adopted in 1981.

The assessments of the data collected during these two phases (1975-1995) revealed several problems and shortcomings related to sampling, analysis, management and interpretation, as well as temporal and spatial coverage that made data comparability difficult (UNEP, 1990, 1996). As a result, the MED POL programme was completely reformulated in 1995 and became fully operational during the year 2000 (Phase III, 1996-2005). More emphasis was given in this Phase to the managerial aspects of pollution control while it had a direct link with the implementation of the relevant protocols especially the Land-based Sources Protocol (UNEP, 1999). This represented a significant evolution from the previous Phases, which were almost entirely directed to pollution assessment.

The programme was divided into the assessment and control components. The assessment component aimed at the accurate evaluation of temporal trends using a small number of fixed coastal stations from the national monitoring programmes. Trend monitoring was also organised in areas under the direct influence of pollution sources: "hot spots" (intensively polluted areas) where control measures were or were to be taken. Monitoring included not only levels of pollutants in the marine environment but also loads and biological effects.

Eleven national programmes were finalized during the 1999-2004 period (Table 1.1). Unfortunately, the geographical coverage of the coastal waters and hot spots of the region was not sufficient for an adequate assessment of pollution (UNEP, 2005). It is particularly worth noting that some of the countries which did not participate in the MED POL monitoring programme were European Countries that already operate well established and functioning monitoring networks.

A major outcome of this Phase was the setting up of the Database. Reference could be made to the previous major documents (UNEP(DEC)/MED WG.202/2, rev.9/4/2002 and UNEP(DEC)/MED WG.243/3, 2003) for the set up of the MED POL Phase III Monitoring Database (MED POL.mdb) and the inter-linked web version (http://195.97.36.231/MED_POL/).

Table 1.1. Status of National Monitoring Programmes

<i>Country</i>	<i>Drafted</i>	<i>Finalized</i>	<i>Revised</i>
Albania	1998	1999	2003
Algeria	2001, 2004	2004	
Croatia	1998	2000	2002
Cyprus	1998	1999	2002
Greece	1999	2000	2003
Israel	2002	2002	
Morocco	1999, 2003	2004	
Slovenia	1998	1999	2002, 2004
Syria	2000, 2003	2003	
Tunisia	2001	2001	
Turkey	1999	2000	2003

The critical analysis of the results, achievements, deficiencies and experience of the MED POL Phase III, particularly those related directly to the practical aspects of pollution control, were used in designing the operational details of a realistic and well focused Phase IV (2006-2010) (UNEP, 2005).

In this respect, monitoring was better integrated into the scope of MAP and used as a tool to:

- (i) continuously assess the state and trends of the marine and coastal environment and the effects of pollution;
- (ii) follow up and assist the countries' implementation of the LBS, Dumping and Hazardous Wastes Protocols; and
- (iii) track the implementation of the SAP and national action plans of pollution reduction.

The incorporation of Spain, France and Italy to the monitoring network during Phase IV represented a significant step forward in the implementation of MED POL. Data collection and handling, reporting and data management policies and procedures were enforced.

Moreover, the conceptual approach of the Program was updated with the recent knowledge and experience generated by the scientific community, considering the marine environment in a holistic way, i.e. as an integral whole consisting of the sea area and the adjacent terrestrial coastal area, as well as with the application of the ecosystem-based approach to environmental protection, including pollution control. Figure 1 shows the proposal of dividing the Mediterranean into four eco-regions.

These new approaches would require the formulation and adoption of additional Mediterranean-wide environmental quality objectives (EQO) and standards (EQS) for different "uses" of the sea to determine locally applicable effluent standards and input limits. The adoption of specific Mediterranean EQOs and EQSs are essential for the risk assessment of the pollution. However, they are not available and their definition is not an easy task.



Figure 1. The Mediterranean MED POL eco-regions

1.2 Objectives and scope

The scope of the present work is to prepare a Thematic Assessment Report on Hazardous Substances in the Mediterranean coastal environment, including sediment and biota contamination, and comment on possible pollution trends (not necessarily statistically supported).

The analysis will cover the full period of MED POL Phase III and Phase IV, until 2010, and will be based on the MED POL database. The database (MED POL.mdb) is hosting at the moment monitoring data from 15 Mediterranean countries (Annex I) and although the content is highly variable and the portion of data for each component and country is uneven it constitutes a relevant source of information. However, international literature will also be reviewed and where longer-term time series are available, in country databases or in earlier phases of MED POL, relevant data will be considered in the assessment.

An initial assessment of the state of marine pollution in the Mediterranean based on the MED POL database was made in 2009 and presented in the MED POL Review meetings of monitoring activities (UNEP/MAP, 2009a). However, following the recommendations from the countries, this assessment was to be up-dated using additional data (2009 and 2010 monitoring), revised monitoring data from previous years, which were sent by the countries, as well as additional scientific information included in the four sub-regional assessment reports, which were prepared in the framework of the gradual application of the Ecosystem Approach for the management of human activities in the Mediterranean by MAP (UNEP/MAP, 2010b).

The final Thematic Assessment Report on Hazardous Substances Pollution in the Mediterranean Sea will be a major building block, in view of the preparation of a Quality Status Report for the Mediterranean by UNEP/MAP in the next biennium (2012-2013).

2. Methodology

2.1 Data compilation

Data from the MED POL Database regarding trace metals, chlorinated pesticides and PCBs in the different marine compartments have been assembled for the purpose of the present assessment (Annex II).

In principle, the attention was focused on sediments and biota, as these are the most indicative compartments of the state of the environment. Moreover, only representative data was used, basically considering the existence of sufficiently large datasets. In this respect, sediments were only found suitable for the assessment of trace metals at regional level. Although it is difficult to assure the quality of the provided data, apparently inconsistent or not traceable data were disregarded.

The selected data is summarized in sections 2.1.1 (trace metals), 2.1.2 (chlorinated pesticides) and 2.1.3 (PCBs), where an overview of the number of samples by matrix and country, as well as the temporal series by country is provided. In order to enlarge the data base, data from the Italian and French monitoring systems¹ were incorporated (see Tables 2.4 and 2.7).

In the case of biota, the Database includes a large number of marine species, but the more abundant groups are bivalves and fish (Table 2.2). The present report is focused on one bivalve (*Mytilus galloprovincialis*) and one benthic fish (*Mullus barbatus*) as they are the more common and widely analyzed species in the region, with the exception of the clam *Mactra corallina* (MC) that has been primarily used in Israel, and *Donax trunculus* (DT) in Egypt..

As far as the parameters are concerned, the selection of metals has been performed on the basis of those considered of priority concern, namely Cd, Hg, Pb, Zn and Cu. Among chlorinated hydrocarbons, the study has been primarily focused on the families of DDTs and PCBs, as the more representative POPs. Lindane has also been considered taking into account the total number of samples and the geographical and temporal coverages.

In order to harmonize the database, all concentrations are recorded in $\mu\text{g g}^{-1}$ or ng g^{-1} dry weight. In cases where they were reported in other units or in wet weight basis, they were converted accordingly.

In general, the DDT concentrations refer to the sum of p,p'-DDT, p,p'-DDE and p,p'-DDD, although individual concentrations were also considered. In the case of PCBs, concentrations have been reported historically as Aroclor equivalents, using the mixture that better compares with the observed profiles. However, since the 80's, concentrations have been mainly expressed as individual congeners. Then, in order to harmonize data, the seven individual congeners proposed by ICES (PCB IUPAC # 28, 52, 101, 118, 153, 138 and 180) were selected. The conversion was performed by means of the average weight composition of individual PCB congeners in the Aroclor formulations (Safe *et al.*, 1985; Schultz *et al.*, 1989; Larsen *et al.*, 1993; Frame *et al.*, 1996; Frame, 1997). According to this, for example, the seven PCB congeners recommended by ICES represent, as an average, 29.2% of the Aroclor 1260 mixture.

¹ SIDIMAR (www.minambiente.it); RNO (<http://wwz.ifremer.fr/envlit/>)

In summary, after selection and harmonization of data, a total of **34738 observations**, corresponding to more than 400 stations monitored during the MED POL Phases III and IV (1999-2010) have been included in the present assessment. Moreover, an in-depth literature survey was performed in order to complement the national observations.

2.1.1 Trace metals

2.1.1.1 Selected data

Table 2.1. Number of samples of selected trace metals by matrix and country.

Country	SEDIMENTS					Total	BIOTA					Total
	Cd	HgT	Pb	Zn	Cu		Cd	HgT	Pb	Zn	Cu	
ALB							41	41	41	41	41	205
CRO	24	20	24	24	22	114	212	172	212	212	199	1007
CYP							35	52	31	18	18	154
EGY	30	30	29	30	29	148	17	16	17	17	17	84
FRA	13	13	13			39	512	512	512	512	512	2560
GRE	197	151	189	165	157	859	367	81	92	407	407	1354
ISR	143	152	144	186	161	786	282	1255	44	1390	1381	4352
ITA	113	496	555	555	554	2273	508	472	503	506	508	2497
MOR	5		12	12	12	41	58	50	56	5	5	174
SLO						0	80	84				164
SPA						0	356	343	356	356	356	1767
SYR	8		8	8	8	32	6		6	6	6	24
TUN	13	13	13			39	119	118	114			351
TUR	219	233	135	233	217	1037	352	352	38	352	351	1445
Total	765	1108	1122	1213	1160	5368	2945	3548	2022	3822	3801	16138

Table 2.2. Number of biota samples for selected trace metals by classes of species.

Class / Sp.	Cd	HgT	Pb	Zn	Cu	Total
BIVALVES	2245	2610	1955	2585	2565	11960
<i>AT</i>	9	1		11	11	32
<i>CH</i>	3			3	3	9
<i>DT</i>	45	315	53	314	309	1041
<i>MC</i>	108	324		388	379	1199
<i>MCC</i>	16	21		38	38	113
<i>MCS</i>	12	2		12	12	38
<i>ME</i>	24	24	24			72
<i>MG</i>	1916	1814	1771	1791	1780	9072
<i>PA</i>	3	2	3	17	17	42
<i>PP</i>	1	1	1	6	6	15
<i>RD</i>	100	98	95	1	1	295
<i>VA</i>	8	8	8	4	4	32

FISH	692	932	61	1229	1228	4142
BB	149	9	10	154	154	476
DS	25	41		49	49	164
EAE	2	1		4	4	11
LM	12	44		56	56	168
MB	487	775	50	911	910	3133
MS	7	18		24	24	73
PGE		18		6	6	30
SAU		6	1	1	1	9
SR	4	5		15	15	39
UM	6	15		9	9	39
Total	2937	3542	2016	3814	3793	16102

AT	<i>Acanthocardia tuberculata</i>		
CH	<i>Chrysaora hysoscella</i>		
DT	<i>Donax trunculus</i>	BB	<i>Boops boops</i>
MC	<i>Mactra corallina</i>	DS	<i>Diplodus sargus</i>
MCC	<i>Mactra corallina corallina</i>	EAE	<i>Epinephelus aeneus</i>
ME	<i>Mytilus edulis</i>	LM	<i>Lithognathus mormyrus</i>
MG	<i>Mytilus galloprovincialis</i>	MB	<i>Mullus barbatus</i>
PA		MS	<i>Mullus surmuletus</i>
PP	<i>Perna perna</i>	PGE	<i>Pagellus erythrinus</i>
RD	<i>Ruditapes decussatus</i>	SAU	<i>Sparus auratus</i>
VA		SR	<i>Siganus rivulatus</i>
		UM	<i>Upeneus moluccensis</i>

Table 2.3. Temporal series of metals in biota (B) and sediments(S) by country.

Country	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010
ALB			B		B	B	B	B	B			
CRO		B	B	B, S	B, S	B, S	B, S				B, S	
CYP	B		B			B	B					
EGY								B, S			B, S	B, S
FRA	B	B	B	B	B	B	B	B, S	B, S	B, S	B, S	
GRE	B, S	B				B, S	B, S	B	B, S			
ISR	B, S											
ITA			B, S									
MOR	B	B	B	B	B	B	B	B	B, S			
SLO	B	B	B	B	B	B	B	B	B	B	B	B
SPA						B	B	B	B			
SYR									B, S			
TUN			B	B	B	B	B	B, S	B, S	B, S	B, S	
TUR	B, S				B, S							

2.1.2 Chlorinated pesticides

2.1.2.1 Selected data

Table 2.4. Number of biota samples (MG and MB) for OCPs by country.

Parameter	ALB	CRO	CYP	FRA ⁺	ITA*	SPA	TUR	Total
DDD	41	84	46	200	332	237	121	1061
DDE	41	108	63	207	372	276	113	1180
DDT	40	104	73	200	301	179	128	1025
DDTs	40	108	46	243	374	276	152	1239
ALD		56	45	9	169	9	66	354
DIE		44	13	8	205	229	88	587
END			13			27		40
HCB		35	53		252	60	110	510
LIN	29	52	45	225 ⁺	194	56		601
Total	191	591	397	1092	2199	1349	778	6597

⁺ Data obtained from the RNO database.

* Data obtained from the SIDIMAR database.

Table 2.5. Number of biota samples for OCPs by species.

Parameter	MG	MB	Total
DDD	927	134	1061
DDE	1028	152	1180
DDT	857	168	1025
DDTs	1087	152	1239
ALD	258	96	354
DIE	499	88	587
END	27	13	40
HCB	374	136	510
LIN	556	45	601
Total	5613	984	6597

Table 2.6. Temporal series of OCPs in biota (B) by country.

Country	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010
ALB			B	B	B	B		B	B			
CRO	B	B		B	B	B	B				B	
CYP	B	B	B	B								
EGY											B	B
FRA	B	B	B	B	B	B	B	B	B		B	
ITA*			B	B	B	B	B	B				
SPA						B	B	B	B			
TUR					B		B	B	B	B	B	

2.1.3 PCBs

2.1.3.1 Selected data

Table 2.7. Number of biota samples (MG and MB) for PCBs by country.

Parameter	ALB	CRO	CYP	ITA*	FRA	SPA	TUR	Total
CB28	14	15	42		183	175	142	571
CB52	23	19	32	332	206	208	149	969
CB101	11	24	5		206	273	149	668
CB118		8			207	266	149	630
CB138	18	27	34	444	207	276	149	1155
CB153		27	33	459	207	276	149	1151
CB180	18	25	34		207	237	149	670
CB7	33	60	49		254	276	149	821
Total	117	205	229	1235	1677	1987	1185	6635

* Data obtained from the SIDIMAR database.

Table 2.8. Number of biota samples for Σ 7PCBs by species and country.

Country	MG	MB	Total
ALB	33		33
CRO	60		60
CYP		48	48
FRA	254		254
SPA	276		276
TUR	51	98	149
Total	674	146	820

Table 2.9. Temporal series of PCBs in biota (B) by country.

Country	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009
ALB					B	B		B	B		
CRO		B		B	B						B
CYP		B	B	B	B		B				
ITA	B	B	B	B	B	B	B				
FRA			B	B	B	B	B	B			B
SPA						B	B	B	B		
TUR					B	B	B	B	B	B	B

2.2 Overall data assessment

A general scheme of the data handling from the Database (MED POL.mdb) to the final outputs of the report is shown in Figure 2. The key component for the present and future assessments of the Database will be the generated Excel-selected data file.

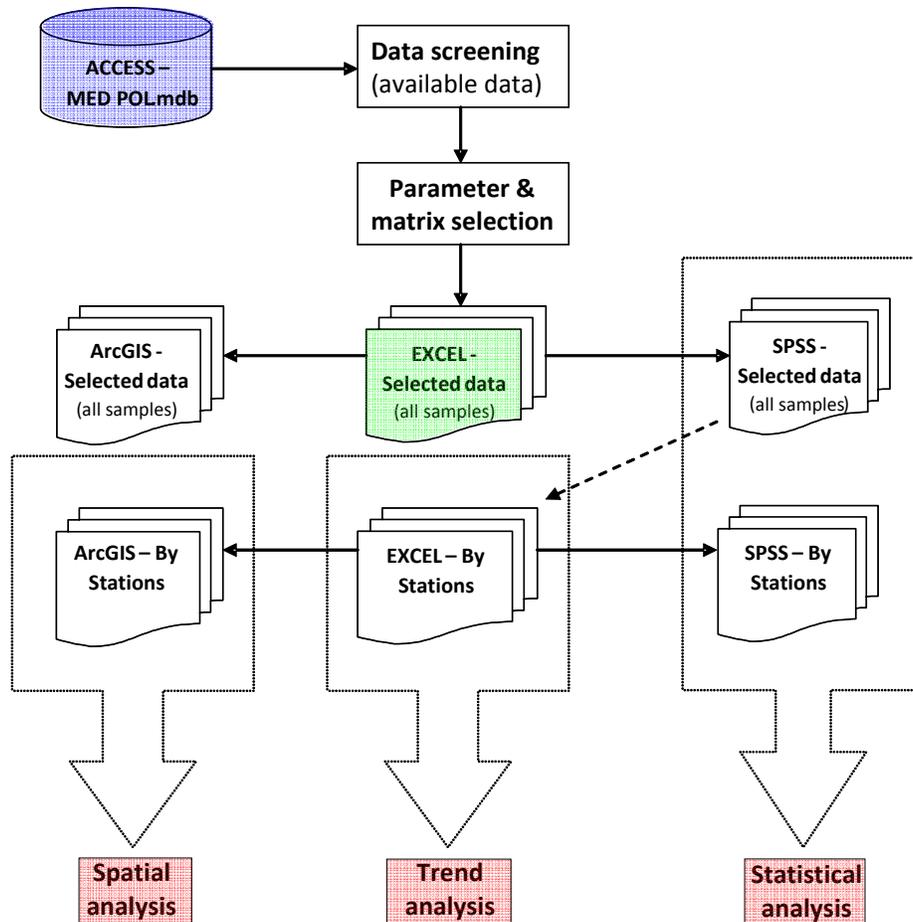


Figure 2. Flow chart of MED POL data analysis.

2.2.1 Spatial analysis

The geographical position of each sample (longitude and latitude) was obtained directly from the database.

Geographical analysis of the data included the use of GIS based mapping methods (software ArcGis 9.2), where information on positions was considered according to the World Geodetic System 84 (WGS-84).

As shown in Figure 3, good coverage was found for the Northern part of the Mediterranean basin while only a small number of sampling points was found in the Southern riparian countries.

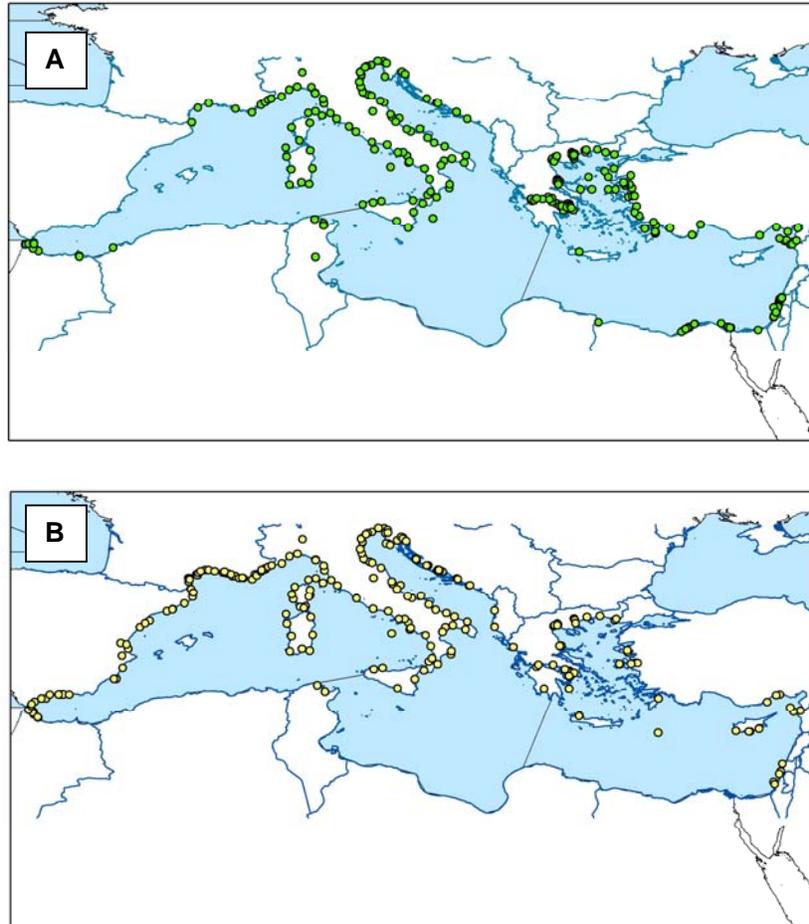


Figure 3. Map of MED POL stations. A) sediments. B) biota

The mapping of the concentrations of the selected contaminants in the different matrices was based on the mean values for each station.

The final assessment has taken into account the four sub-regions defined by the Contracting Parties to fulfil the ecosystem approach process (Figure 1).

Supporting information on eco-regions has been obtained from ICES Data Centre (<http://www.ices.dk/aboutus/icesareas.asp>).

2.2.2 Temporal trends

Nowadays, a variety of tests are available to analyze data records for trends assessment. The OSPAR Convention proposed three methods (Mann-Kendall, linear

regression, lowess smoother) for this purpose. Mann-Kendall is the most robust to outliers, but in case of a linear trend, the linear regression has more power and therefore is selected first. Since nature is not always linear, the smoother is taken to detect a non-linear trend. The idea is to take the benefits of all methods, because there is not only one method which always offers the best analysis.

To this end, the software called Trend-y-tector was particularly used in their assessments of monitoring data (OSPAR, 2000). The software was developed by the Netherlands National Institute for Coastal and Marine Management (RIKZ) and adopted by the ICES Working Group on Statistical Aspects of Environmental Monitoring (WGSAEM).

The Trend-y-Tector is easy to use and offers a structured methodology for deciding whether there is a significant (downward) trend or not. Some stations that are believed to be representative of the trends in each country/eco-regions have been selected for indicative purposes.

2.2.3 Statistical data analysis

Statistical data treatment has been carried out with the SPSS 17.0 package (SPSS Inc., Chicago, U.S.A.). Concentration ranges, means, medians and lower and upper quartiles (Box-and-Whisker plots) for each chemical were analysed in order to characterise the populations and identify the extreme values. Mean values were used for characterizing the different stations. On the other hand, median values were generally considered as a better representation of the population position in largely asymmetric distributions as those found at regional/national level in the present survey. Values located in the upper limit at a distance higher than 1.5 times the interquartile range of data (IQR, grouping the 50% of data, between the 75th and the 25th quartile) were considered as outliers.

The normality of the datasets for the different populations has been evaluated by means of the Kolmogorov-Smirnov test. Since concentration values were not normally distributed, non-parametric statistics was applied. The Mann-Whitney test was used for comparing two data sets and the Kruskal-Wallis test when more than two groups were considered. In these two non-parametric tests, the differences among groups of data were considered statistically significant when the p-value was lower than 0.05 (probability of 95%). When studying the relationship between two variables, the non-parametric Spearman correlation coefficient was used, setting the level of probability also at 95%. On the other hand, the Pearson correlation coefficient was calculated for assessing linear regressions of log-transformed data, which were checked to be normally distributed.

3. Results and discussion

3.1 Occurrence and geographical distribution

A comparison of the levels of trace metals found in sediments and marine biota, and chlorinated pesticides, notably DDTs and lindane, and PCBs, in marine biota of the different Mediterranean countries and eco-regions is summarized in the following sections (3.1.1 to 3.1.3).

In general, good data coverage is found for the Northern part of the Mediterranean basin while only a small number of sampling points are present in the Southern riparian countries. Therefore, data has been complemented with available information from the literature as well as other MAP reports. In comparing data, special use is made of the median concentration values because they constitute a better representation of the left-skewed populations studied.

3.1.1 Trace metals in sediments

3.1.1.1 General distribution

An overview of the concentration ranges of Cd, total Hg, Pb, Zn and Cu in coastal sediments of the region is summarized in Table 3.1. As it is shown, Cd and Hg exhibit values several orders of magnitude lower than Zn, being Pb and Cu in an intermediate and similar position. The median values can be used as reference for comparison purposes. In general, these values are in the lower range than those reported in previous assessments derived from MEDPOL I and II (UNEP, 1996). For example, the reported values for Cd, Pb, Zn and Cu were, respectively, of 0.02-64, 3-3000, 1.7-6200 and 0.6-1890 $\mu\text{g g}^{-1}$ dw (UNEP, 1996).

Table 3.1. Statistics of trace metal concentrations ($\mu\text{g g}^{-1}$ dw) in sediments

Metal	n	Mean	Median	Min	Max	SD
Cd	765	0.46	0.13	0.01	18.50	1.40
HgT	1108	0.76	0.13	0.001	166.89	6.74
Pb	1122	27.64	14.30	0.03	1032.60	52.73
Zn	1213	82.23	55.76	0.05	1505.35	125.47
Cu	1160	26.12	15.62	0.01	962.68	58.37

The analysis of levels by countries, shown in Table 3.2 and Figure 4, reveals the occurrence of some stations with high values of Hg, Pb, Cu and Zn in Croatia. A wide range of values for all metals was found in Italy, particularly for Pb. Average values of Cd, Zn and Cu in the higher range were found in Morocco and Syria whereas in Egypt and Israel were in the lower end. Values of Pb and Zn were also high in Greece, and of Zn and Cu in Turkey. However, as data has not been normalised to the sediments lithology (e.g. grain size or lithium and aluminium contents), the values given cannot be adequately assessed. Moreover, some of them should be quality assured as they are far outliers from the normal distributions. For example, the very high levels of Hg and Pb found in some stations in Croatia (Figure 4) could deserve further evaluation.

Table 3.2. Mean and median concentrations of trace metals in sediments ($\mu\text{g g}^{-1}$ dw) by country.

Country	Cd		HgT		Pb		Zn		Cu	
	Mean	Median	Mean	Median	Mean	Median	Mean	Median	Mean	Median
CRO	0.64	0.24	16.73	0.55	118.90	38.59	215.63	102.09	57.33	35.86
EGY	0.19	0.20	0.98	0.25	5.57	5.26	21.60	19.09	8.81	7.91
FRA	0.05	0.05	0.04	0.01	23.15	26.00	-	-	-	-
GRE	0.41	0.19	0.23	0.12	45.21	30.17	114.90	54.23	35.09	20.10
ISR	0.32	0.08	0.22	0.10	8.94	5.27	34.47	10.07	10.96	2.31
ITA	0.99	0.21	0.71	0.10	22.40	12.28	73.83	53.10	16.01	11.56
MOR	2.78	1.60	-	-	16.65	16.02	89.59	91.45	50.96	45.40
SYR	3.38	3.78	-	-	30.59	30.87	110.00	102.92	22.35	17.14
TUN	0.65	0.48	0.26	0.19	40.68	28.36	-	-	-	-
TUR	0.18	0.06	0.22	0.14	32.65	26.16	109.95	90.81	54.62	27.22

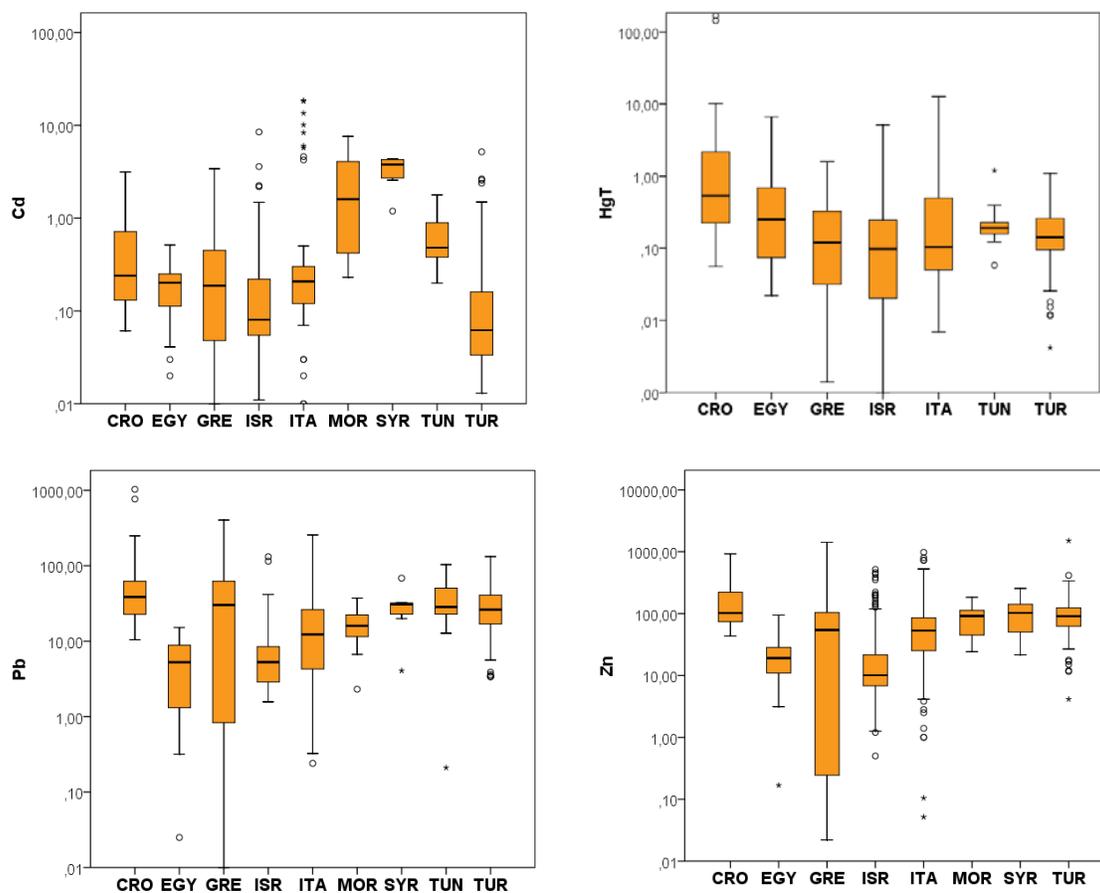


Figure 4. Concentrations of trace metals in sediments ($\mu\text{g g}^{-1}$ dw) by country.

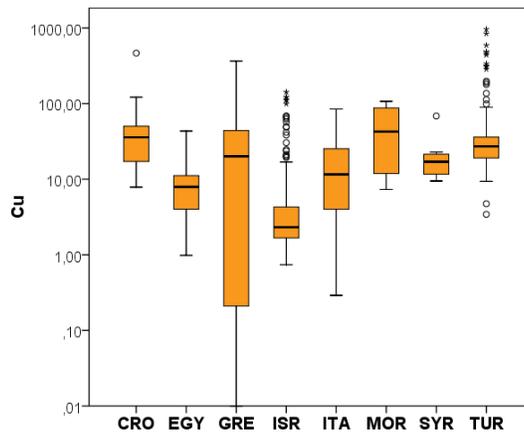


Figure 4 (cont.). Concentrations of metals in sediments ($\mu\text{g g}^{-1}$ dw) by country.

A closed look of the mean levels of Cd, total Hg, Pb, Zn and Cu in all stations is shown in Figure 5. As it can be seen, the spatial coverage is very sparse; the Western Mediterranean and the southern Central and Aegean-Levantine eco-regions being underrepresented. A detailed discussion of these data is made in the following section.

Figure 5. Mean concentrations of trace metals in sediments ($\mu\text{g g}^{-1}$ dw).

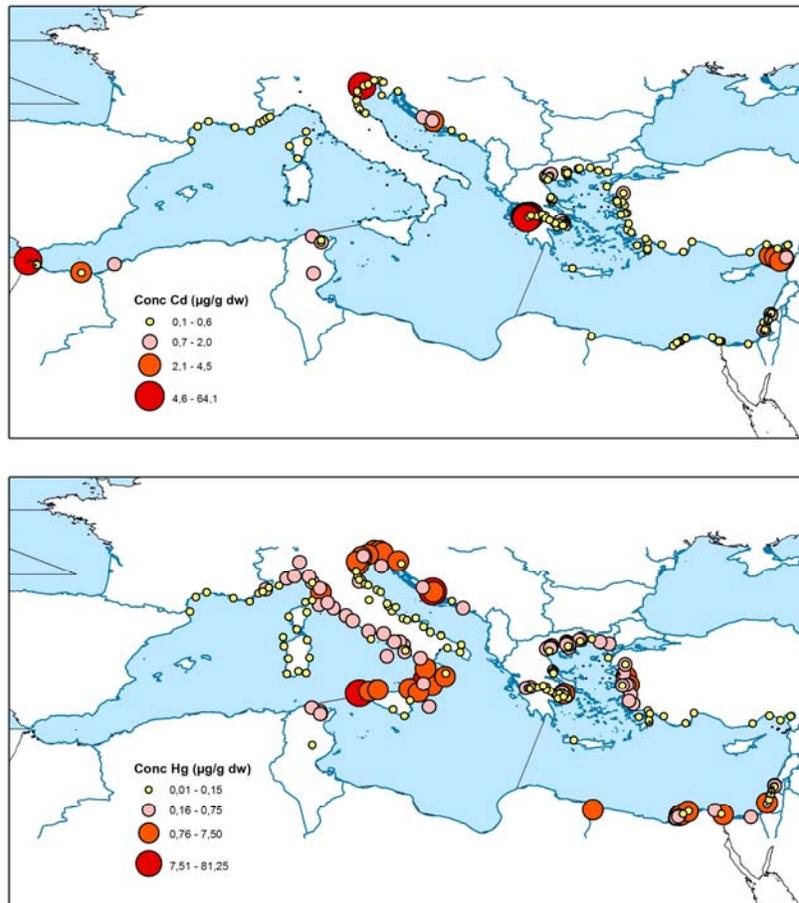
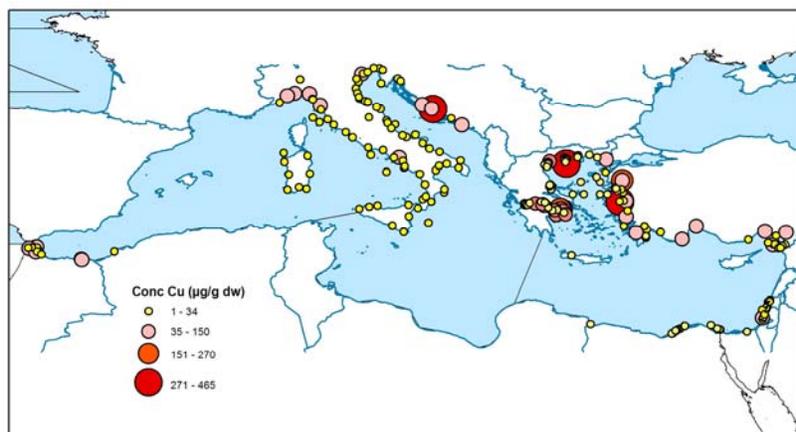
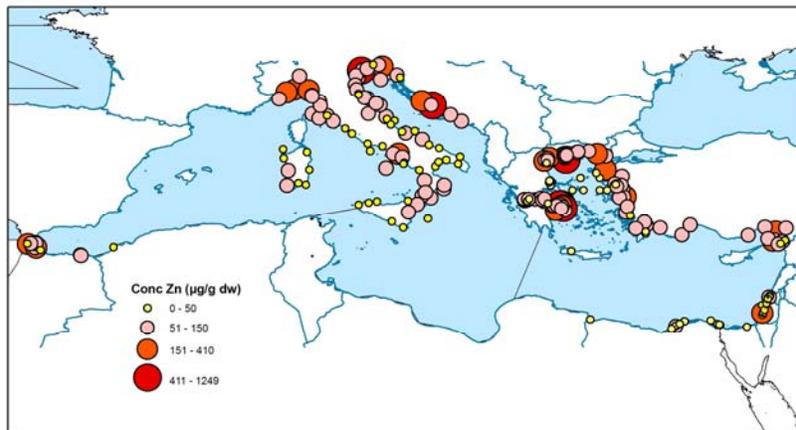
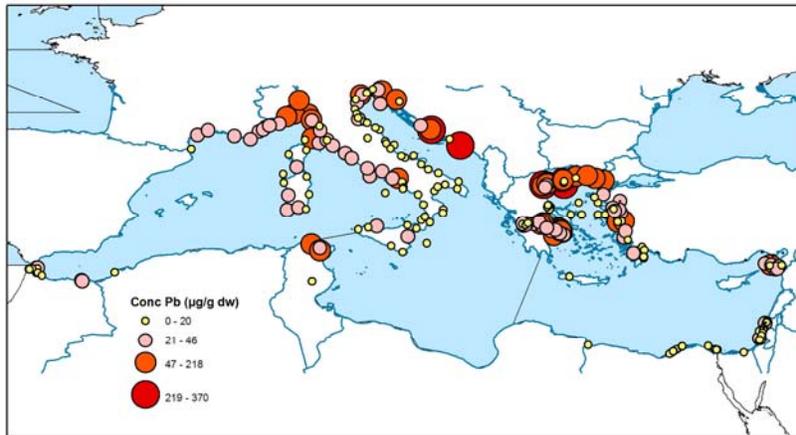


Figure 5 (cont.). Mean concentrations of trace metals in sediments ($\mu\text{g g}^{-1}$ dw).



3.1.1.2 Distribution of trace metals in sediments within eco-regions

A summary of the data corresponding to trace metals in sediments from the four Mediterranean eco-regions is shown in Table 3.3 and Figure 6. In general, the Western Mediterranean is the region where concentrations are higher, followed by the Aegean-Levantine and the Adriatic. However, these values are far from representative of the sub-basins because data is missing from large areas in the Western and Central Mediterranean (e.g. Spain, Algeria and Lybia), as well as in the Aegean Levantine Sea (e.g. Algeria and Lebanon). Only the Adriatic is well covered.

Angelidis *et al.* (2011) have recently reported data from two sediment cores collected at the deep Balearic basin (Western Med) and the Cretan Sea (Eastern Med), providing evidence for the accumulation of Cd, Pb and Zn in the top few centimetres. From the bottom data, background values can be estimated (Table 3.3). It is interesting to notice the close similarity of these values with those issued from the medians of the whole data set, which may provide a guideline for the assessment of hotspots. For comparison purposes, the general accepted values for the earth crust are also given. As it can be seen, the high levels of Zn and Cu found in some areas can be attributed to natural factors.

Table 3.3. Median and concentration ranges of trace metals in sediments ($\mu\text{g g}^{-1}$ dw)

Eco-region	Cd	HgT	Pb	Zn	Cu
WESTERN MEDITERRANEAN	1.60 (0.23-7.61)	0.16 (0.02-12.6)	19.40 (0.24-256)	50.10 (1.0-731)	13.90 (0.68-107)
ADRIATIC	0.21 (0.01-18.5)	0.10 (0.01-166.9)	9.83 (0.39-1033)	67.00 (5.0-980)	16.31 (1.39-465)
CENTRAL MEDITERRANEAN	0.09 (0.01-2.80)	0.05 (0.00-6.00)	4.39 (0.01-103.4)	32.66 (0.02-205)	5.35 (0.01-67.5)
AEGEAN- LEVANTINE	0.10 (0.01-8.47)	0.15 (0.00-6.70)	16.89 (0.01-404)	59.34 (0.02-1505)	18.52 (0.01-962)
Western Med*	0.07	-	15.0	63.2	36.8
Eastern Med*	0.08	-	14.3	54.6	37.5
Earth crust	0.15	-	11.5	92	23

* Estimated background values, from Angelidis *et al.* (2011)

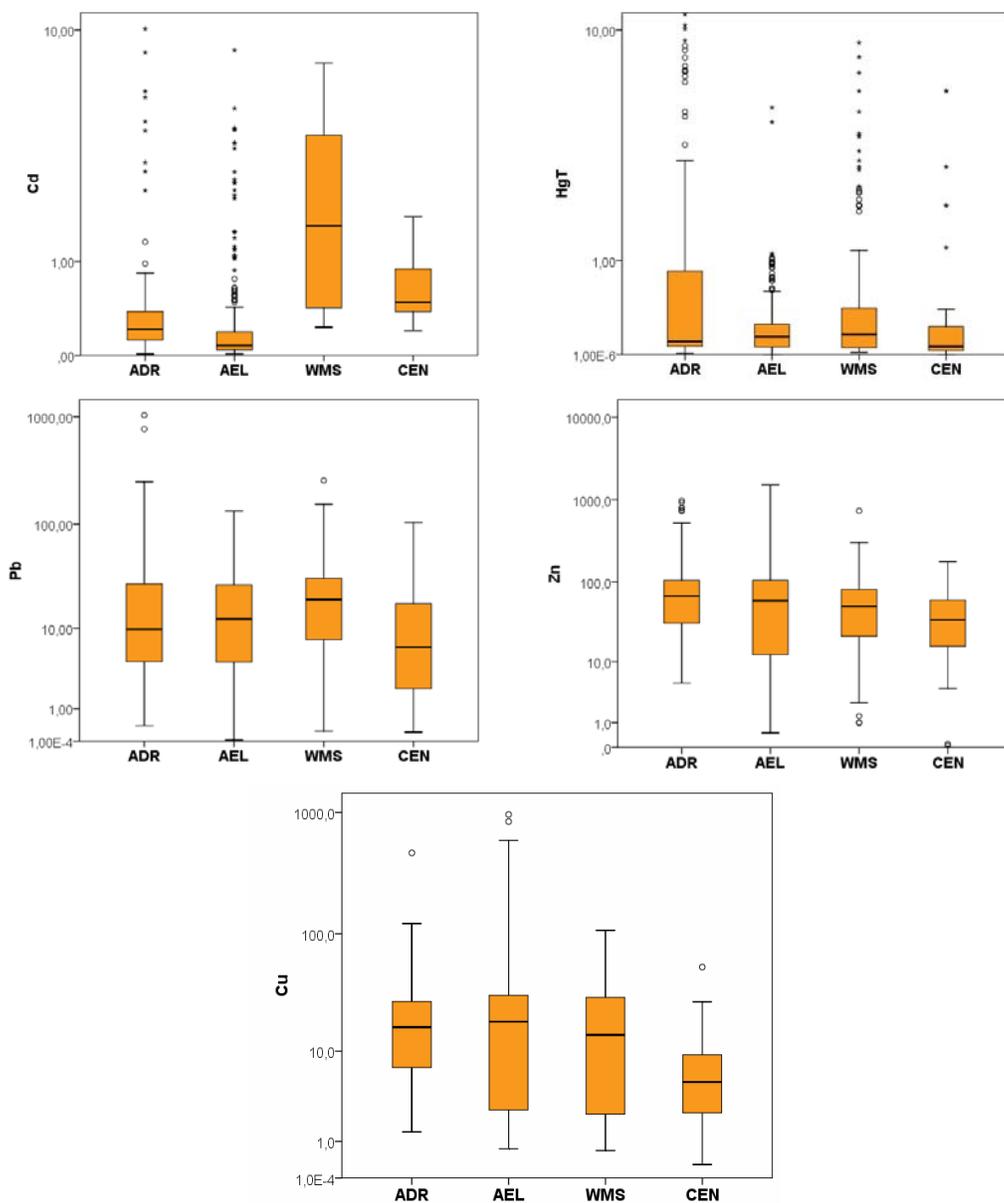


Figure 6. Concentrations of trace metals in sediments ($\mu\text{g g}^{-1}$ dw) from the different Mediterranean sub-regions.

Additional data available from the literature, summarized in Table 3.4, enable to complement the whole picture. Besides a few studies covering open costal areas of the NW Mediterranean and Aegean Sea (Lafabrie et al., 2007; Roussiez et al., 2006; Bertolotto et al., 2005; Karageorgis et al., 2005) or Algeria, Egypt and Israel (Goldsmith et al., 2001; Kress et al., 2004; Alomary et al., 2007; El Nemr et al., 2007), studies have been focused on coastal lagoons (e.g. Mar Menor, Berre, Venice, Akyatan, Mariut, etc.) and gulfs and bays (e.g. Vlora, Taranto, Tunis, Sfax, Ismir, Abu Qir, El-Mex, etc.), which have been recognized as the most contaminated sites. However, the lack of standardized methods in these studies, including normalization parameters (e.g. sediment lithium or aluminium contents), makes it difficult to compare data from different sources in order to draw consistent conclusions about spatial trends.

Table 3.4. Concentrations of trace metals in sediments in other studies (*Mean values. **Industrial site. ***Polluted site)

Location/Area	Year(s)	Stations / Samples	Concentration ($\mu\text{g/g dw}$)					Reference
			Cd	Hg	Pb	Zn	Cu	
Mar Menor Lagoon	2005	16 / 64	0.5 – 9.1	-	< D.L – 6.97	< D.L – 7.13	< D.L – 74	Maria-Cervantes et al. (2009)
Berre Lagoon	2002	17 / -	0.16 - 1.61	0.15 – 0.55	18 - 82	50 - 151	12 - 48	Accornero et al. (2008)
NW Mediterranean	2004 – 2005	3 / 9	0.03 – 0.40	0.02 – 0.56	18.67 – 44.50	-	-	Lafrabrie et al. (2007)
Gulf of Lions	2002	51 / -	0.36 (0.22 – 0.82)	-	39.2 (20.6 – 69.7)	102.2 (65.6 – 144.9)	22.7 (9.0 – 45.8)	Roussiez et al. (2006)
Ligurian Sea	1995	75 / -	0.03 – 1.13	0.02 – 0.47	15 – 150	13 – 610	2.3 – 74	Bertolotto et al. (2005)
Pozzuoli Gulf (IT)**	2004	37 / -	0.71 (0.01 – 4.70)	0.58 (0.01 – 2.90)	260 (21 – 1288)	539 (111 – 2525)	40 (6 – 165)	Romano et al. (2009)
Corsica coast	1999 - 2001	25 / -	0.02 – 0.14	0.001 – 0.269	6.0 – 53.8	6 – 84	0.8 – 31.1	Galgani et al. (2006)
Sardinia coast	2001 - 2006	8 / 96	0.07 (<0.03 – 0.13)	0.05 (<0.03 – 0.08)	5.6 (1.0 – 19.3)	10.8 (1.4 – 21.0)	2.7 (0.5 – 8.5)	Schintu et al. (2009)
Algerian coast	2005	18 / -	1.1 (0.1 – 2.3)	-	5.7 (1.3 – 11.5)	20.4 (5.3 – 45.7)	4.7 (1.1 – 10.4)	Alomary et al. (2007)
Venice Lagoon	1996	18 / -	0.2 – 5	1.2 – 1.4	38 – 114	101 – 1115	-	Bellucci et al. (2002)
Vlora Bay	2007-2008	34 / -	0.05 – 0.29	<D.L. – 3.06	9.65 – 20.1	59.3 – 109	22.3 – 46.1	Rivaro et al. (2011)
Taranto Gulf	-	19 / -	-	0.12 (0.04 – 0.41)	57.8 (44.7 – 74.8)	102.3 (86.8 - 129.0)	47.4 (42.4 – 52.3)	Buccolieri et al. (2006)
Sfax Bay	2005	67 / -	5.9 (5.5 – 7)	-	32 (18 – 88)	58.9 (39 – 117)	16 (13 – 29)	Gargouri et al. (2011)
Gulf of Tunis	2004 – 2005	38 / -	0.27 (0.07 – 0.67)	0.27 (0.09 – 0.53)	54.1 (18.8 – 98.8)	125 (75 – 249)	20.90 (7.28 – 89.30)	Ennouri et al. (2010)
NW Aegean Sea	1997 – 1998	112 / -	-	-	52 (17 – 265)	120 (33 – 429)	34 (4 – 108)	Karageorgis et al. (2005)
Lesvos Island	2004	4 / -	0.047 – 0.285	0.030 – 0.110	20.5 – 42.9	38 – 113	7.4 – 39.3	Aloupi et al. (2007)
Crete Island	1991	88 / 264	0.27 (0.04 – 1.86)	0.19 (0.03 – 2.04)	24.26 (8.26 – 63.00)	42.7 (6.9 – 125.3)	27.11 (2.07 – 126.1)	Poulos et al. (2009)
Izmir Bay	1996	25 / -	-	-	-	76.5 (25.6 – 154)	22 (4.26 – 45.2)	Kontas (2008)
Izmir Bay	1997 - 2002	28 / -	0.005 – 0.82	0.05 – 1.3	14 – 113	-	-	Kucuksezgin et al. (2006)
Izmit Bay	2002	8 / -	6.3 (3.3 – 8.9)	-	94.9 (23.8 – 178)	754 (500 – 1190)	89.4 (60.6 – 139)	Pekey (2006)
Akyatan Lagoon	2007 – 2008	7 / 21	< 0.005	-	19.5 – 33.6	54 – 102	14.4 – 31.3	Davutluoglu et al. (2010)
Southern coast Turkey	1996	2 / 20	-	0.17 – 4.75	6.23 – 413.5	< DL	5.19 – 143.4	Dogan-Saglamtimur et al. (2010)
Israeli coast ***	1992	20 / -	< D.L – 3.12	0.01 – 1.36	1.66 – 43.8	30.0 - 668	9.40 – 242	Kress et al. (2004)
Israeli coast	1995	16 / -	0.16 – 0.36	-	18.4 – 37.4	83.1 – 137.0	28.7 – 57.6	Goldsmith et al. (2001)
Egyptian coast	1999	20 /120-160	11.83 (3.07 – 53.41)	-	92.9 (49.9 – 219)	75.3 (16.6 – 166.5)	41.71 (6.94 – 192.5)	El Nemr et al. (2007)
Abu Qir Bay	2006	30 / -	1.08 (<0.200 – 3.54)	0.85 (<0.36 – 2.70)	34.6 (2.71 – 129)	71.4 (5.77 – 717)	15.1 (<0.320 – 127)	Khairy et al. (2010)

In any case, it is known that the north-western Mediterranean and the Adriatic are highly affected by intense human activities that can cause chemical contamination of coastal areas (EEA, 1999). Besides urban and industrial inputs, rivers and streams are major contributors of metals of anthropogenic or natural origin to coastal areas. Moreover, land-based sources of specific trace metals are not only set down at coastal sediments but may end up deep in the marine canyons (Palanques *et al.*, 2008). Atmospheric deposition may also contribute to the contamination of deep sea sediments (Angelidis *et al.*, 2011).

In the **Western Mediterranean**, major hotspots for metals, particularly Cd, Pb, Hg and Cu, are located in the areas of Marseilles-Fos and Toulon, in France (Sauzade *et al.*, 2007), and Cartagena-Valle Escombreras-Portman, in Spain (Benedicto *et al.*, 2008), holding important urban-industrial and mining areas, respectively. Sensitive areas are represented by the coastal lagoons (e.g. Berre, Thau and Mar Menor) (Accornero *et al.*, 2008; Maria-Cervantes *et al.*, 2009).

An extensive study performed in 2005, by IFREMER, in the framework of the French RNO, including 48 stations from the Gulf of Lions (Figure 7), showed high levels of Hg and Pb in the area of Marseille. The Thau Lagoon also exhibited high levels of Cd, Pb and Cu. The area influenced by the Rhone River discharges exhibited increased levels of all metals except Hg.

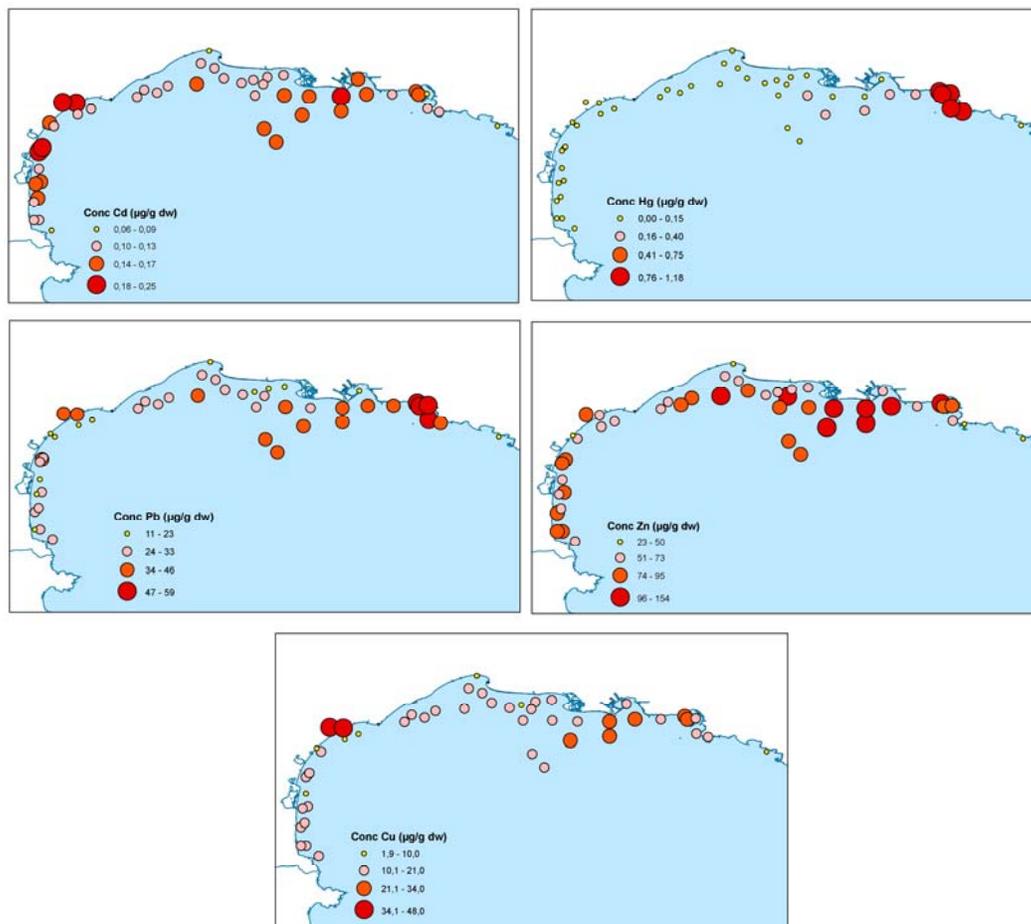


Figure 7. Concentrations of trace metals in sediments ($\mu\text{g g}^{-1}$ dw) from the Gulf of Lions (RNO, IFREMER, 2005).

The levels of Pb are important along the Italian coast, especially in areas around the Gulf of Genova (e.g. Savona, La Spezia, Genova, Livorno) that also show evidence of Zn accumulation, while high levels of Pb are found in Naples and of Cd and Zn in Palermo (Tranchina *et al.*, 2008; Lafabrie *et al.*, 2007). These pollution sources can be related to industrial and domestic wastes and harbour activities. Mercury levels are also high in the area of Messina and Palermo and Regio Calabria (Figure 8). The contributions related to tectonic sources are important in volcanic and geothermal sources near as southern Tyrrhenian Sea. These contributions could explain natural important levels of mercury in some islands of the basin. Two sites in SW Sardinia are also characterized by high levels of Cd, Pb and Zn that can be attributed to industrial and past mining activities (Mitis *et al.*, 2005).

Significant concentrations of Zn, Cu and particularly Cd (Figure 5) have been reported on the coast of Morocco (Tangier-Martil and Nador) (Benaoui *et al.*, 2004), whereas higher levels of Hg were found in Algeria (Algiers) (Taleb *et al.*, 2007; Soualili, 2008).

Trace metals have been extensively monitored in the **Adriatic Sea**. The Po River, draining the most industrialized part of northern Italy, is an important pollution vector in the area. As shown in Figure 7 and the close-up view of Figure 8, high levels of Cd, Hg, Pb and Zn are found in the northern Adriatic, including the Venice and Grado Lagoons. Mercury and Pb are also critical contaminants in the Gulf of Trieste and Rijeka Bay (Figure 8) (Covelli *et al.*, 2001; Horvat *et al.*, 1999; Hines *et al.*, 2000).

Data reveals higher values in the eastern than in the western coast. The occurrence of some discrete stations with high levels of Hg, Pb, Cu and Zn are found in Croatia, such as at Kastela and Martinska Bays. In Albania, Hg contamination from a former inland chlor-alkali plant is evidenced in sediments of Vlora Bay (Rivaro *et al.*, 2011).

The **Central Mediterranean** exhibits elevated levels of Hg in the Gulf of Taranto and in both the Tunisian and Italian coasts of the Strait of Sicily (Figure 5). However, an extensive study in the Ionian Sea revealed that mercury levels were generally comparable to those from other Mediterranean areas (around 50 ng g⁻¹ dw) (Di Leonardo *et al.*, 2008). The Tunis and Bizerta Lakes (Tunisia) also exhibits high contents of Pb (Figure 8). In the Hellenic coastal zone, the most elevated levels of such contaminants in sediments (e.g. Amvrakikos and Patraikos Gulfs) can be associated with the main sewage (domestic and industrial) outfalls (SoHeIME, 2005). Very high values of Cd have been found close to Patras (Figure 9). Information is lacking for the Lybian coast.

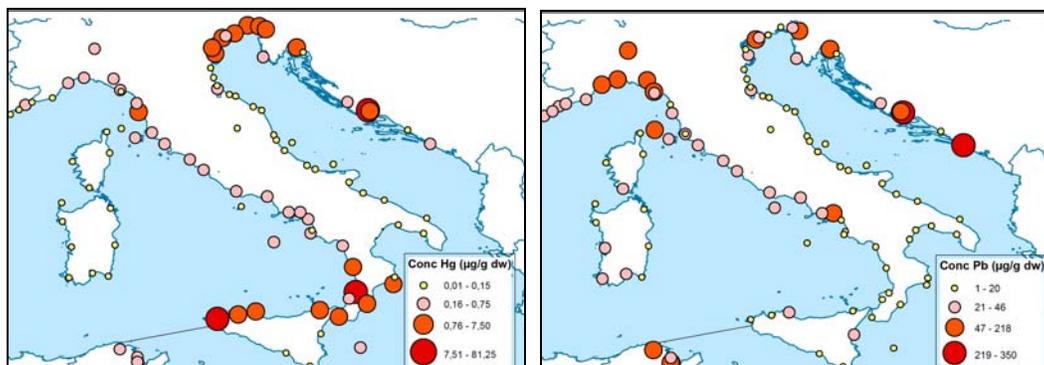


Figure 8. Mean concentrations of Hg and Pb in sediments ($\mu\text{g g}^{-1}$ dw) from the Tyrrhenian and Adriatic Seas.

The analysis of representative trace metals in sediments of the **Aegean – Levantine basin** revealed that, in general, values are in the lower range than those reported in previous assessments (derived from UNEP/MAP-MED POL I and II).

The coast of Turkey has been extensively monitored in MEDPOL III and IV, and high values of Cd and Zn were found at Iskenderun Bay (Figure 9). Very high levels of Cd, Pb, Zn and Cu have been found in Izmit Bay (Pekey, 2006). Lead was also high in Ismir Bay (Figure 9). Most of the stations, particularly Ismir, Edremit and Candarli Bays, exhibited moderate levels of Cu and Zn. Low to moderate levels were found in Israel, with high values of Hg and Zn in some stations (e.g. Haifa Bay), whereas Cd was high in the northern coast of Syria. Conversely, and with the exception of Hg, relatively low values were found in Egypt, around the mouth of the Nile River. The distribution patterns reflect anthropogenic sources originating from point and diffuse land-based sources, providing useful information on the identification of hotspots in the area although not fully comprehensive.

In Greece, monitoring of metals in sediments carried out for the Report “*State of the Hellenic Marine Environment*” (SoHelME, 2005) revealed a pollution gradient across the coastal areas, indicating different pollution fingerprints, with higher concentrations of Cu and Pb in the areas of influence of Thessaloniki and Athens. The values reported in the database (Figure 9) also exhibit high concentrations of Pb, Cu and Zn in the Saronikos, Strymonikos and Kavala Gulfs.

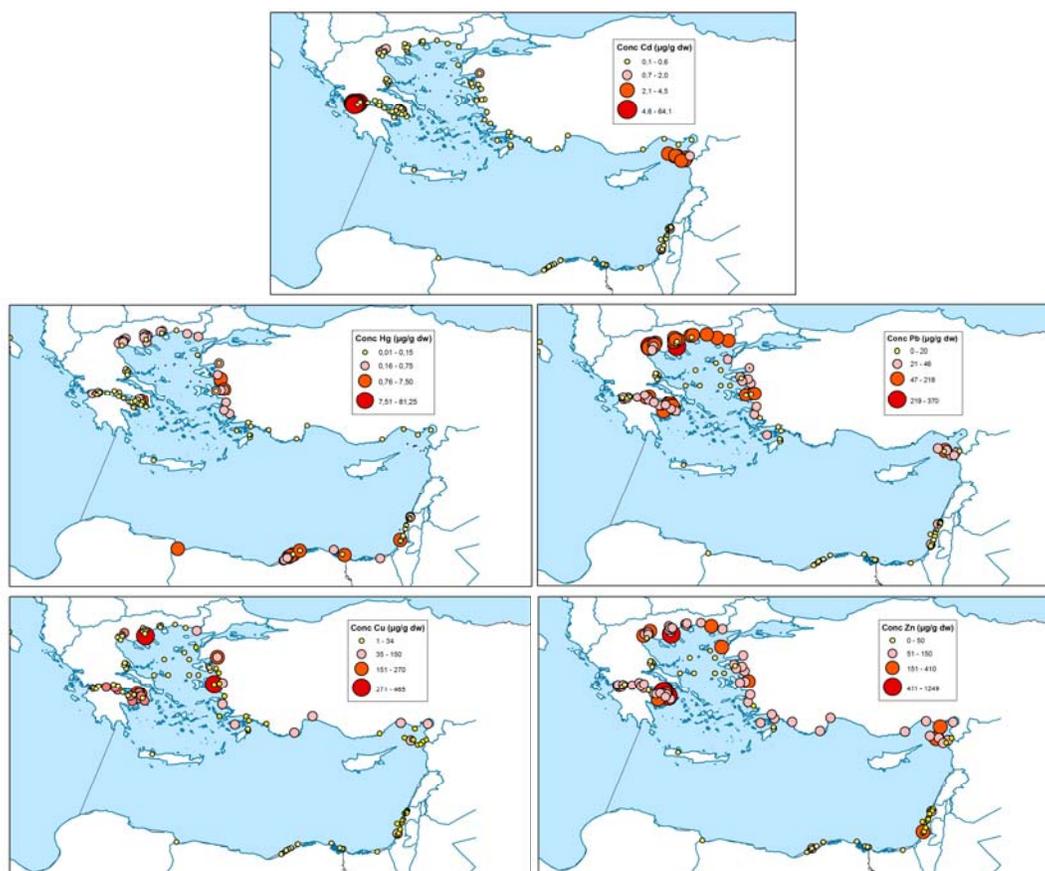


Figure 9. Mean concentrations of trace metals in sediments (µg g⁻¹ dw) from the Aegean-Levantine basin coast.

3.1.2 Trace metals in biota

3.1.2.1 General distribution

A summary of all data corresponding to the indicator species, the bivalve *Mytilus galloprovincialis* and the benthic fish *Mullus barbatus*, is shown in Table 3.5 that provides the corresponding range and average values (means and medians) for each one of the selected metals. Here, the reported values are, excluding the hotspots, of the same order of magnitude than those obtained during the MED POL I and II.

Table 3.5. Statistics of trace metal concentrations ($\mu\text{g g}^{-1}$ dw) in *Mytilus galloprovincialis* (MG) and *Mullus barbatus* (MB).

Specie	TM	n	Mean	Median	Min	Max	SD
MG	Cd	1916	0,85	0,70	0,010	10,00	0,71
	HgT	1814	0,25	0,13	0,003	8,45	0,69
	Pb	1771	2,99	1,80	0,010	79,10	5,08
	Zn	1791	142,20	126,00	0,000	5337,01	156,78
	Cu	1736	9,51	6,64	0,050	113,60	10,99
MB	Cd	487	0.80	0.07	0.000	35.49	2.98
	HgT	775	0.26	0.15	0.003	5.66	0.47
	Pb	50	0.42	0.27	0.010	1.25	0.41
	Zn	911	29.33	16.64	0.000	325.72	40.67
	Cu	910	5.05	2.11	0.546	55.68	6.97

The accumulation is higher in mussels than in fish and the levels follow a similar trend to that of sediments $\text{Cd} < \text{HgT} < \text{Pb} < \text{Cu} \ll \text{Zn}$ (Table 3.1). In fact, the concentrations of trace metals in sediments and mussels exhibit a fair correlation except in the case of Zn, which can be attributed to a major influence of the sediment lithology. Like in the case of sediments, the median values can be used as a reference for a preliminary assessment of spatial trends and identification of hot spots.

The corresponding data for the different countries are summarized in Table 3.6. The mussel *Mytilus galloprovincialis* has been more widely used than the fish *Mullus barbatus*, although in the case of Israel and Egypt, data are referred to other bivalve species (e.g. the clam *Macra corallina*). The bioaccumulation patterns of these organisms are different, so that the corresponding values cannot be included in general comparisons.

Table 3.6. Mean concentrations of trace metals in bivalves and benthic fish by country (in $\mu\text{g g}^{-1}$ dw).

Country	MG					MB				
	Cd	HgT	Pb	Zn	Cu	Cd	HgT	Pb	Zn	Cu
ALB	0.52	0.35	4.02	124.12	12.03					
CRO	0.87	0.63	2.15	137.62	10.71					
CYP						0.05	0.34	0.42	34.19	1.74
EGY	0.83*	0.15*	2.08*	46.48*	0.86*					
FRA	0.88	0.13	2.05	107.93	7.61					
GRE	0.77	1.13	1.03	150.48	8.22	2.27	0.66	0.43	55.90	2.28
ISR	1.87**	0.29**		124.9**	10.25**	0.54	0.31		23.27	2.32
ITA	0.86	0.30	3.56	159.43	12.51					
MOR	0.23	0.18	0.88	72.00	4.40					
SLO	0.85	0.12								
SPA	0.86	0.16	4.53	183.92	6.48					
SYR						0.51		0.37	170.35	9.88
TUN	0.32	0.19	0.91							
TUR	1.14	0.09	2.87	88.57	13.86	0.14	0.16		20.71	11.70

*Values corresponding to the bivalve *Donax trunculus*

**Values corresponding to the bivalve *Macra corallina*

As shown in Figure 10, the concentrations of trace metals in bivalves exhibit a large span of values for all countries but the average values are, with some exceptions, of the same order of magnitude (Table 3.6). Overall, the levels found in Morocco and Tunisia samples are below the mean, whereas they are higher for Hg in Croatia and Greece, and lower for Zn in Spain and Turkey. A number of stations with Hg and Pb values well above the mean are also found in Italy (Figure 10). High levels of Cd and Hg have also been found in Israel, but as mentioned before, these correspond to the clam *Macra corallina* that lives in the sediment. Conversely, the values were lower in *Donax trunculus* (Egypt) that could also be due to a different bioaccumulation rate of trace metals.

In the case of the fish *Mullus barbatus*, data has only been reported for the eastern Mediterranean. The concentrations of Cd and Hg were generally higher in Greece, whereas Zn was higher in Greece and Syria and Cu in Syria and Turkey.

A proper picture of the spatial distribution of the different elements in *Mytilus galloprovincialis* is given in Figure 11, where the mean concentrations of trace metals for each station is illustrated. In most cases, low or moderate levels were found, in particular for Cd, Hg and Zn, and to a lesser degree, for Pb and Cu. In this respect, Cd, Hg and Zn display a rather even distribution along the coasts of the different eco-regions, with a few hotspots in the coasts of Spain, in the areas of Cartagena (Cd) and Alboran (Zn), Italy, in the coasts of Sardinia (Cd) and Naples (Cd, Hg and Zn), and Ismir Bay (Cd). Mercury hotspots are also found in Croatia (Rijeka Bay) and Greece (Patras). A large number of stations exhibit high values of Pb, usually associated to urban areas (e.g. Barcelona, Marseille, Genova, Fiumicino and Naples) and mining spots (Portman, Spain). High levels of Cu have been found in the Ligurian (e.g. Genova) and Thyrenian Seas (e.g. Fiumicino and Naples) as well as in the coast of Sardinia.

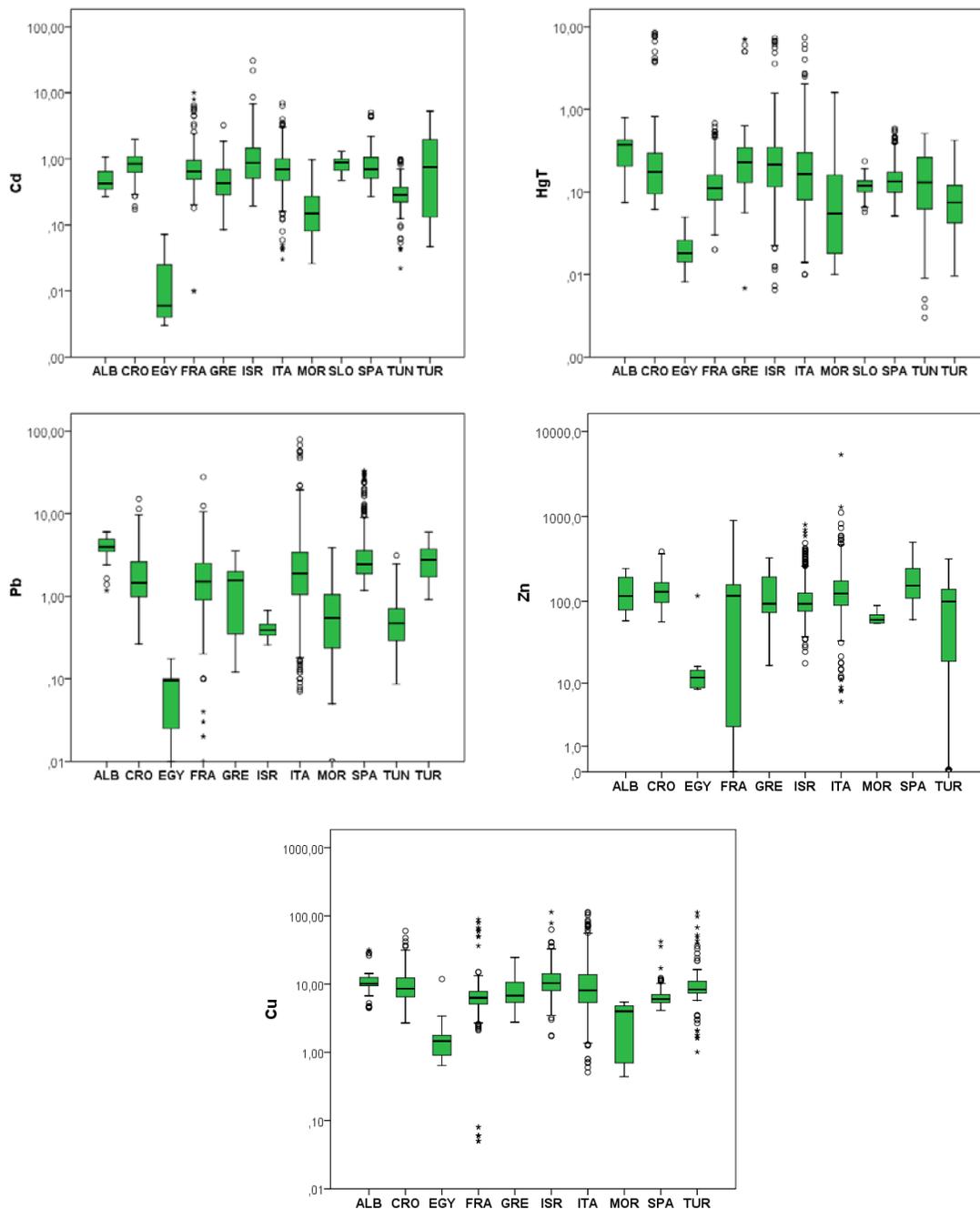


Figure 10. Concentrations of trace metals in bivalves ($\mu\text{g g}^{-1} \text{ dw}$) by country.

Figure 11. Mean concentrations of trace metals in *Mytilus galloprovincialis* ($\mu\text{g g}^{-1}$ dw).

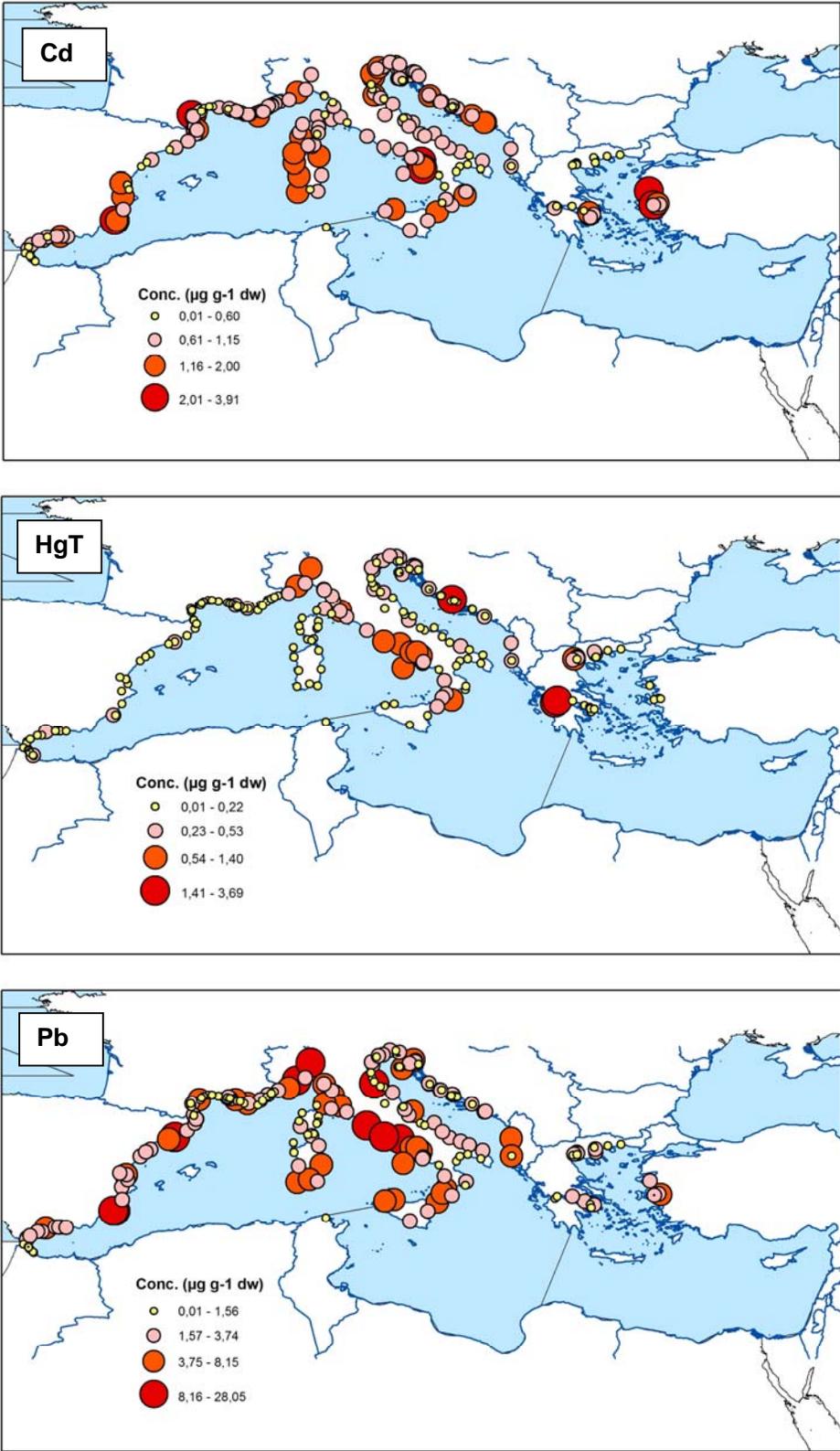
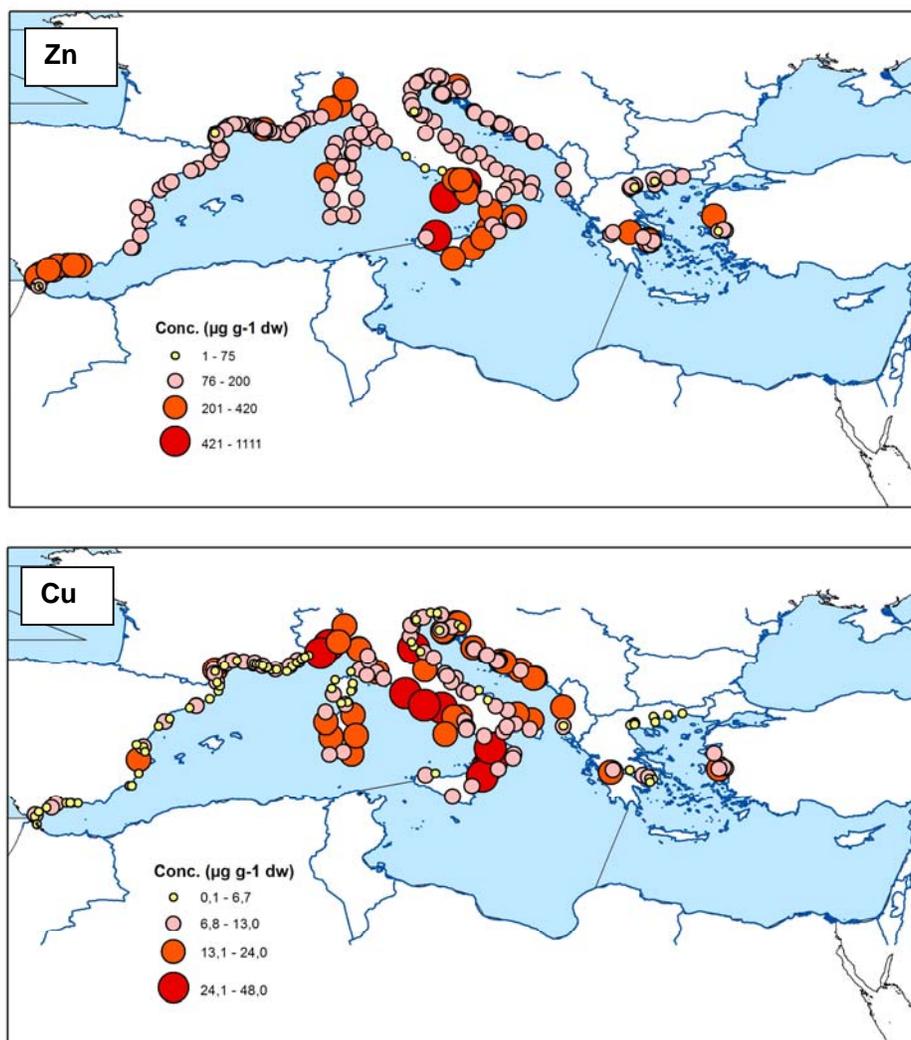
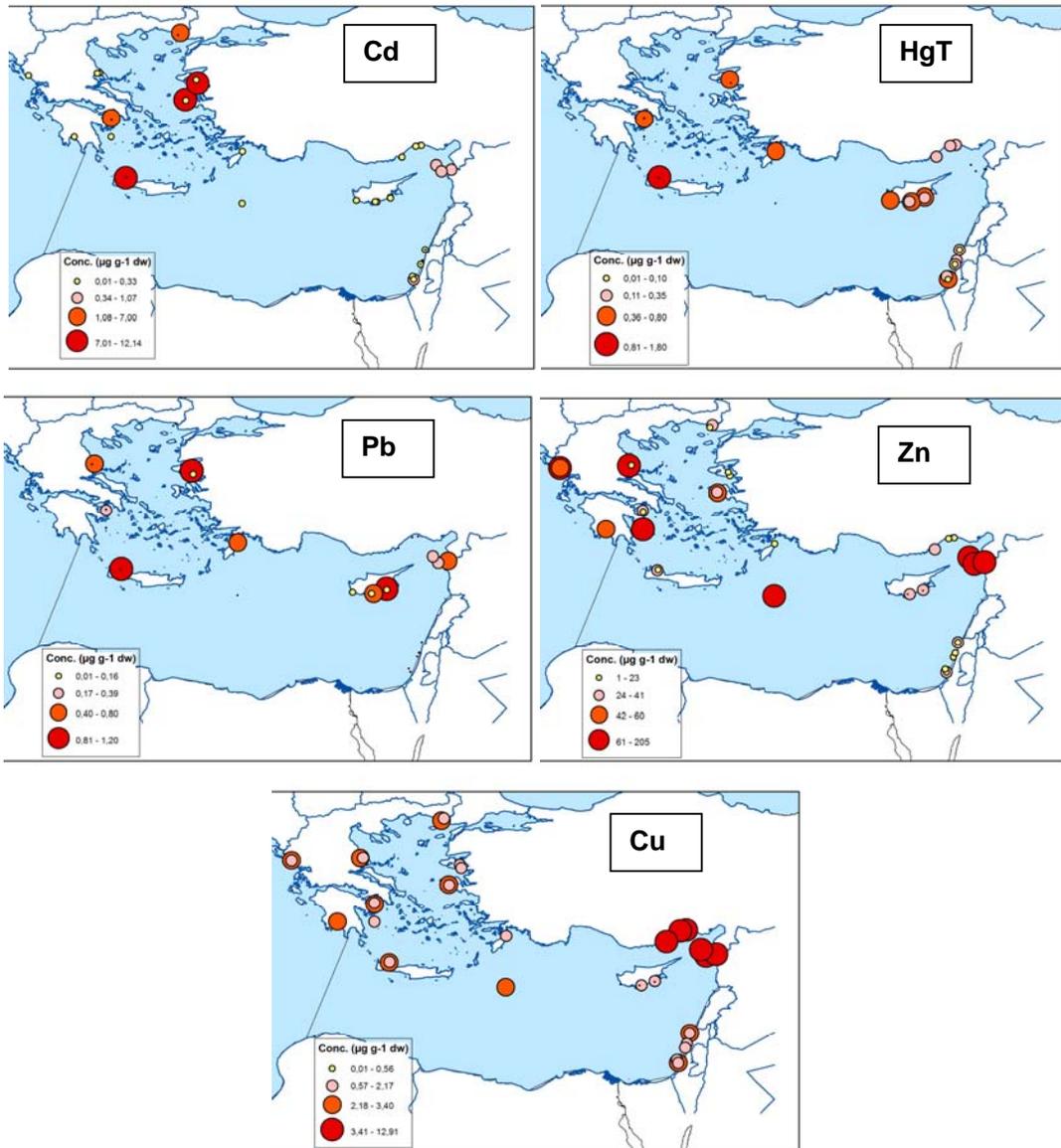


Figure 11 (cont.). Mean concentrations of trace metals in *Mytilus galloprovincialis* ($\mu\text{g g}^{-1}$ dw)



The mean concentrations of trace metals in *Mullus barbatus* for each station are shown in Figure 12. The most significant feature is the uneven distribution of Cd, with significant hotspots in the stations of Greek Islands (Crete, Mytilini and Chios). High values of Hg and Pb are also found in Crete (Chania Bay) and Cyprus (Larnaca and Limassol Bays). The levels of Zn are also high in the Avramikos, Saronikos and Thermaikos Gulfs (Greece). The area of Mersin (Turkey) exhibit high values of Cu and the mullets collected in the northern coast of Syria exhibit high concentrations of Zn and Cu. However, the limited number of stations precludes any further consideration.

Figure 12. Mean concentrations of trace metals in *Mullus barbatus* ($\mu\text{g g}^{-1}$ dw).



3.1.2.2 Distribution of trace metals in biota within eco-regions

A summary of the data corresponding to trace metals in mussels (*Mytilus galloprovincialis*) from the four Mediterranean eco-regions is shown in Table 3.7 and Figure 13. Data for fish (*Mullus barbatus*) was only available for the Aegean-Levantine basin, so no comparison was possible with the other regions.

Table 3.7. Median and concentration ranges of trace metals in *Mytilus galloprovincialis* ($\mu\text{g g}^{-1}$ dw)

Eco-region	Cd	HgT	Pb	Zn	Cu
WESTERN MEDITERRANEAN	0.66 (0.01-10.0)	0.13 (0.01-7.4)	2.00 (0.01-79.1)	130 (0.01-5337)	6.30 (0.05-114)
ADRIATIC	0.80 (0.03-2.73)	0.14 (0.01-8.45)	1.53 (0.07-67.5)	122 (5.7-467)	8.01 (0.51-81.6)
CENTRAL MEDITERRANEAN	0.43 (0.09-3.40)	0.16 (0.00-7.00)	0.81 (0.07-5.36)	87 (11.6-565)	9.32 (1.36-70.5)
AEGEAN-LEVANTINE	0.75 (0.05-5.27)	0.08 (0.01-0.63)	2.28 (0.84-5.97)	131 (0.00-325)	7.84 (1.01-112)

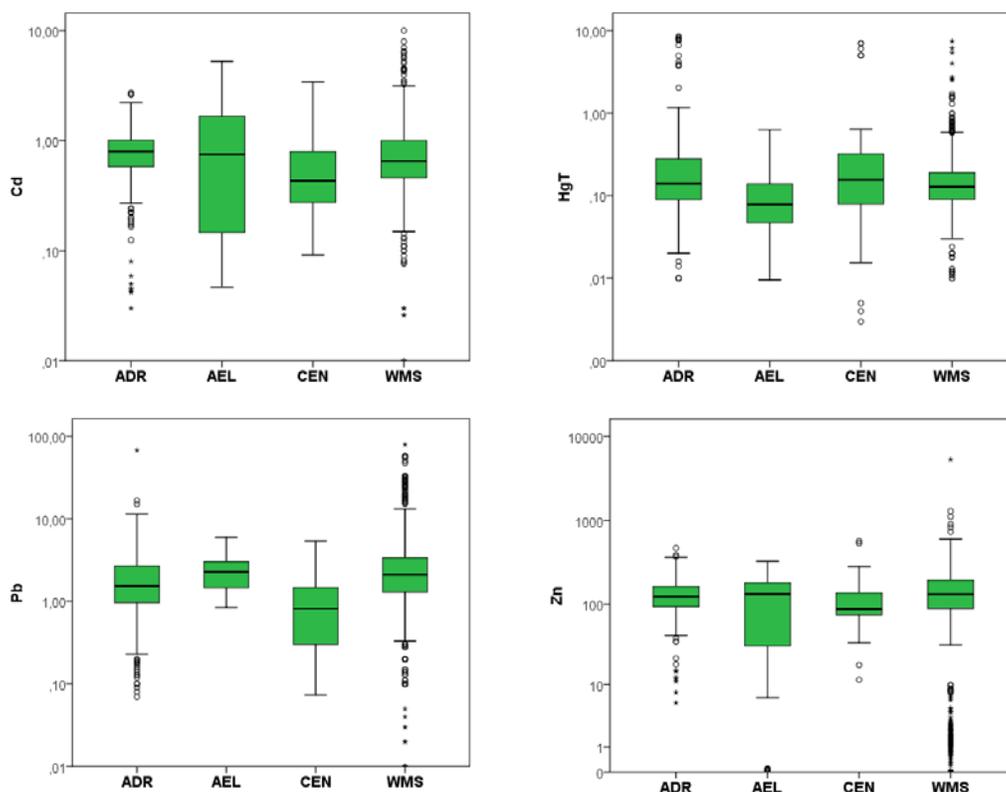


Figure 13. Concentrations of trace metals ($\mu\text{g g}^{-1}$ dw) in *Mytilus galloprovincialis* from the different Mediterranean sub-regions.

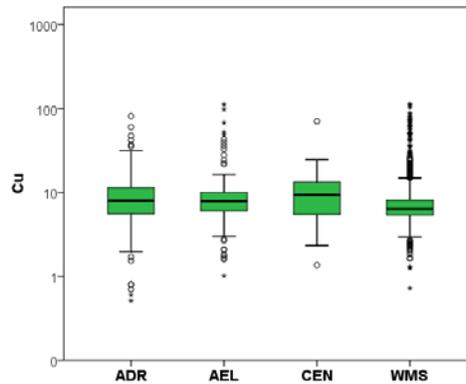


Figure 13 (cont.). Concentrations of trace metals ($\mu\text{g g}^{-1}$ dw) in *Mytilus galloprovincialis* from the different Mediterranean sub-regions.

As it was observed for sediments, concentrations of trace metals in mussels are higher for the Western Mediterranean and the Adriatic Sea, although the data set is lacking information from several eastern and southern countries.

In order to look for the consistency of the values in the Database and also enlarge the spatial coverage, the concentrations of trace metals in *Mytilus galloprovincialis* and *Mullus barbatus* in other recent studies published in the open literature were assembled and reported in Table 3.8.

Studies have been basically conducted on coastal enclosures like the Portman Bay (Spain), Venice Lagoon (Italy), Vlora Bay (Albania), Izmir Bay (Turkey), etc., and the Gulfs of Taranto (Italy) and Saronikos (Greece) among others. A few studies have also been focused on open areas of the Western Mediterranean, Adriatic and Aegean Sea.

As it can be seen, the levels are in general within similar ranges as those found in the Database, with high values of Cd, Pb and Cu in the Venice Lagoon and Portman Bay, and of Hg, Pb, Zn and Cu in the Eastern Adriatic.

Table 3.8. Concentrations of trace metals in MG and MB in other studies.

Species	Location/Area	Year(s)	Stations / Samples	Concentration ($\mu\text{g/g dw}$) ^a					Reference
				Cd	Hg	Pb	Zn	Cu	
MG	Portman Bay (ES)	1991 - 2005		1.21 – 3.00 ^a	0.09 – 0.58 ^a	37 – 421 ^a	147 – 300 ^a	4.2 - 16 ^a	Benedicto et al. (2008)
	Balearic Islands	2005	15 /	2.82 (1.56 - 3.37)	0,20 (0.16-0,26)	2.43 (1.82-3.34)	233 (223-251)	4.76 (4.30 – 5.15)	Deudero et al. (2009)
	Iberian coast	2003-2004	16 / 96	0.28 – 1.67	0.08 – 0.78	1.82 – 57.83	91 - 431	5.98 – 11.31	Fernández et al. (2010)
	Western Med.	2001-2006	123	1.33 (0.46 – 2.89)	0.10 (0.04 – 0.33)	1.4 (0.5 – 8.3)	-	-	Benedicto et al. (2011)
	Tyrrhenian Sea	2000	6 / 87	0.38 (0.33 - 0.49)	-	0.46 – 1.31	152 (123 – 180)	8.73 (5.51 – 11.5)	Conti and Cecchetti (2003)
	Venice Lagoon	1993	6 / 60	0.05 – 4.54	0.11 – 0.27	6.18 – 80.26	82 – 185	5.08 – 20.89	Widdows et al. (1997)
	Central Adriatic	2001- 2004	1 / 39	0.409 – 1.60	0.022 – 0.186	0.292 – 2.95	23.2 – 153	1.08 – 8.12	Fattorini et al. (2008)
	Western Adriatic	2005	6 / -	0.448 – 2.41	0.014 – 0.064	0.503 – 1.28	57.6 - 158	1.77 – 4.01	Fattorini et al. (2008)
	Mali Ston Bay (HR)	1998-2005	4 / 39 - 85	1.15 (0.39 – 2.4)	0.15 (0.08 – 0.28)	1.09 (0.24 – 3.69)	139 (49 – 418)	5.61 (1.98 – 10.98)	Kljakovic-Gaspic et al. (2007)
	Eastern Adriatic (HR)	2006	14 / -	0.99 (0.48 – 1.72)	1.05 (0.12 – 10.3)	5.45 (0.99 – 14.79)	181 (93 – 563)	40.19 (5.92 – 369.3)	Kljatovic-Gaspic et al. (2010)
	Eastern Adriatic (HR)	2001-2005	4 / 20	0.189 – 0.526 ^a	-	-	7.00 – 59.52 ^a	3.79 – 15.58 ^a	Dragun et al. (2010)
	Vlora Bay (Albania)	2006	2 / 100	0.22 – 0.85 ^b	0.17 - 0.26 ^b	0.6 – 1.7 ^b	-	-	Corsi et al. (2010)
	Gulf of Taranto (IT)	1999-2000	2 / 50	0.23 – 0.95 ^b	-	1.19 – 4.29 ^b	68 - 109 ^b	6.35 – 15.72 ^b	Cardellicchio et al. 2008
	Hellenic waters	1993 - 2002	-/1200	0.02 - 8.57	0.04 – 1.36	0.03 – 22.0	-	0.01 - 24.9	SoHelME, 2005
	Saronikos Gulf	2004-2006	3 / 360	0.20 – 0.40	-	0.53 – 7.89	-	-	Vlahogianni et al. (2007)
Izmir Bay (TK)	2004	2 / 20-40	0.028 (0.025 – 0.03) 0.011 (0.009 – 0.013)	0.018 (0.017-0.02) 0.017 (0.017- 0.02)	0.4 (0.38-0.42) 0.08 (0.07-0.08)	27.7 (26.9-28.5) 17.9 (17.8-17.9)	5.28 (5.26 – 5.30) 3.67 (3.561 – 3.78)	Kucuksezgin et al. (2008)	
Aegean Sea (TK)	2002-2003	6 / 720	0.21 – 2.74 ^a	-	2.58 – 9.05 ^a	84.79 – 195.50 ^a	5.00 – 9.74 ^a	Sunlu (2006)	

MB	Portman Bay (ES)	2004		5.0 – 5.41 ^a	0.33 – 0.50 ^a	0.25 – 0.33 ^a	15.83 – 17.50 ^a	1.63 – 1.79	Benedicto et al. (2008)
	Adriatic Sea	2006		0.08 (0.04-0.17)	2.92 (0.21 – 11.5)	0.25 (0.17 – 0.75)	-	-	Storelli et al. (2008)
	Vlora Bay (Albania)	2006	2 / 40	< 0.02 – 0.08 ^b	0.09 – 1.06 ^b	< 0.1 – 0.3 ^b	-	-	Corsi et al. (2010)
	Hellenic waters	1993 - 2002	-/1200	-	0.03 – 0.05	0.05 – 3.75	0.01 - 72.5	0.01 - 5.14	SoHelME, 2005
	North-East Med (TK)	2003	10 / 4 seasons	1.9 – 3.1	-	5.8 – 9.4	26.7 – 34.5	10.9 – 17.5	Çogun et al. (2006)
	Aegean Sea (TK)	1994-1998	12 / -	0.01 (0.002-0.02) ^a	0.38 (0.07-0.83) ^a	0.53 (0.17-0.86) ^a	-	-	Kucuksezgin et al. (2001)
	Izmir Bay (TK)	1997 - 2002		0.0004 – 0.042 ^a	0.058 – 2.167 ^a	0.011 – 1.992 ^a	-	-	Kucuksezgin et al. (2006)
	Izmir Bay (TK)	2005	186	-	0.018 – 0.658 ^a	-	-	-	Gonul & Kucuksezgin (2007)
	Eastern Med (TK)	1996	2 / 15	-	0.71 ^a	6.08 ^a	< DL	< DL	Dogan-Saglamtimur & Kumbur (2010)
	Eastern Med (TK)	2005	1 / 10	0.49	-	0.56	5.87	-	Turan et al. (2009)

^a Values converted into dw assuming 19% and 24% DW for MG and MB, respectively

^b Mean values.

In the **Western Mediterranean** some stations are chronically polluted with trace metals, according to the levels found in mussels, like those at the Ligurian Sea, close to Genova, and Naples (Figure 14). In addition to these, certain sites can be considered as hot spots for Cd and Pb (e.g. Sardinia coast, Palermo and Cartagena-Portman) (Benedicto *et al.*, 2008; Magni *et al.*, 2006; Tranchina *et al.*, 2008; Lafabrie *et al.*, 2007). Significant concentrations of Pb and Cu were reported in Piombino (Ligurian Sea) (Bocchetti *et al.*, 2008). A number of stations, usually associated to urban areas or to river inputs, also exhibit high values of Pb (e.g. Barcelona, Malaga, Marseille, Genova, Fiumicino and Naples).

High levels of Cu have been found in the Ligurian (e.g. Genova) and Tyrrhenian Seas (e.g. Fiumicino and Naples) as well as in the coasts of Calabria and Sardinia. Higher levels of Hg were found in coastal waters of the Tyrrhenian Sea (e.g. Fiumicino, Naples and Messina). All these stations are close to mining and/or urban areas. Finally, the levels of Zn are also high in the Alboran coast of Spain and the southern coast of Italy (Calabria and Sicilia).

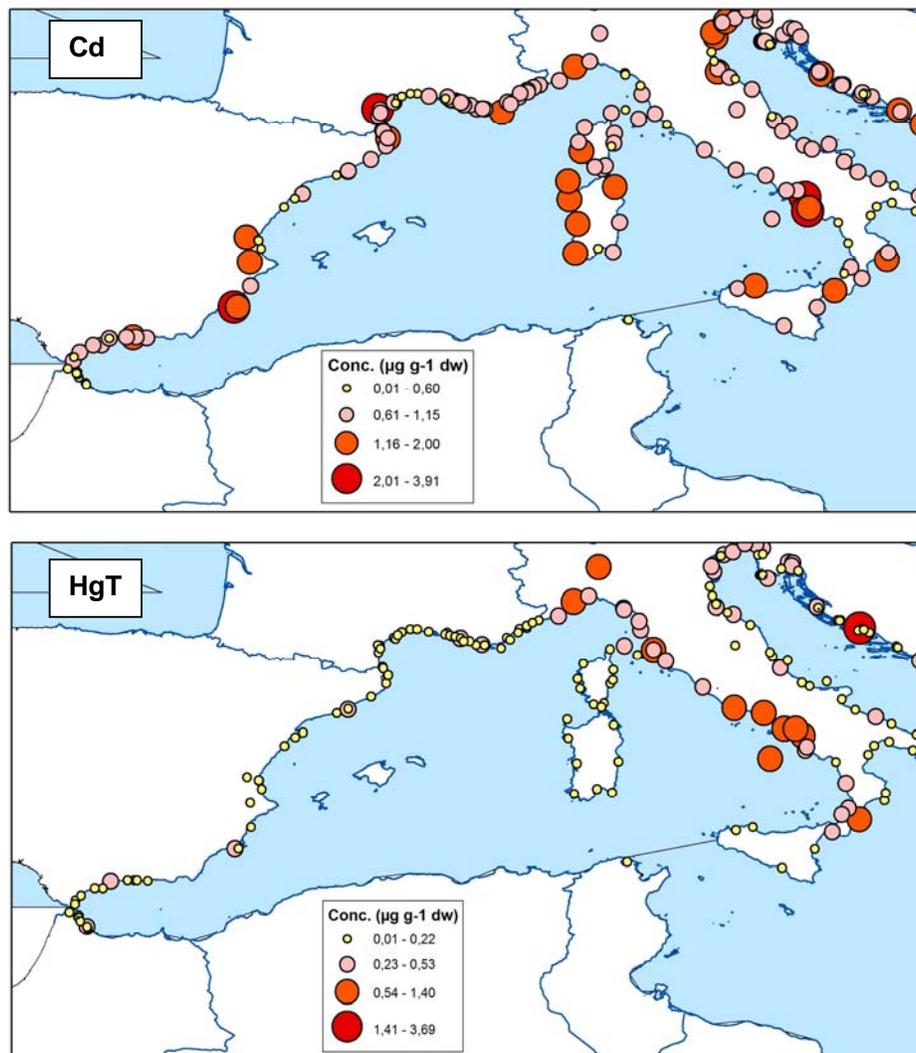


Figure 14. Mean concentrations of trace metals ($\mu\text{g g}^{-1}$ dw) in *Mytilus galloprovincialis* from the Western Mediterranean.

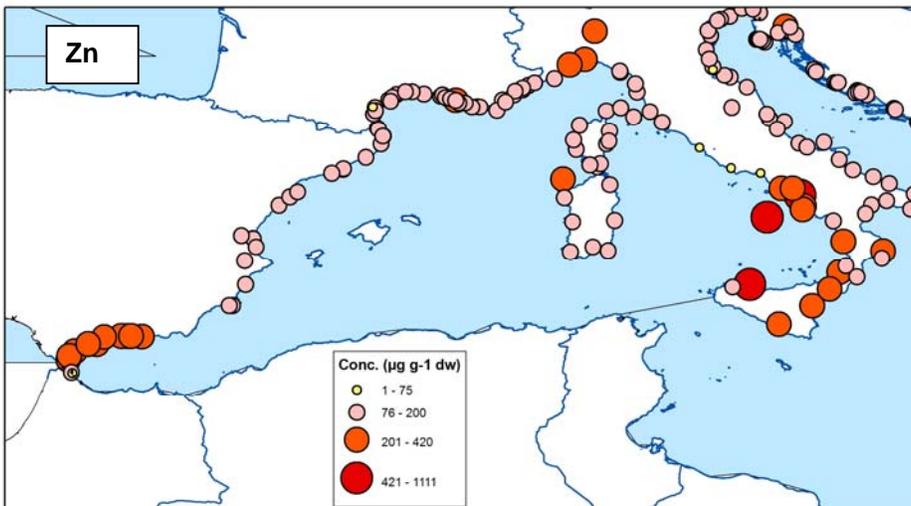
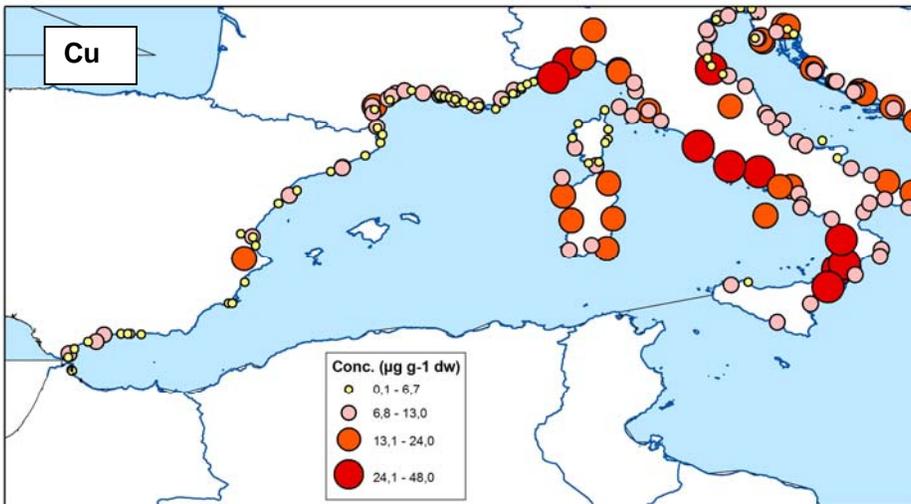
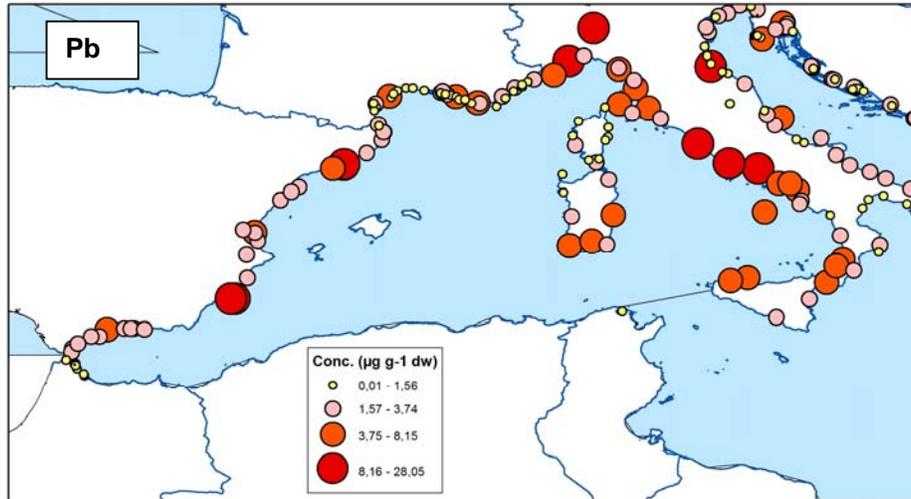


Figure 14 (cont.). Mean concentrations of trace metals ($\mu\text{g g}^{-1}$ dw) in *Mytilus galloprovincialis* from the Western Mediterranean.

An interesting study was carried out recently by Benedicto *et al.* (2011) using transplanted mussels (*Mytilus galloprovincialis*) in 123 coastal stations of the Western Mediterranean. The distributions of Pb, Cd and Hg in the different sub-basins are shown in Figure 15 that, in general, confirm the previous findings.

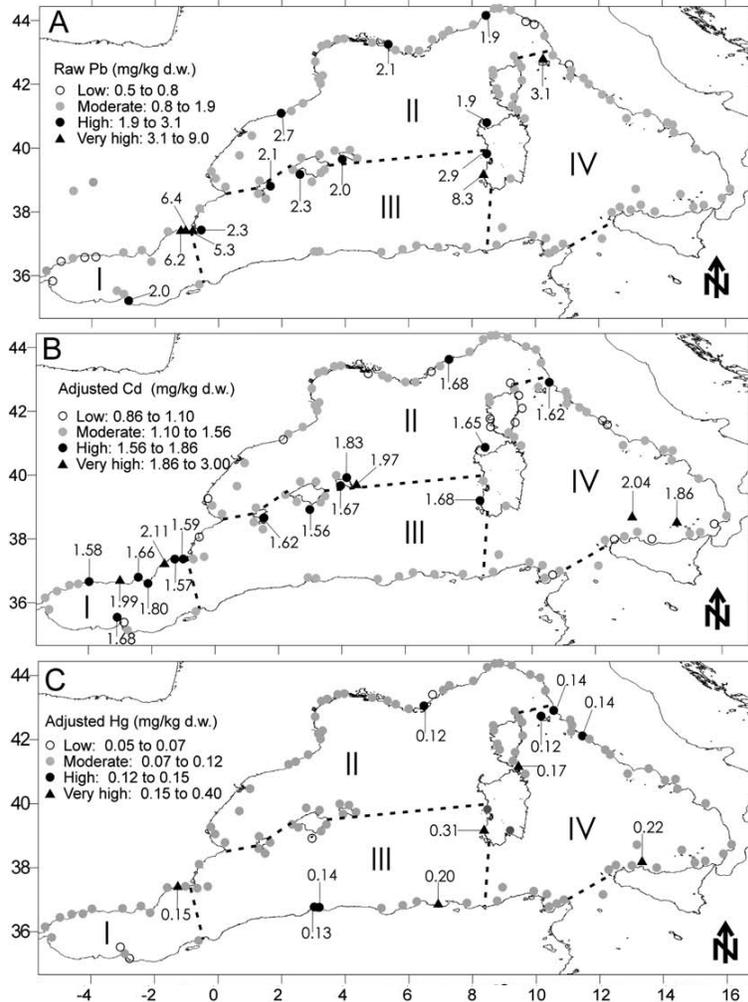


Figure 15. Spatial distribution of the adjusted concentrations of Pb (A), Cd (B) and Hg (C) in caged *Mytilus galloprovincialis*, in the Western Mediterranean. Upper left legend shows the concentration ranges for each metal (Benedicto *et al.*, 2011).

The Pb concentrations recorded in the Western Mediterranean basin were generally categorized as moderate (Figure 15A). Higher levels have been reported for polluted areas of singular sites of the NW Italy (Schintu *et al.*, 2008; Frenzilli *et al.*, 2004), in Portoscuso (Sardinia) and in three sites located along the SE coast of Spain (Cabo del Agua, El Portus and Portman). The coast of the Balearic Islands (e.g. Palma de Mallorca), the south coast of France (Huveaune), the northern coast of Italy (Savona), the coast of Sardinia (Oristano and Porto Torres) and the coast of Morocco (Nador) exhibited moderate levels, mainly attributed to industrial sources.

Adjusted Cd concentrations (Figure 15B) were categorized as low to moderate levels. Many of the “high levels” of Cd were recorded at sites along the south and south-

eastern coast of Spain (Cabo de Gata, Almeria and El Portus), an intensely mineralised region; the coasts of Balearic Islands and Morocco, and at few sites along the coast of Italy (Piombino, and western Sardinia) and France (Nice).

Mercury concentrations are shown in Figure 15C. Very high Hg concentrations (ranging from 0.15 to 0.31 $\mu\text{g g}^{-1}$ dw; adjusted data) were measured at five scattered sites along the Western Mediterranean basin (Portoscuso, Palermo and Maddalena-Sardinia Island, in Italy; Skikda in Algeria, and El Portus in Spain). There was also a predominance of high Hg values at Algiers Bay, along the north-west Italian coast (Piombino, Portoferraio) and in the French coast at Hyeres ouest. The assessment of potential sources for the environmental occurrence of Hg point out to the anthropogenic activities.

The study allowed also identifying the contributions of direct river/sewage inputs. Along the coast of France, mussels moored in the mouth of the Huveaune River, currently considered to represent a significant source of trace metals into the Marseille Gulf, showed high Pb levels. Likewise, high Pb concentrations measured in the mouth of the Llobregat River (NE Spain) may be explained by the influence of the Barcelona metropolis. The high level of Hg at Hyeres Bay (France), near the mouth of the small river Gapeau, could be also related to sewage inputs.

Trace metals are extensively monitored throughout the **Adriatic Sea** using mussels as bioindicators, mostly confined in urban and industrial areas. Concentrations are from low to moderate in Northern, NE and NW part of the basin. High concentrations of Pb and Cu were found in the Venice Lagoon (Widdows *et al.*, 1997) and in the areas affected by the Po River discharges, consistent with urban/industrial discharges. The Central Eastern and Western Adriatic has also been extensively monitored. Concentrations of Pb and Cu are relatively high in one area of the Dalmatian coast. High levels of Cd, Hg and Cu are found in the Kastela and Rijeka Bays, due to the discharge of untreated urban and industrial wastewaters. A monitoring survey carried out during 2001-2005 in the Mali Ston Bay (Croatia), determined that Pb and Hg were significantly elevated in the urban and industrial areas, while Cd was more uniformly distributed across the monitored sites, being also high in mussels from rural areas located far away from anthropogenic sources of pollution (Kljakovic-Gaspic *et al.*, 2007). In Albania, Pb and Cu contamination above the mean levels were found in mussels collected in Vlora Bay and close to Durres, a known dumping site of industrial residues.

The **Central Mediterranean** is not well represented in the Database. A few samples from south Italy and Greece are available. High levels of Hg were found in the Gulf of Patras (Greece) and moderately high levels of Cu were also found at the Sicily coast.

Trace metal analysis in biota in the **Eastern Mediterranean** exhibited low values in the case of *Mytilus galloprovincialis*, with the exception of Cd, Pb and Cu in Turkey (Akçay and Ismir Bays) and Cd and Zn in Greece (Piraeus). In Israel, *Mactra corallina* was used as indicator organism which exhibited relatively high values of Cd and Cu in the northern coast (Haifa). Information is missing for the southern countries.

Analysis of *Mullus barbatus* showed uniform metal bioaccumulation through the area but certain stations from Greek Islands (e.g. Crete) exhibited high levels of Cd, Hg, Pb and Cu. Copper and Zn were also high in Syria, and Pb and Hg in Cyprus.

An extensive monitoring of metals in mussels and red mullets, collected during 1990-2002 from several Hellenic coastal areas, revealed low levels (SoHeIME, 2005). Exceptions concern areas subject to heavy anthropogenic activities, such as the Saronikos Gulf (Zn, Pb), the Thermaikos Gulf (Hg) and the Pagasitikos Gulf (Zn).

3.1.3 Chlorinated pesticides in biota

3.1.3.1 General distribution

Chlorinated pesticides have been extensively analyzed in Mediterranean biota since the inception of MED POL (UNEP, 1990). Mussels and mullets have been the most widely studied organisms in the whole basin as part of many case studies published in the literature and recently assessed on the occasion of the implementation of the Stockholm Convention (UNEP, 2002). However, it has been only since the last decade that they have been continually monitored, and data gathered in the MED POL Database.

A summary of the mean concentrations of organochlorinated pesticides (OCPs) in *Mytilus galloprovincialis* is shown in Table 3.9. Aldrin, dieldrin, endrin, hexachlorobenzene (HCB) and lindane have been monitored in few countries. Concentrations were in the low ng g⁻¹ range with the exception of aldrin, dieldrin and HCB in some stations from Turkey, and of lindane in Albania.

Concentrations of DDTs were one order of magnitude higher (Tables 3.9 and 3.10), with p,p'-DDE being, in general, the predominant component, indicating old sources of DDT. However, recent inputs in some areas cannot be ruled out.

Concentrations up to 9779 ng g⁻¹ dw of total DDTs were found in mussels from the Albania coast, probably as a result of the existence of large stockpiles of obsolete pesticides in the country, including DDT as well as lindane, as it has been reported elsewhere (UNEP, 2002).

Table 3.9. Mean concentrations of OCPs in *Mytilus galloprovincialis* (ng g⁻¹ dw) by country.

Country	ALD	DIE	END	HCB	LIN	DDD	DDE	DDT	DDTs
ALB	-	-	-	-	35.28	268.78	762.84	546.52	1581.5
CRO	0.93	1.10	-	0.26	0.69	3.10	5.65	3.94	11.89
FRA	1.70	1.73	-	-	0.99	7.48	10.09	2.44	17.21
ITA	0.68	0.74	-	0.61	0.94	6.99	12.40	8.49	23.22
SPA	1.10	0.93	1.56	0.50	0.72	5.90	11.83	8.03	22.10
TUR	7.05	11.64	-	2.93	-	29.99	29.31	2.02	20.25

Table 3.10. Statistics of DDTs in *Mytilus galloprovincialis* (ng g⁻¹ dw) by country.

Country	n	Mean	Median	Min	Max	SD
ALB	40	1581.49	1298.95	28.50	9778.69	1908.72
CRO	108	11.89	9.00	2.01	42.50	7.61
FRA	243	17.21	10.39	0.60	157.20	19.92
ITA	374	23.22	13.76	0.01	345.00	37.86
SPA	276	22.10	6.32	0.24	188.66	34.94
TUR	46	20.25	9.85	1.01	173.11	31.44

The concentration ranges are shown in Figure 16. In general, the average values are of the same order of magnitude, but large span of values are found in Turkey for aldrin and dieldrin, and for lindane in Albania, with largely skewed distributions, indicating the occurrence of significant hot spots. The DDT values in Albania are also several orders of magnitude higher.

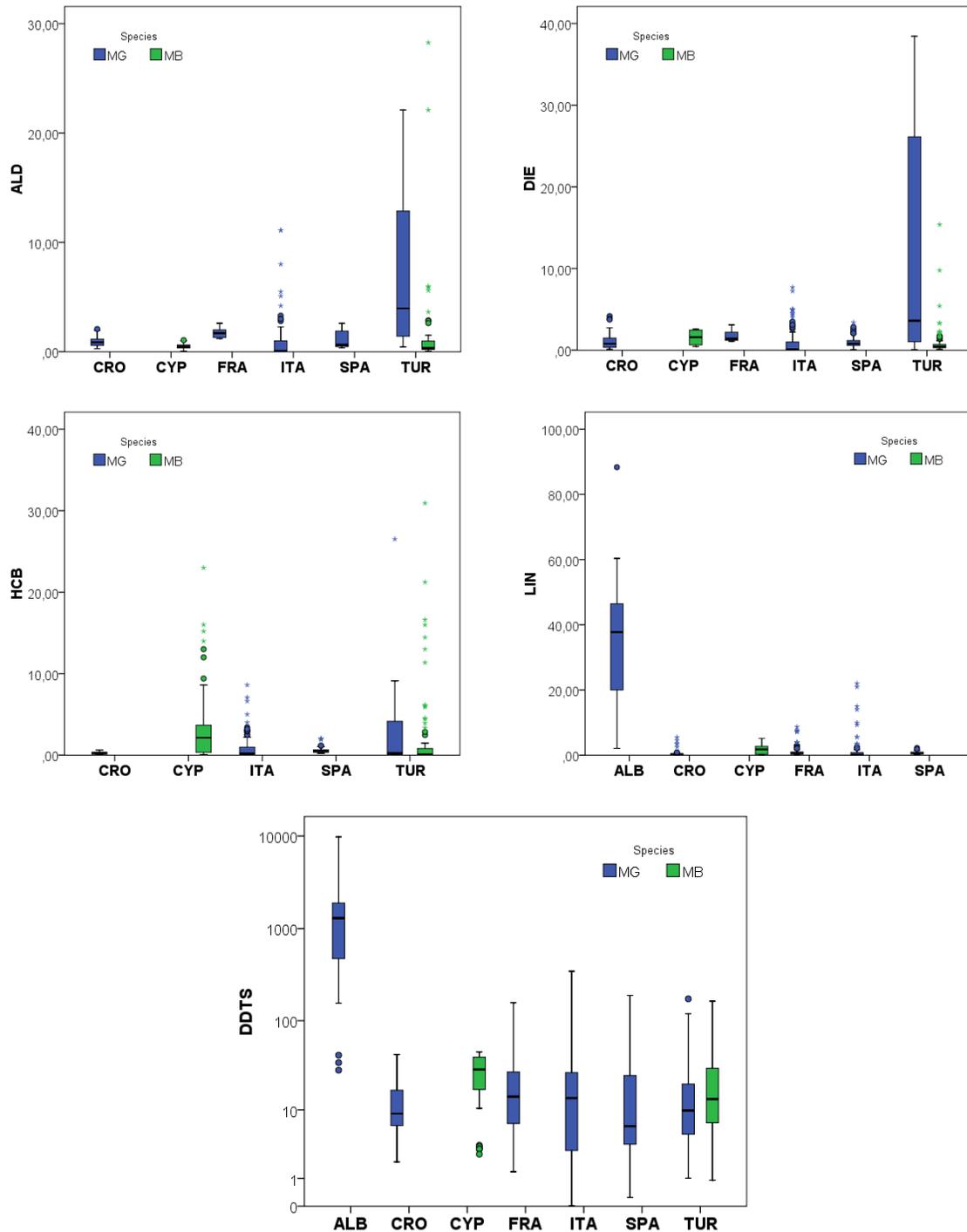


Figure 16. Concentrations of OCPs and DDTs in MG and MB, by country.

The mean concentrations of OCPs in mussels for each station are illustrated in Figures 17 and 18. Moderate concentrations of aldrin were recorded in the NW Mediterranean and the western Adriatic coast whereas mussels from the Ligurian coast exhibited high concentrations of HCB. Lindane was also moderately present in the Northern Adriatic (Trieste Gulf), and DDTs in a few stations of Spain (Ebro River and Barcelona), France (coastal lagoons of Leucate and Thau), Italy (Liguria and Marche regions) and Turkey (Ismir Bay). Much higher levels were recorded in mussels collected in the Albania stations (Durrës and Vlora Bay), where the existence of large stocks of obsolete pesticides have been reported (UNEP, 2002). The levels of aldrin, dieldrin and HCB were also high in mussels from the Ismir Bay (Turkey).

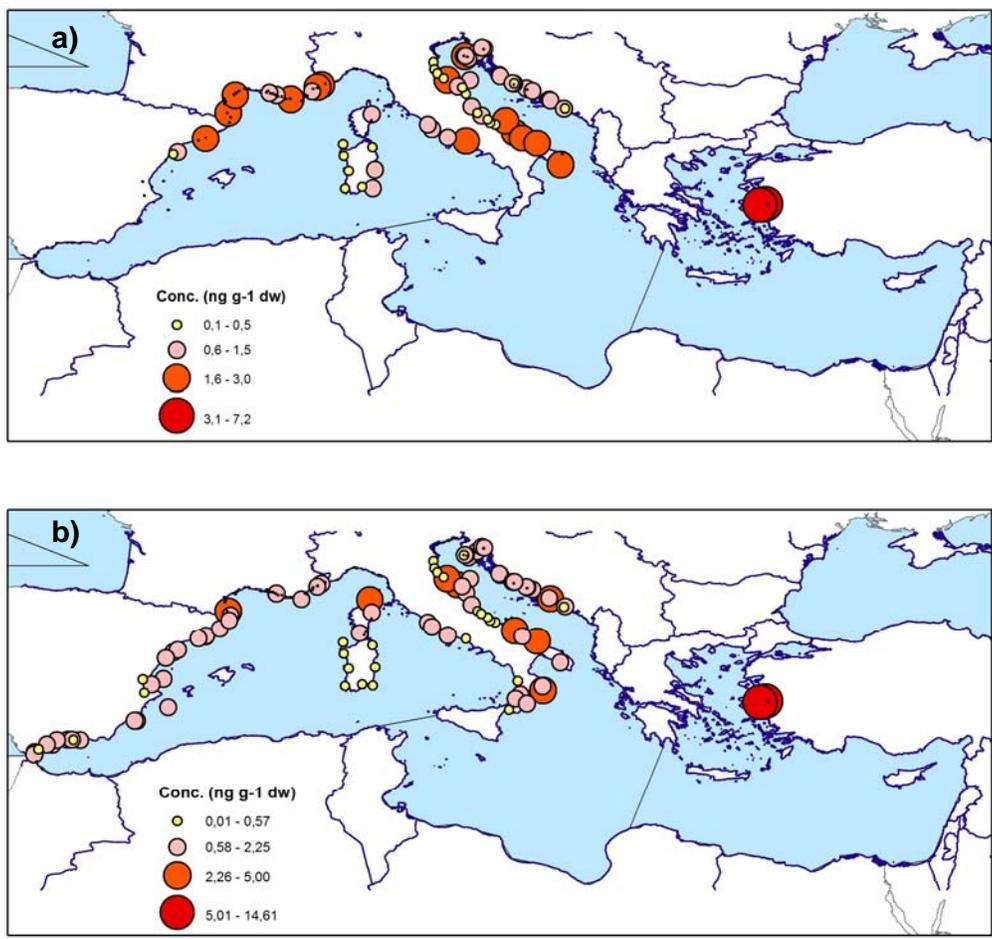


Figure 17. Mean concentrations ($\text{ng g}^{-1} \text{ dw}$) of (a) aldrin and (b) dieldrin in *Mytilus galloprovincialis*.

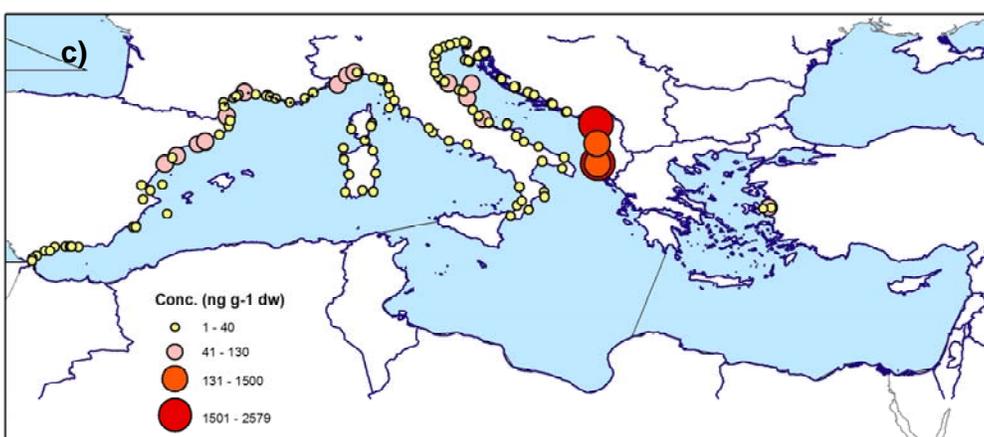
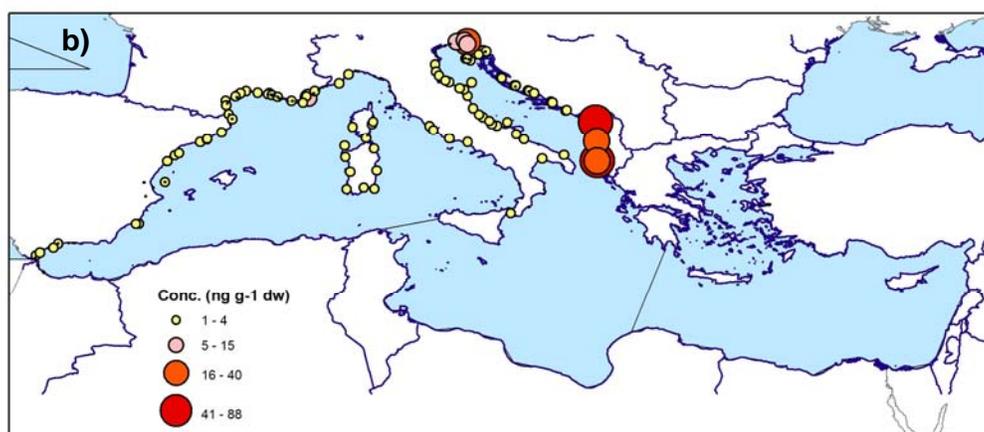
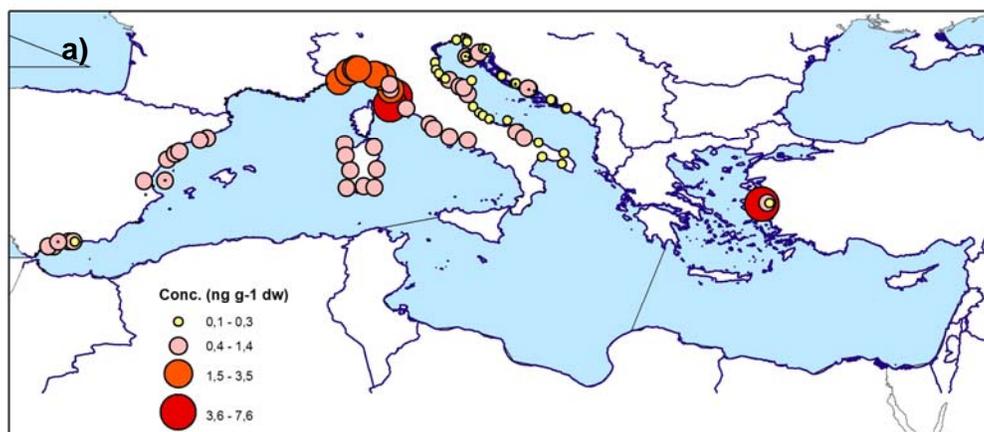


Figure 18. Mean concentrations (ng g⁻¹ dw) of (a) HCB, (b) lindane and (c) DDTs in *Mytilus galloprovincialis*.

Mullus barbatus was also the indicator species used in the Aegean-Levantine Sea (Tables 3.11 and 3.12). A large survey of the red mullet (*Mullus barbatus*) carried out between 1986 and 1991 in 8 coastal stations of the Aegean Sea revealed concentrations of α and γ -HCH of 0.1-0.5 ng g⁻¹ ww and 0.6-3.5 ng g⁻¹ ww, respectively. Lindane was also found in red mullet collected between 1993-99 in the Cyprus coast (0.6-1.3 ng g⁻¹ dw) (UNEP, 2002).

Another survey conducted from 1986 to 1995 in both the Greek and Turkish coasts determined a relatively low level of pollution in this area (UNEP, 2002), although moderate levels of DDTs and HCB were found in some stations of Turkey and Cyprus (Figure 19).

Table 3.11. Mean concentrations of OCPs in *Mullus barbatus* (ng g⁻¹ dw) by country.

Country	ALD	DIE	END	HCB	LIN	DDD	DDE	DDT	DDTs
CYP	0.45	1.55	5.12	3.77	1.77	3.81	15.00	13.67	27.29
TUR	1.90	1.04	-	2.11	-	2.39	32.16	2.54	31.34

Table 3.12. Statistics of DDTs in *Mullus barbatus* (ng g⁻¹ dw) by country.

Country	n	Mean	Median	Min	Max	SD
CYP	46	27.29	29.00	2.65	45.52	13.24
TUR	106	31.34	13.40	0.91	163.50	40.07

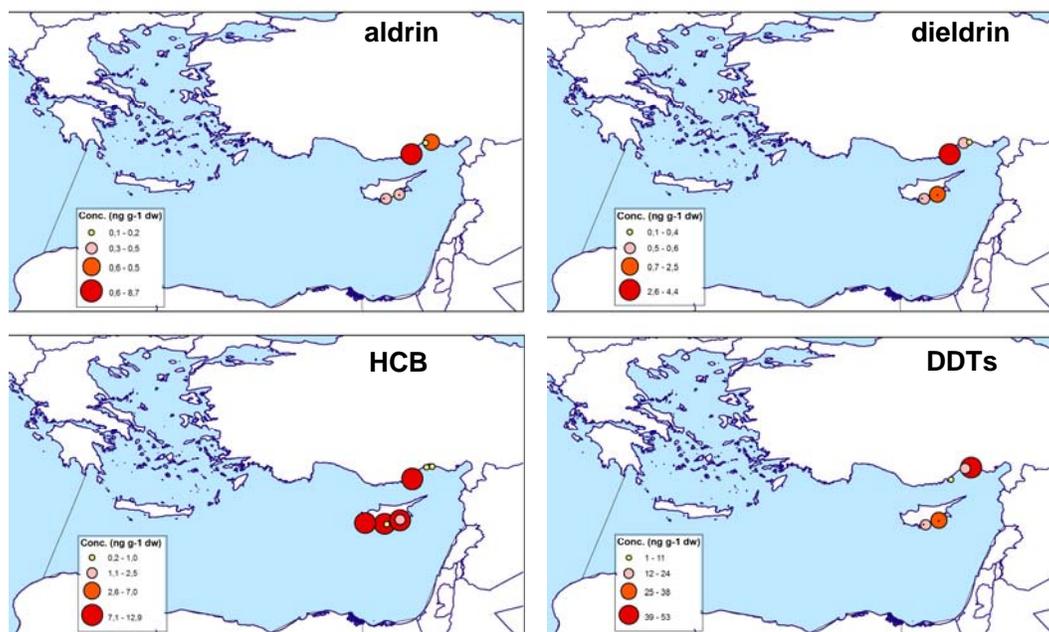


Figure 19. Mean concentrations of organochlorinated pesticides (ng g⁻¹ dw) in *Mullus barbatus*.

3.1.3.2 Distribution of chlorinated pesticides in biota within eco-regions

A summary of data corresponding to aldrin, dieldrin, HCB, lindane and DDTs in mussels (*Mytilus galloprovincialis*) from the four Mediterranean eco-regions is shown in Table 3.13 and Figure 20. Concentrations are generally low, higher in the Aegean-Levantine for aldrin and dieldrin, and in the Western Mediterranean for HCB and lindane. However, the highest values of HCB and lindane are found in Turkey and Albania, with a number of important hot spots, as shown above (Figures 17 and 18). In the case of DDTs, their median values are similar for the four eco-regions, around 10 ng g⁻¹ dw, but again extremely high values are found in the Adriatic (Figure 20). However, data are lacking for most of the southern countries. Data on fish (*Mullus barbatus*) was only provided for the Aegean-Levantine basin, so no comparison is possible with the other regions.

An additional number of individual studies reported in the literature on the occurrence of DDTs in *Mytilus* and *Mullus* sp. are referenced in Table 3.14 for comparison purposes. As it can be seen, the levels are in general within similar ranges although the samples collected during the 90s exhibit much higher values than the more recent ones. This may clearly reflect a decreasing trend resulting from the ban of this pesticide in the region, as will be described in the following section. Consequently, the assessment of the spatial distribution of DDTs in the Mediterranean should be based on recently published data, around the last 5 years.

Table 3.13. Median and concentration ranges of chlorinated compounds in *Mytilus galloprovincialis* (ng g⁻¹ dw)

Eco-region	Aldrin	Dieldrin	HCB	Lindane	Σ DDTs
WESTERN MEDITERRANEAN	0.34 (0.01-11.10)	0.76 (0.01-4.54)	0.54 (0.07-8.60)	0.60 (0.01-20.95)	10.80 (0.24-322)
ADRIATIC	0.45 (0.001-8.00)	0.37 (0.001-7.70)	0.10 (0.005-3.00)	0.24 (0.001-88.4)	11.00 (0.01-9779)
CENTRAL MEDITERRANEAN	1.63 (0.27-3.00)	1.0 (0.4-26.0)	0.15 (0.12-0.15)	0.11 (0.1-1.40)	10.24 (0.40-26.0)
AEGEAN-LEVANTINE	3.96 (0.44-22.11)	3.61 (0.04-38.45)	0.25 (0.01-26.52)	--	9.85 (1.01-173)

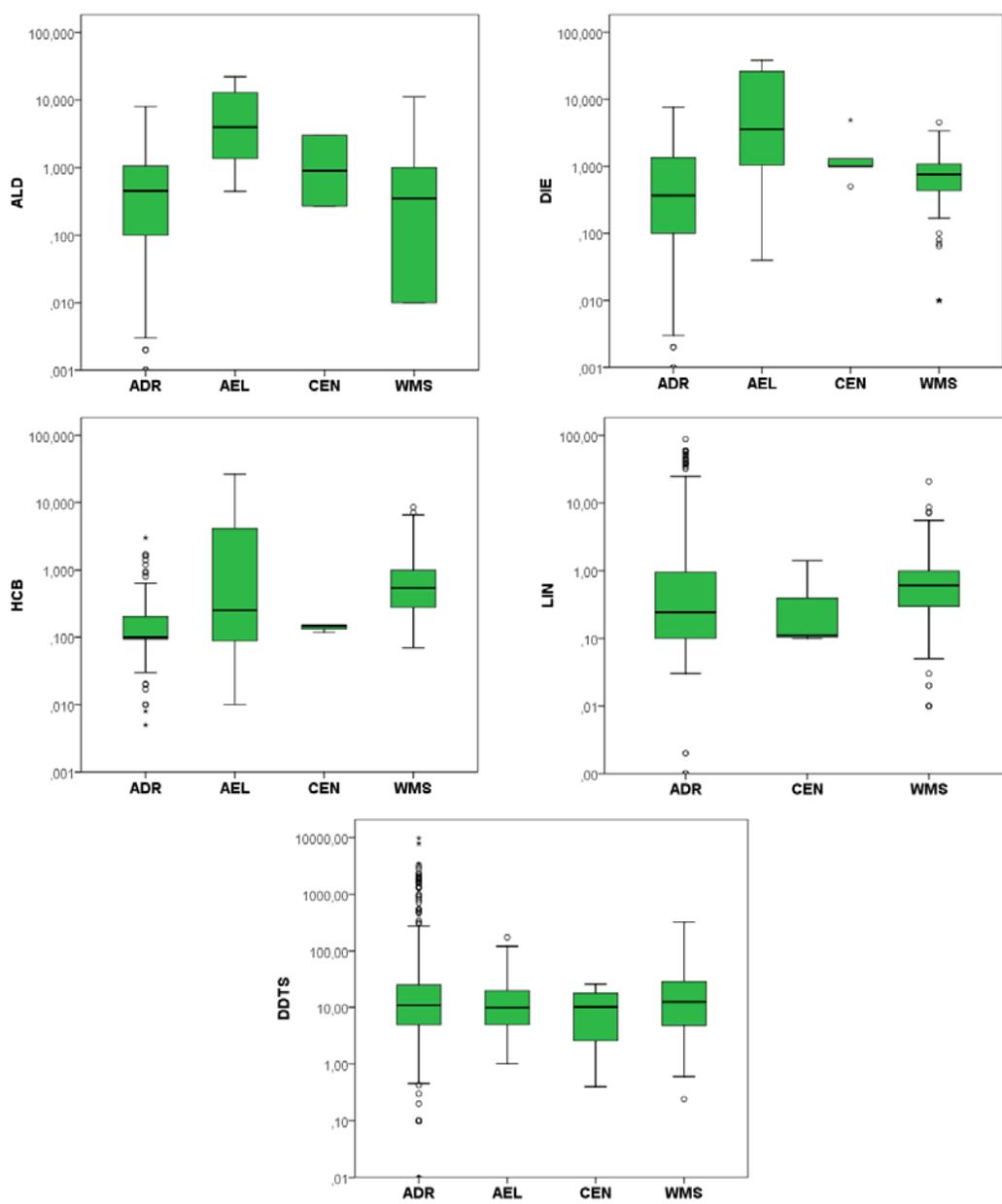


Figure 20. Concentrations of aldrin, dieldrin, HCB, lindane and DDTs (ng g⁻¹ dw) in *Mytilus galloprovincialis* from the different Mediterranean sub-regions.

Table 3.14 Concentrations of total DDTs in MG and MB in other studies.

Sp.	Location/Area	Year	Samples	DDTs (ng/g dw) ^a	Reference
MG	Morocco coast	1988-1990	15	101^{b,e}	El-Hraiki <i>et al.</i> (1994)
	Balearic Islands	1995-2000	5	15 (8-35)	Deudero <i>et al.</i> (2007)
	NW Med	1988-1989	11	137 (21-663)	Villeneuve <i>et al.</i> (1999)
	Ebro Delta	1989-1991	23	132 (11-758)	Sole <i>et al.</i> (2000)
	Spanish coast	2003-2004	96	20.4^b	Fernández <i>et al.</i> (2010)
	Western Med	2004-2006	99	1.2-16.6^e	Scarpato <i>et al.</i> (2010)
	Mali Ston Bay (HR)	2005-2007	80	1.59^b	Kozul <i>et al.</i> (2009)
	Adriatic Sea (HR)	1986-1992	32	6-50^b	Picer & Picer (1995)
	Adriatic Sea (IT)	1997-1998	18	8-16^{b,c}	Bayarri <i>et al.</i> (2001)
	Adriatic Sea (IT)	2002	453	9.9-10^b	Perugini <i>et al.</i> (2004)
Adriatic Sea (IT)	1992	123	2.2-11.3^{b,e}	Stefanelli <i>et al.</i> (2004)	
MB	Morocco coast	1988-1990	15	73^{b,e}	El-Hraiki <i>et al.</i> (1994)
	NW Med	1995	53	0.6 - 230	Porte <i>et al.</i> (2002)
	Ebro Delta	1992	29	18-70^b	Pastor <i>et al.</i> (1996)
	Adriatic Sea (IT)	1997-1998	28	34-41^{b,c}	Bayarri <i>et al.</i> (2001)
	Adriatic Sea (IT)	1997	211	14-32^{b,e}	Stefanelli <i>et al.</i> (2004)
	Adriatic Sea (IT)	2002	118	6.89-8.87^b	Perugini <i>et al.</i> (2004)
	Aegean Sea (GR)	1988-1991	697	50-93^b	Giouranovits-Psyllidou <i>et al.</i> (1994)
	Aegean Sea (TK)	1994-1998	18	68 (45-94)^d	Kucuksezgin <i>et al.</i> (2001)
	Marmara Sea	2003		146.3	Coelhan <i>et al.</i> (2006)

^a Values converted into dw assuming 19% and 24% DW for MG and MB, respectively

^b Mean values

^c ppDDE

^d DDE+DDD

^e ppDDT+ppDDE+ppDDD

Despite the similarity of the mean DDT levels for the four eco-regions (Figure 20), available data indicate that contaminants are not uniformly distributed throughout the sub-regions. In the **Western Mediterranean**, areas of particular concern include estuaries (Rhône, Ebro), ports, bays and gulfs (Barcelona, Fos, Bays of Algiers and Tunis, Naples, etc.) (Gomez-Gutierrez *et al.*, 2007).

An extensive monitoring study using caged mussels and covering the whole Western Mediterranean basin, carried out between 2004 and 2006, was able to determine the spatial distribution of chlorinated compounds, particularly DDTs and PCBs, in the basin (Scarpato *et al.*, 2010). A general presentation of the DDT results is shown in Figure 21. The levels of lindane were always below the detection limit in all sampling stations.

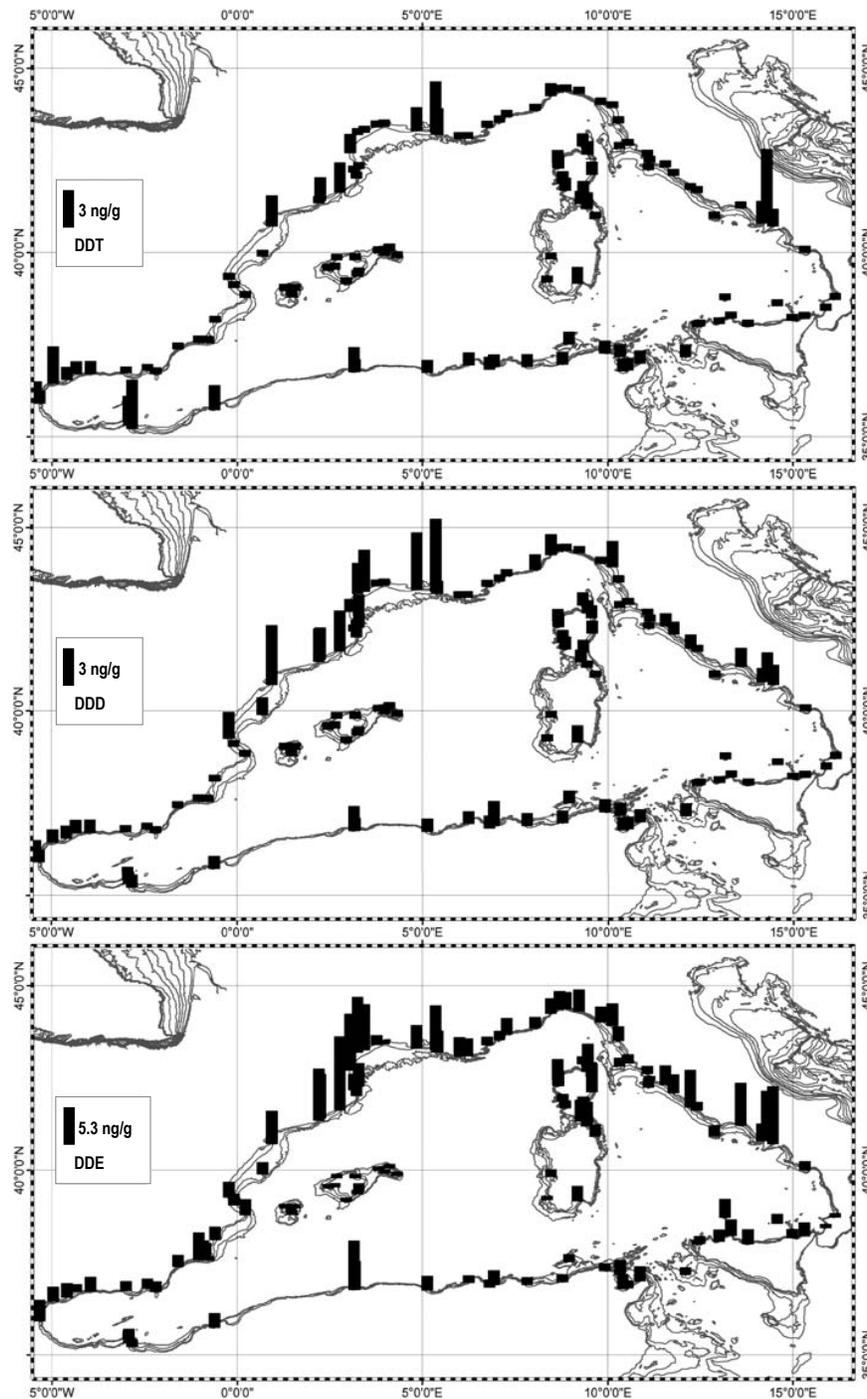


Figure 21. Levels of DDT, DDE and DDE ($\text{ng g}^{-1} \text{ dw}$) in mussels caged along the coast of the Western Mediterranean basin (Scarpato *et al.*, 2010).

Higher accumulation values are found at the main metropolitan areas and at the mouth of important rivers. There is evidence that river inputs represent the most important source of pesticides entering the Western Mediterranean Sea.

In terms of total DDTs accumulation, the most contaminated samples were recovered at the Marseille coastal area and close to the Rhone river mouth (9.9 - 16.6 ng g⁻¹). In the remaining part of the northwestern coast, from Marseille down to Barcelona, the levels oscillated around 10 ng g⁻¹. In the Barcelona metropolitan area, at the Besos and Llobregat River sites, values were similar to those found at Marseille (10.6 - 16.2 ng g⁻¹). Comparable values were also found in the Gulf of Naples (15.3 ng g⁻¹) and less than 10 ng g⁻¹ were found in the rest of the Italian peninsula.

In the Southern part of the W. Mediterranean, significant accumulation values of DDT were recorded at the Nador Lagoon (Morocco) and in Algiers Bay.

Coastal waters of the **Adriatic Sea** belong to unpolluted areas of chlorinated pesticides. Concentrations of aldrin, dieldrin, endrin, lindane and hexachlorobenzene in *Mytilus galloprovincialis* are in the low ng g⁻¹ range, with the exception of some stations from Albania. Concentrations up to 9779 ng g⁻¹ dw of total DDTs were found at the Durres Bay, most probably due to the presence of stockpiles of obsolete pesticides in the country.

Moderate concentrations of Lindane and DDTs were found in the Gulf of Trieste and the Marches region (Ancona), respectively, although they were banned in the 70s.

Information on the **Central Mediterranean** is almost lacking, although the region seems relatively free of hotspots of chlorinated hydrocarbons in marine bivalves, at least according to the limited availability of data.

In the **Aegean-Levantine** eco-region, organochlorine compounds, mainly DDTs, were determined in mussels and red mullets across the Greek coastal waters (SoHelME, 2005) and in some stations of Turkey and Cyprus. In all cases the concentrations of DDTs were quite low.

The highest concentrations of DDTs in mussels collected in the Aegean and Ionian Seas were found in the Amvrakikos Gulf (81.0 ± 45.8 ng g⁻¹), which according to these data seems to be moderately contaminated (Figure 22). This pollution is probably attributed to the intense agricultural activities in the area. In the Saronikos and Thermaikos Gulfs similar values were recorded, which were significantly lower (22.7 - 23.2 ng g⁻¹).

On the other hand, the spatial distribution of DDTs in *Mullus barbatus* (Figure 22) revealed a homogeneous pattern (av. 12.4 ng g⁻¹), indicating no point sources of pollution, in accordance with the banning of these compounds since the 1970s. The values for Cyprus and Turkey (Table 3.12) can also be considered in the low range.

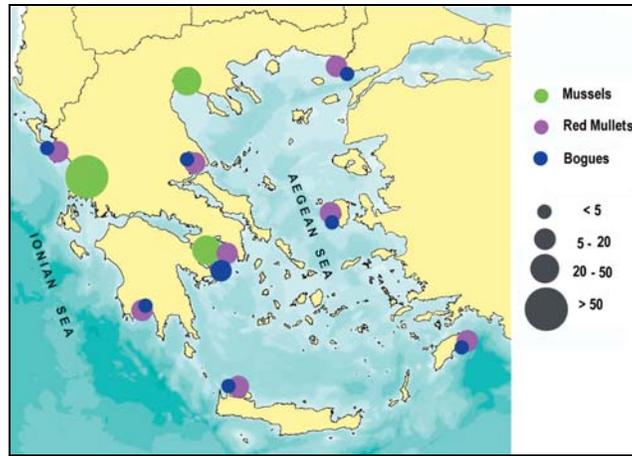


Figure 22. DDTs concentrations in mussels and fishes (ng g⁻¹ ww) in Greek seas.
(SoHeIME, 2005)

3.1.4 PCBs in biota

3.1.4.1 General distribution

As already stated in section 2.1, only data reported as the 7 ICES PCB individual congeners have been considered. A summary of their distributions in the indicator organism *Mytilus galloprovincialis* is shown in Tables 3.15 and 3.16. In addition, the information existing in the Italian Database SIDIMAR has been retrieved, taking into account the large number of samples and stations included, although the PCB congeners determined were not those corresponding to the 7 of reference (Table 3.17).

Table 3.15. Mean concentrations of PCB congeners in *Mytilus galloprovincialis* (ng g⁻¹ dw) by country.

Country	CB28	CB52	CB101	CB118	CB138	CB153	CB180	CB7
ALB	75.18	91.33	82.98		142.99		115.10	263.99
CRO	4.87	11.00	11.32	0.19	12.93	14.55	10.76	82.17
FRA	0.58	1.71	6.14	5.00	19.96	26.24	2.89	53.21
ITA ^a		6.91			20.49	25.43		
SPA	0.21	0.33	6.98	0.87	2.57	3.48	0.52	14.63
TUR	0.04	2.37	2.37	0.51	4.23	6.65	6.39	22.55

^a Data from SIDIMAR

Table 3.16. Statistics of PCBs (Σ 7CB) in *Mytilus galloprovincialis* (ng g⁻¹ dw) by country.

Country	n	Mean	Median	Min	Max	SD
ALB	33	263.99	162.36	31.08	874.71	225.74
CRO	60	82.17	66.18	2.23	286.89	75.32
FRA	254	53.21	30.20	3.75	1500.65	105.46
SPA	276	14.63	4.51	0.30	139.48	23.32
TUR	51	22.55	2.61	0.18	134.88	37.53

Table 3.17. Concentrations of PCB congeners (ng g⁻¹ dw) in mussels from Italian coasts (SIDIMAR database, 2001-2006).

Parameter	n	Mean	Median	Min	Max	SD
CB52	332	6.91	2.17	0.05	298.40	19.52
CB77	344	18.72	0.78	0.01	676.20	59.01
CB81	262	5.88	0.31	0.01	256.00	18.82
CB128	358	3.82	1.44	0.03	45.70	6.55
CB138	444	20.49	7.48	0.09	411.80	43.62
CB153	459	25.43	7.50	0.05	602.80	55.46
CB169	200	1.61	0.10	0.01	57.40	5.32
7PCBs	472	118.14	21.80	0.09	2380.00	269.19

A summary of the geographical distribution of the average values and ranges is shown in Figure 23. The higher levels in mussels were found in Albania, ranging from 100 to 874 ng g⁻¹ dw, although values up to 1500 ng g⁻¹ dw were found in one station of France, in 2004.

A spatial representation of the mean concentrations of the different stations is shown in Figure 24. A rather good coverage of the Western Mediterranean and the Adriatic Sea is obtained. However, there is no information in the database about the southern and eastern parts of the basin.

PCBs occur in the vicinity of industrial and urban sites, as well as in major river mouths. As it can be seen, the higher values are found in the NW Mediterranean, in the vicinities of the cities of Barcelona, Marseille and Genova, and in the Eastern Adriatic (Croatia and Albania). A number of stations around Sardinia exhibited also moderate levels of PCBs.

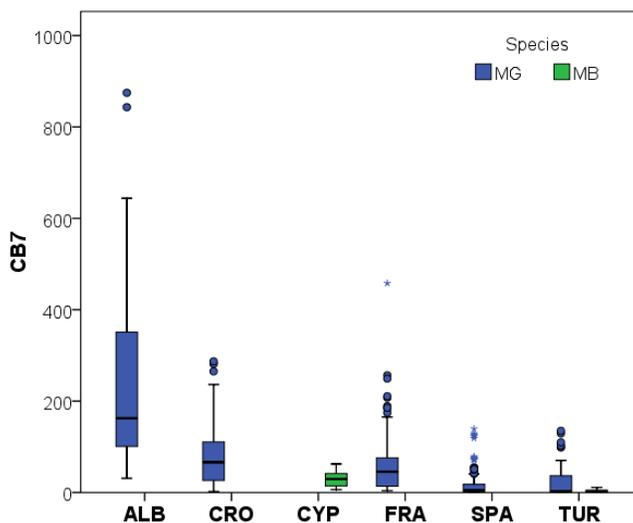


Figure 23. Concentrations of Σ7PCBs (ng g⁻¹ dw) in MG and MB, by country.

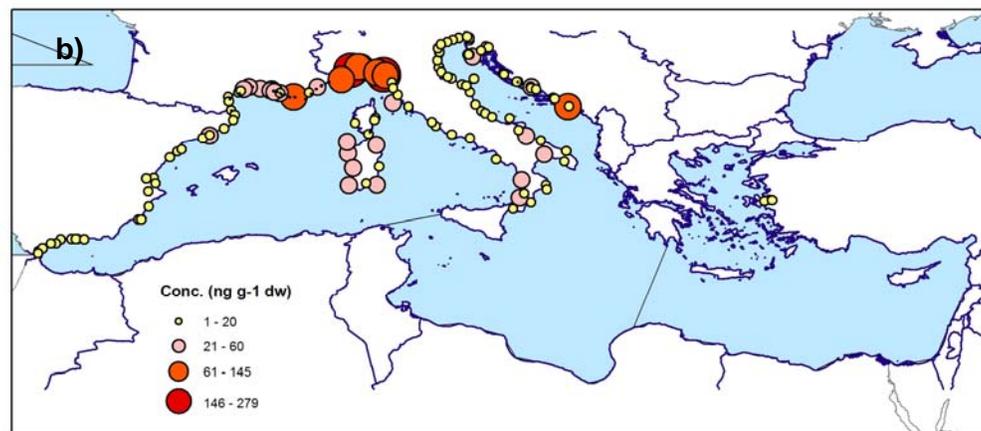
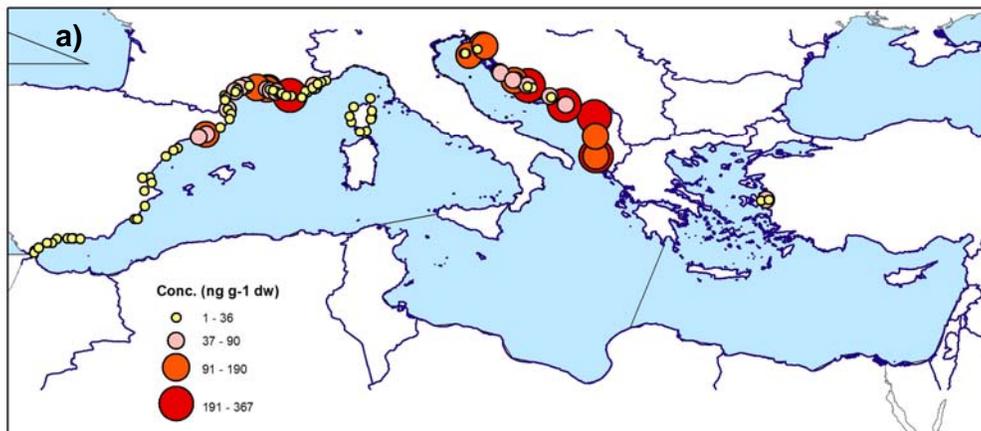


Figure 24. Mean concentrations (ng g⁻¹ dw) of a) Σ7CBs, and b) CB153 in *Mytilus galloprovincialis*

As far as fish is concerned, data on *Mullus barbatus* is limited to Cyprus and Turkey (Tables 3.18 and 3.19). Although the average values are similar in both countries, the median values are higher for Cyprus. This is reflected in the graphical representation of the mean concentrations of the different stations shown in Figure 25.

Table 3.18. Mean concentrations of PCB congeners in *Mullus barbatus* (ng g⁻¹ dw) by country.

Country	CB28	CB52	CB101	CB118	CB138	CB153	CB180	CB7
CYP	6.06	2.79	21.58	-	9.28	9.14	11.06	29.49
TUR	0.03	2.34	2.34	0.51	4.19	6.59	6.32	22.33

Table 3.19. Statistics of PCBs (Σ 7CB) in *Mullus barbatus* (ng g⁻¹ dw) by country.

Country	n	Mean	Median	Min	Max	SD
CYP	48	29.49	29.40	6.28	62.55	17.59
TUR	98	22.33	2.50	0.22	134.88	37.56

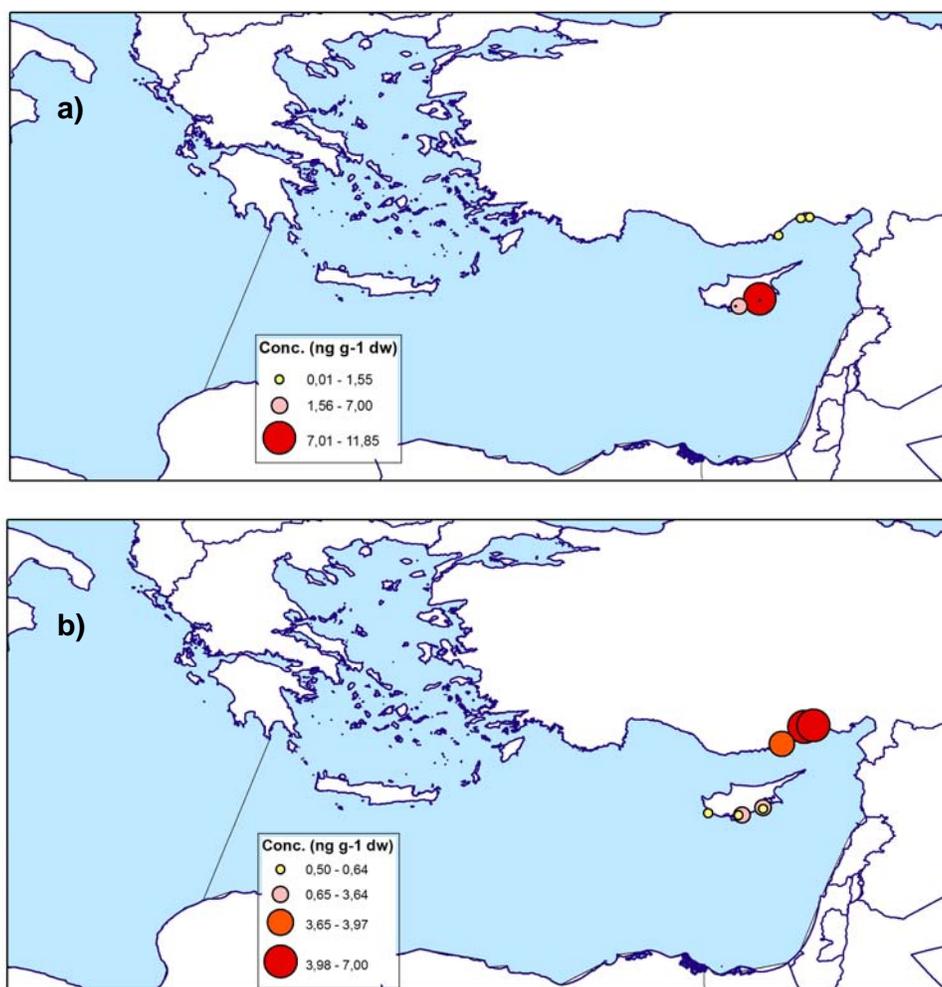


Figure 25. Mean concentrations (ng g⁻¹ dw) of a) CB153, and b) Σ 7CBs in *Mullus barbatus*.

3.1.4.2 Distribution of PCBs in biota within eco-regions

The congeners CB138 and CB153 together with the 7 ICES congeners have been selected for the comparison of values within regions, as they are the most representative both in the MEDPOL database and in the open literature. An overview of the data corresponding to mussels (*Mytilus galloprovincialis*) for the different eco-regions is shown in Table 3.20 and Figure 26. Data show concentrations much higher in the Western than in the Eastern Mediterranean, also according to a recent assessment of persistent toxic substances in the Mediterranean (UNEP, 2002). Data is very limited for the Central Mediterranean and is biased in the case of the Adriatic by the high levels of mussels from Albania.

Table 3.20. Median and concentration ranges of chlorinated compounds in *Mytilus galloprovincialis* (ng g⁻¹ dw)

Eco-region	CB138	CB153	Σ7CBs
WESTERN MEDITERRANEAN	4.20 (0.07-566)	6.18 (0.16-603)	12.6 (0.30-1501)
ADRIATIC	6.10 (0.1-350)	4.80 (0.05-85.5)	90.84 (2.23-875)
CENTRAL MEDITERRANEAN	5.00 (0.30-23.0)	4.27 (0.70-38.0)	--
AEGEAN-LEVANTINE	0.49 (0.03-25.3)	0.77 (0.05-39.8)	2.61 (0.18-135)

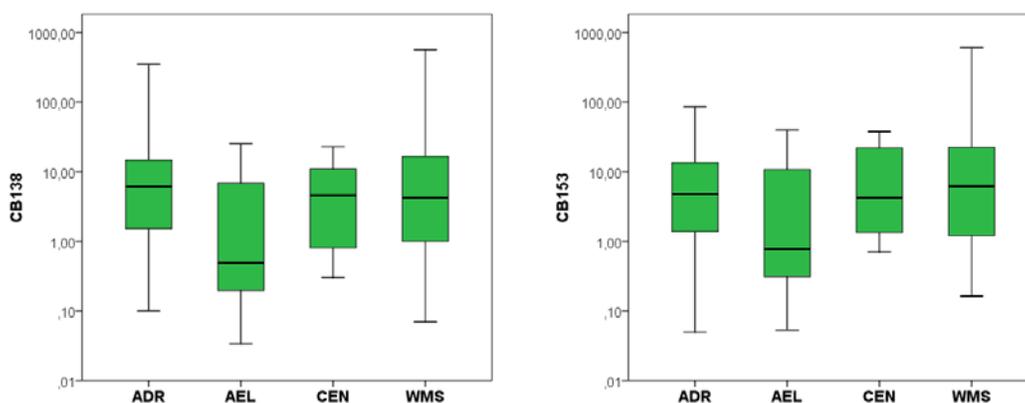


Figure 26. Concentrations of PCBs (ng g⁻¹ dw) in *Mytilus galloprovincialis* from the different Mediterranean sub-regions.

A number of recent studies published in the literature are summarized in Table 3.21. Concentrations of the 7 ICES congeners in mussels generally range from below 10 to around 100 ng g⁻¹, depending on the level of environmental pressure on the stations. Higher values correspond to an evidence of direct PCB inputs.

Table 3.21. Concentrations of $\Sigma 7\text{PCBs}$ (ng g^{-1} dw) in MG and MB in other studies.

	Location/Area	Year	CB28	CB52	CB101	CB118	CB138	CB153	CB180	$\Sigma 7\text{PCBs}$	Reference
MG	Iberian Med. coast	2003-2004	-	-	-	-	-	-	-	58	Fernández <i>et al.</i> (2010)
	Catalan coast	1986	0.21 – 15.68	0.42 – 23.37	1.26 – 44.58	1.74 – 44.32	4.32 – 77.37	3.16 – 47.74	0.37 – 15.11	11.5-269	Porte and Albaiges (1994)
	NW Med	1988-1989	-	1.1 - 37	4 – 130	-	4.8 – 270	7.7 – 120	-	-	Villeneuve <i>et al.</i> (1999)
	Ebro Delta	1989-1991	3.7	2.6	8.9	17.4	30.0	17.9	4.2	95	Sole <i>et al.</i> (2000)
	Balearic islands	1996-2005	2.6 (2.1-7.9)	4.2 (2.1-17.4)	6.8 (1.6-16.8)	14.2 (2.1-59.5)	18.2 (2.1-46.3)	27.9 (2.1-66.8)	5.8 (2.1-17.9)	80 (14-194)	Deudero <i>et al.</i> (2007)
	Thau Lagoon	2005	0.12-0.17	0.09-0.25	0.96-3.54	0.80-3.33	2.30-12.23	5.24-18.67	0.32-0.50	10 - 39	Castro-Jiménez <i>et al.</i> (2008)
	Gulf of Naples	2003	0.5 (ND-2.4)	1.3 (ND-2.2)	7.3 (ND-12.3)	15.4 (7.1-21)	23 (10-35)	29 (13-43)	3.5 (1.9-6.4)	80 (32-122)	Naso <i>et al.</i> (2005)
	Western Med.	2004-2006	ND-7.7	-	ND-18	ND-12	6.0-27.6	4.0-42.3	-	1.1-83.8	Scarpato <i>et al.</i> (2010)
	Tyrrhenian and Adriatic Sea		<d.l.–7.6	<d.l.–2.2	<d.l.–62.5	<d.l.–29.8	6.3–263.2	3.21–315.8	<d.l.–42.1	-	Pinto <i>et al.</i> (2008)
	Adriatic Sea	2002	ND	4-8.9	3.7-10.3	1.6-3.4	4.7-11.1	3.7-8.2	0.8-2.1	18-52	Perugini <i>et al.</i> (2004)
	Western Adriatic	1997-98	-	-	-	-	-	-	-	7 - 97	Bayarri <i>et al.</i> (2001)
	Eastern Adriatic	2006	1.74	4.99	1.61	3.86	11.92	10.21	1.21	35.57	Kljatovic-Gaspic <i>et al.</i> (2010)
	Mali Ston Bay (Eastern Adriatic)	2005-2007	1.24	3.57	1.38	0.28	1.29	1.75	0.43	9.94	Kozul <i>et al.</i> (2009)
Vlora Bay, Albania (Eastern Adriatic)	2006	-	-	-	-	-	-	-	38 – 74*	Corsi <i>et al.</i> (2010)	

MB	Ebro Delta	1989-1990	0.3-1.5	0.7-7.3	1.3-9.1	2.6-20	9.3-44	11-37	5.3-22	31-141	Sanchez <i>et al.</i> (1993)
	Catalan coast	1986	0.33 – 12.58	0.92 – 25.42	1.25 – 54.58	4.29 – 94.58	11.54 – 236.67	5.58 – 147.50	2.46 – 75.00	27-646	Porte and Albaiges (1994)
	NW Mediterranean	1995	-	-	-	-	-	-	-	1.7 - 450	Porte <i>et al.</i> (2002)
	Gulf of Naples	2003	1.2 (ND-2.2)	ND	1.1 (ND-3.5)	13.5 (ND-17.4)	24 (17-41)	33 (26-42)	19 (11-28)	92 (54-134)	Naso <i>et al.</i> (2005)
	Western Adriatic	1997-98	-	-	-	-	-	-	-	83 - 181	Bayarri <i>et al.</i> (2001)
	Adriatic Sea	2002	ND	9.4-10.8	4.6-5.2	9.5-9.8	26.5-27.1	20-20.3	3.3	76	Perugini <i>et al.</i> (2004)
	Vlora Bay, Albania (Eastern Adriatic)	2006	-	-	-	-	-	-	-	17 – 56*	Corsi <i>et al.</i> (2010)
	Marmara Sea	2003	0.84-4.04	0.75-5.4	0.31-11.94	1.57-11.59	3.32-28	2.82-26	0.90-17.87	10.5-105	Coelhan <i>et al.</i> (2006)

*Sum of 42 PCB congeners (Aroclor 1254, 1260)

In the **Western Mediterranean**, the baseline levels are high and the sites most affected are the areas of Barcelona, Marseille (up to $1500 \text{ ng g}^{-1} \text{ dw}$) and the Ligurian Sea from Livorno to Nice, including the Genova harbour (Figure 24). The levels of PCBs found in mussels and mullets from the Western Mediterranean during the 70s and 80s have been reviewed and discussed by Tolosa et al. (1997). Localised “hot spots” were identified at enclosed locations along the French coast (Toulon, Thau, Sete, Cannes, Monaco), off industrial and highly populated cities (Marseille, Barcelona and Genova), and at the mouths of Rhone and Ebro Rivers. It was clear that in western Mediterranean coastal areas, rivers and wastewater discharges are the major sources of PCBs.

A recent monitoring study using caged mussels and covering the whole Western Mediterranean basin has been able to determine the spatial distribution of PCBs in the basin (Scarpato *et al.*, 2010). A general presentation of the results is shown in Figure 27.

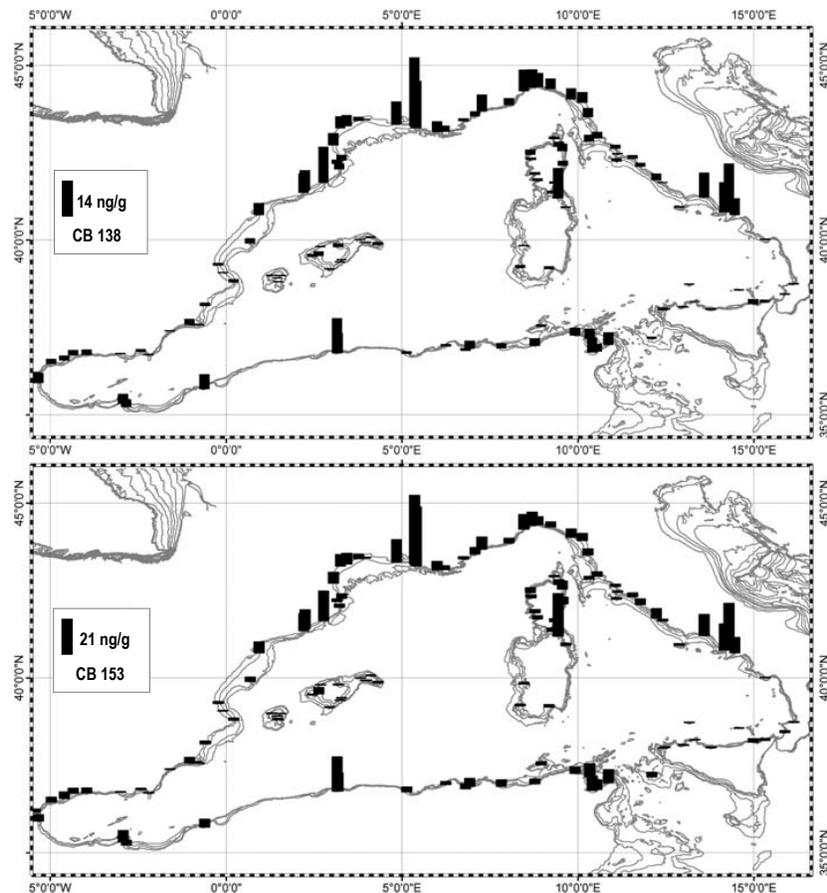


Figure 27. Levels of PCB congeners 138 and 153 ($\text{ng g}^{-1} \text{ dw}$) in mussels caged along the coast of the Western Mediterranean basin (Scarpato *et al.*, 2010).

The PCB congeners 138 and 153 show maximum levels of accumulation along the French coast, particularly at Marseille basin (42.3 ng g^{-1}) and to a lesser extent close to the Rhone mouth. Concentrations are also important along the Italian coast, in Naples

(47.0 ng g⁻¹) and Bagnoli (28.0 ng g⁻¹), in Sardinia (38.0 ng g⁻¹), and in Barcelona (19.2 - 32.1 ng g⁻¹). In southern Mediterranean, significant values were found in Algiers (34.6 ng g⁻¹). For the remaining Mediterranean sites, detected accumulation values for both congeners do not exceed 10 ng g⁻¹.

The **Central Mediterranean** was relatively free of hotspots of chlorinated hydrocarbons in marine bivalves, at least according to the present availability of data.

The **Adriatic Sea** has been extensively monitored for PCBs. It includes from unpolluted to moderately polluted coastal areas. Concentrations are low along the western coast, with areas with high concentrations in the eastern bank, along the coasts of Croatia and Albania.

In the case of **Aegean-Levantine** spatial analysis is limited to Cyprus (fish) and Turkey (mussels and fish). The values on *Mullus barbatus* in the MED POL database can be considered in the medium-low range, taking into account the higher accumulation capacity of fish with respect to mussels.

PCBs in marine biota across the Greek coastal environment were determined within the Greek monitoring system, and the main results are illustrated in Figure 28 (SoHeIME, 2005). The analysis provided evidence of a contamination gradient when mussels were used as an indicator. The highest contamination was observed in mussels collected from the Saronikos Gulf (industrial and urban effluents). On the other hand, bioaccumulation in fish revealed a homogeneous pattern indicating no point sources of pollution.

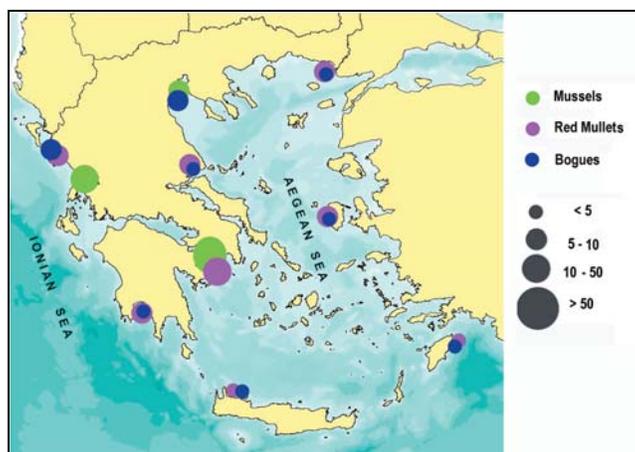


Figure 28. PCBs concentrations in mussels and fishes (ng g⁻¹ ww) in Greek seas. (SoHeIME, 2005)

3.2 Temporal trends

Marine monitoring implies the repetitive observing for defined purposes, of one or more elements of the marine environment, according to prearranged spatial and temporal schedules using comparable methodologies. The temporal trend monitoring starts with the objective to detect trends in concentrations with the aim of monitoring the effectiveness of control measures taken at pollution hot spots. Trends in pollutant or contaminant levels, in general, are also considered as “state” indicators of pollution and are included in most of the regional monitoring programmes to provide inputs to the assessments of the state of the marine environment.

Surface sediments and biota can be used for recognizing possible temporal trends of metals and organochlorine compounds in the marine environment and, thus, can be an important tool for the assessment of the effectiveness of control measures taken at the pollution hot spots and also for state assessment. However, data variability can be influenced by several factors other than contaminant inputs, namely those associated with sampling and the representativeness of the collected samples. In any case, the first requirement is the availability of data series long enough, so that long-term monitoring programmes are maintained in time.

In a previous evaluation of the database for the trend monitoring of contaminants (UNEP, 2005) it was concluded that the MED POL Phase III programme objectives preliminarily set, were not enough to achieve the temporal trend of any selected contaminant for a selected site. The major reason for this was the various difficulties in analysing data, especially when normalization was intended for reducing the variance of the data set by taking into account the differences in morphology (e.g. sediment grain size) or composition (e.g. tissue fat content) of the samples. Both the selected trace metals and the organic contaminants will co-vary strongly with such factors but normalization of the results against any of the normalisers (Reference Methods for Marine Pollution Studies, No.63) was not considered in any of the programmes.

A second aspect to be considered is the time span necessary for trends assessment. In general, the first temporal trend evaluation using sessile marine organisms can be performed with data sets of more than five years ongoing programmes. The use of sediments still require a longer time span (>10yr) for evidencing and assessing significant variations. However, after ten years of the monitoring programme, certain countries still did not have valid and continuous data covering at least five years. As shown in tables 2.3, 2.6 and 2.9, there are very few countries fulfilling this requirement.

Consequently, the analysis has been focused on *Mytilus galloprovincialis* (MG) and *Mullus barbatus* (MB), with the exception of Israel, where the clam *Mactra corallina* (MC) has been considered. Sediments have not been considered due to the limited time span (<10yr) for evidencing and assessing significant variations. However, a few studies have been taken from the literature.

Trends are first demonstrated through linear regression and then more accurately assessed through the Trend-y-Tector methodology for deciding whether there is a significant (downward) trend or not, as indicated in section 2.2.2. Some stations that are believed to be representative of the trends in each country/eco-regions have been selected for indicative purposes, not necessarily supported by a comprehensive statistical analysis. Particular attention has been paid to the assessment of hot spots.

3.2.1 Trace metals in sediments

As shown in Table 2.3, sediments have not been monitored continuously in the MED POL Programme by most countries for evidencing and assessing temporal variations. The only data set with a 10 years time span in that of Israel. Although the levels of metals is rather low in this area, a clear downward trend is observed in the samples exhibiting the higher levels like those from Haifa Bay, as it is shown in Figure 29. The sediments in the surroundings of the centre of the Bay also exhibit the same trend for Zn (Figure 30). In general, there is no observable trend for Cu.

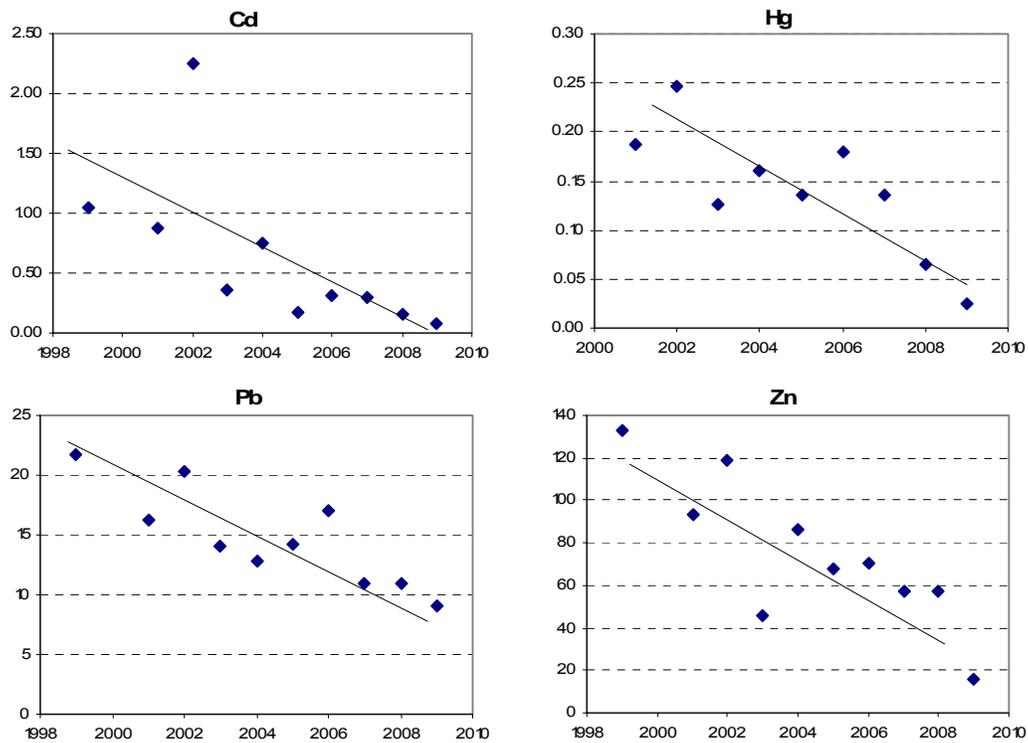


Figure 29. Concentrations of trace metals ($\mu\text{g g}^{-1}$ dw) in station ISRTMH27 from Haifa Bay (Israel)

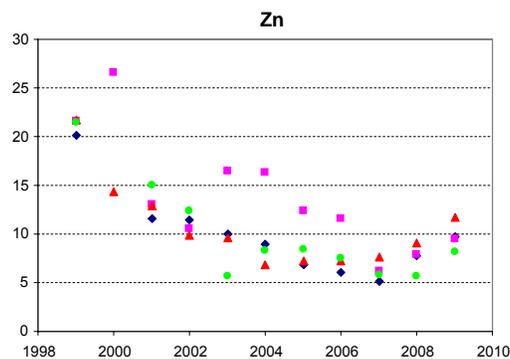


Figure 30. Concentrations of Zn ($\mu\text{g g}^{-1}$ dw) in sediments from different areas of the Haifa Bay (Israel)

Other studies have been taken from the literature. In this respect, within the framework of the French RNO monitoring system, including 48 stations from the Gulf of Lions (Figure 7), 21 of them were monitored in 1995 and 2005. A summary of the average concentrations is shown in Table 3.22. As it can be seen, although the mean and median values have apparently decreased for Pb, Zn and Cu, only a slight evidence of a downward trend is observed for Pb (Figure 31). Unfortunately, there is no information about the most polluted area of Marseille-Fos.

Table 3.22. Average concentrations of trace metals ($\mu\text{g g}^{-1}$ dw) in sediments from the Gulf of Lions (RNO, IFREMER, 2005).

	Year	Cd	Hg	Pb	Zn	Cu
mean	1995	0.12 \pm 0.02	0.06 \pm 0.03	33.17 \pm 6.10	82.61 \pm 9.93	14.73 \pm 4.04
	2005	0.13 \pm 0.03	0.05 \pm 0.02	26.01 \pm 6.32	74.00 \pm 16.02	13.46 \pm 4.76
median	1995	0.11	0.06	33.4	83	15.2
	2005	0.12	0.06	25.3	72	13.3

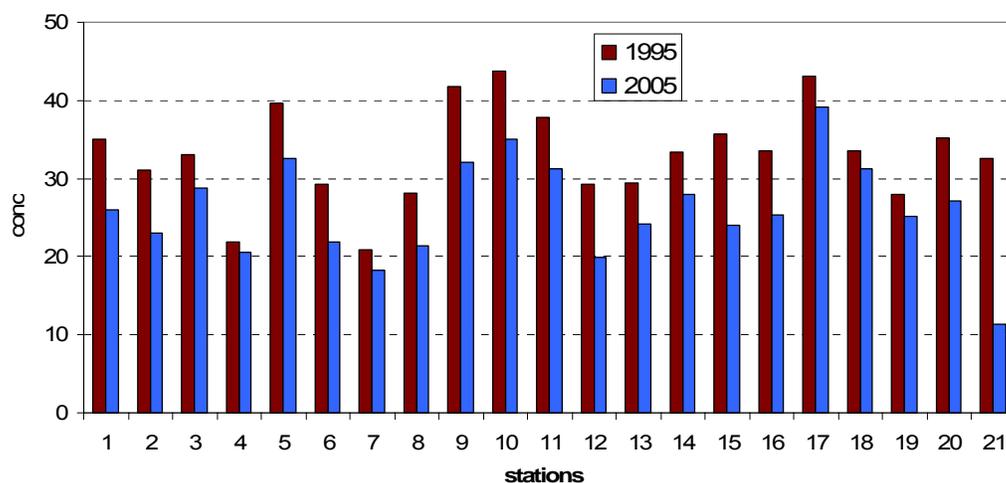


Figure 31. Concentrations of Pb ($\mu\text{g g}^{-1}$ dw) in sediments from different stations of the Gulf of Lions (RNO, IFREMER, 2005).

Analysis of metals in sediments from the Gulf of Tunis (Cd, Pb, Hg, Cu and Zn) performed in 1999, 2003 and 2010 demonstrated that trace metals remain in the same range of concentrations for the last 12 years, except cadmium that decreased more than 10 fold.

Mercury is also a critical contaminant in the Gulf of Trieste (Covelli *et al.*, 2001; Horvat *et al.*, 1999; Hines *et al.*, 2000). Data showed that even 10 years after the closure of the Idrija mercury mine, no decline of mercury concentration in the Gulf was observed.

3.2.2 Trace metals in biota

On the basis of recent UNEP/MAP-MED POL monitoring data, it appears that the overall concentrations of trace metals in bivalves do not exhibit clear trends for the different countries, with the exception of particular cases (Figure 32). For example, in Slovenia, concentrations of Cd and Hg in *Mytilus galloprovincialis* seem to be decreasing during the last decade. A similar trend is observed for Morocco and Tunis (not shown) although the number of stations in these countries is very limited. In other cases (e.g. Italy), although the median levels are more or less steady there is an apparent decline of outlier values, which may reflect a general improvement of the hotspots.

However, a better perspective of these trends can be obtained considering individual stations, as it will be illustrated in the following sections for the different sub-regions.

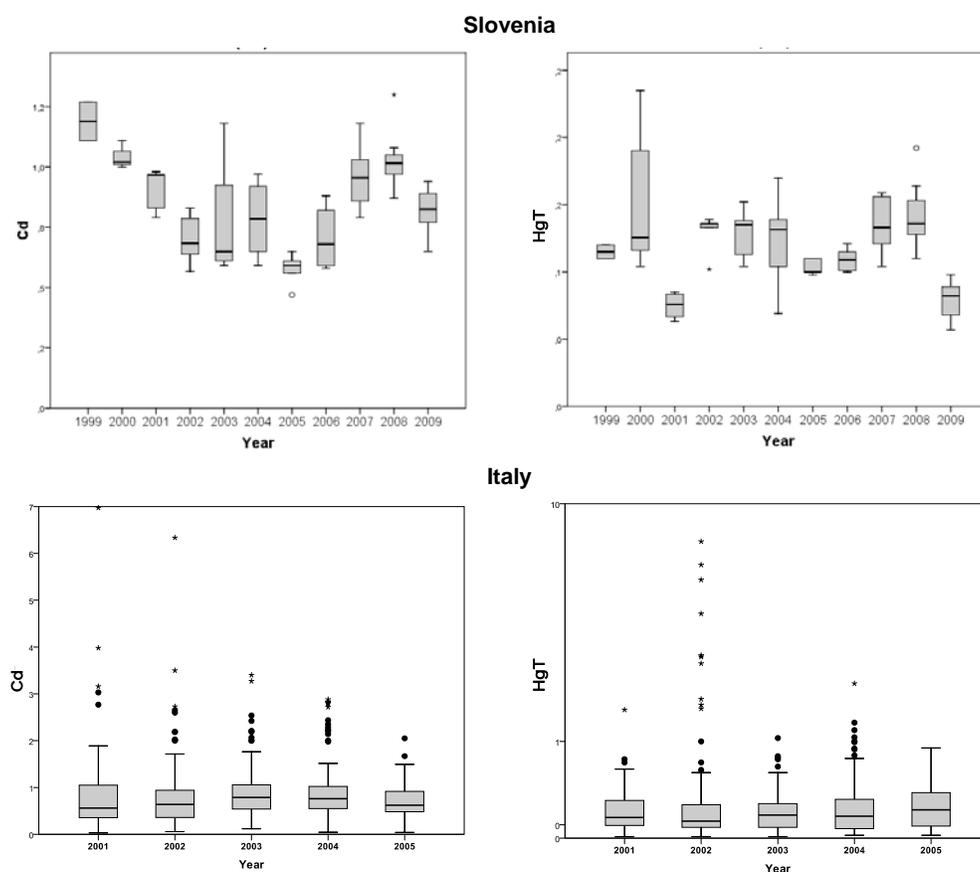


Figure 32. Temporal trends of Cd and total Hg in samples of *Mytilus galloprovincialis* from Slovenia and Italy.

3.2.2.1 Western Mediterranean

The available data for trends assessment in the region is practically limited to the stations located in the French and Italian coasts, and to one station of Morocco (Oued Laou) where a decreasing trend of concentrations is observed, from 1999 to 2005, for Cd and Hg but not for Pb (Figure 33). The Spanish stations have been monitored during 3-4 years and the concentrations were rather stable but no trend analysis can be made.

A complete dataset for 21 stations from the French coast and covering the period 1999-2010 shows stable concentrations of metals in most of them, with some decreasing trends in a few areas (e.g. the costal lagoons of Leucate and l'Ayrolle, and the Corsica stations) (Figure 34A). However, the stations exhibiting the higher values (e.g. the costal lagoons of Thau, Fos and Camargue, or the areas close to Marseille and Toulon) are still showing an increase (Figure 34B). Copper does not show, in general, a clear trend.

In the case of Italy, although the time series are more limited (4-5 years), it appears that the most contaminated stations identified previously (e.g. Genova, Fiumicino and Naples) show evidence of a decline of trace metal levels (Figure 35). However, the levels continue to increase in some of them (e.g. Piombino) (Figure 36).

As far as the Sardinia stations are concerned, there are a variety of situations as reflected in Figure 36. In Cagliari, for instance, the levels are increasing whereas in Olbia and in most of the other stations the trend is the contrary. In general, in the less contaminated stations, concentrations remain stable along the time. This pattern is extensive to most of the other continental stations. Another general feature is that the trends behaviour of all metals is usually similar, except when a specific source can be identified.

Oued Laou (Morocco)

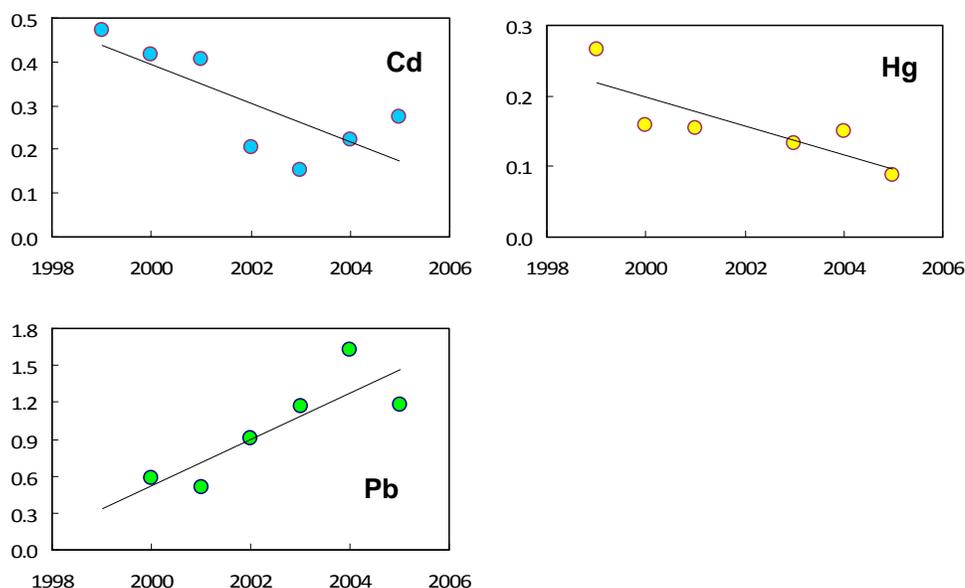
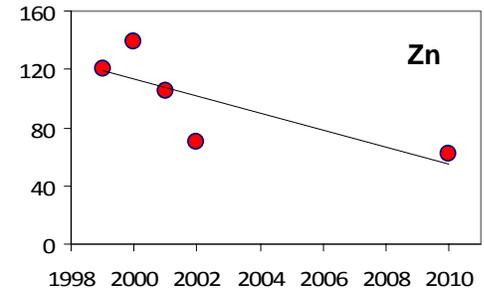
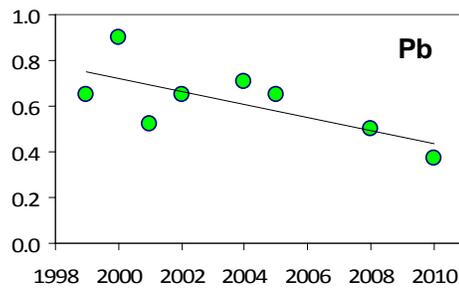
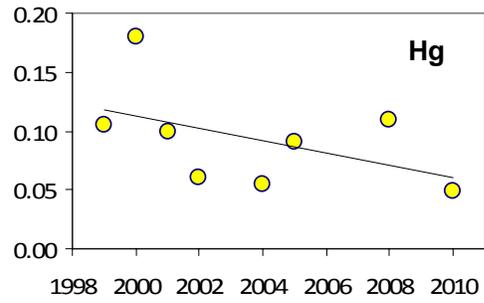
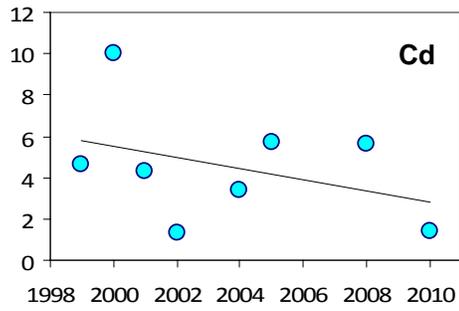


Figure 33. Temporal trends of trace metals in samples of *Mytilus galloprovincialis* from the station at the mouth of the Oued Laou (Morocco).

A - Etang de l'Ayrolle (Narbonne)



B - Les Stes. Maries de la Mer (Camargue)

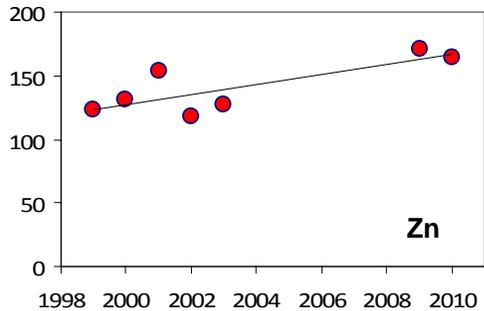
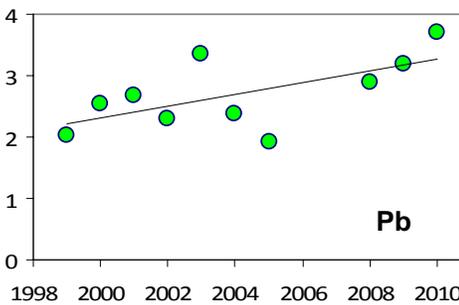
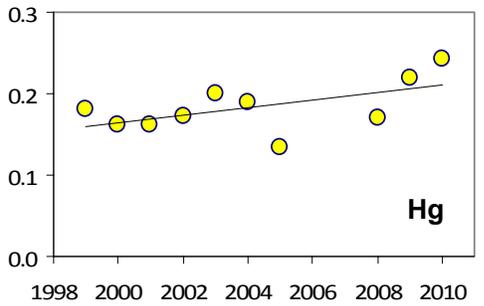
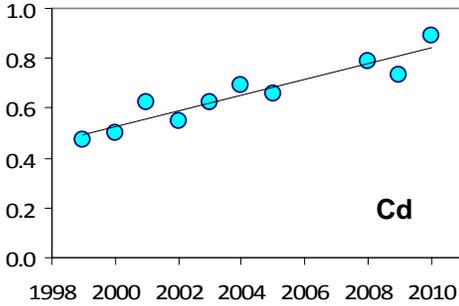
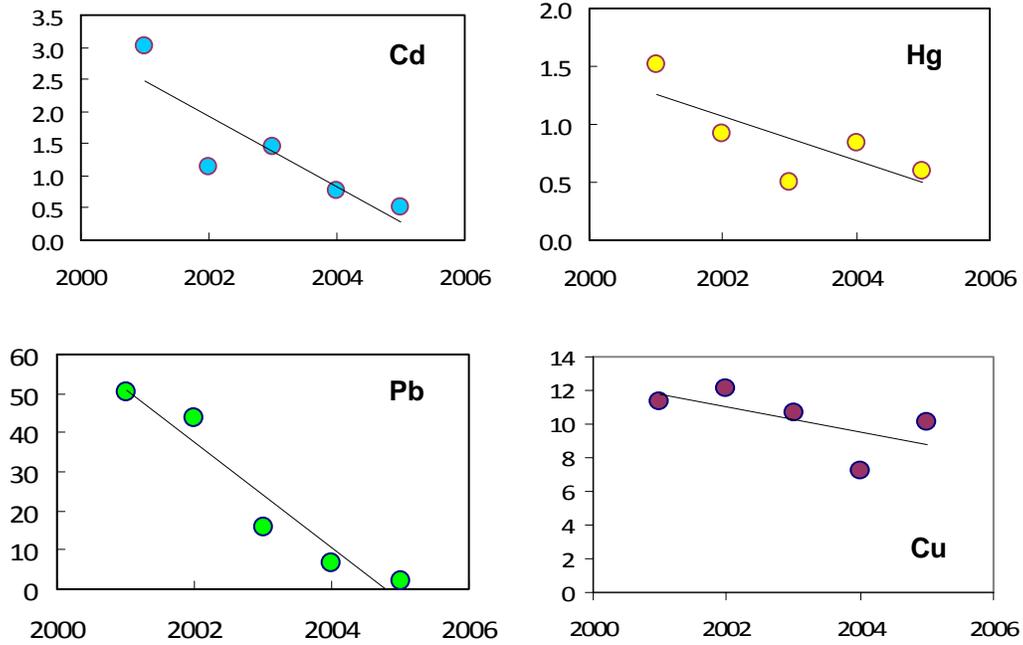


Figure 34. Temporal trends of trace metals in samples of *Mytilus galloprovincialis* from individual stations at the French coast.

Vado Ligure



Fiumicino

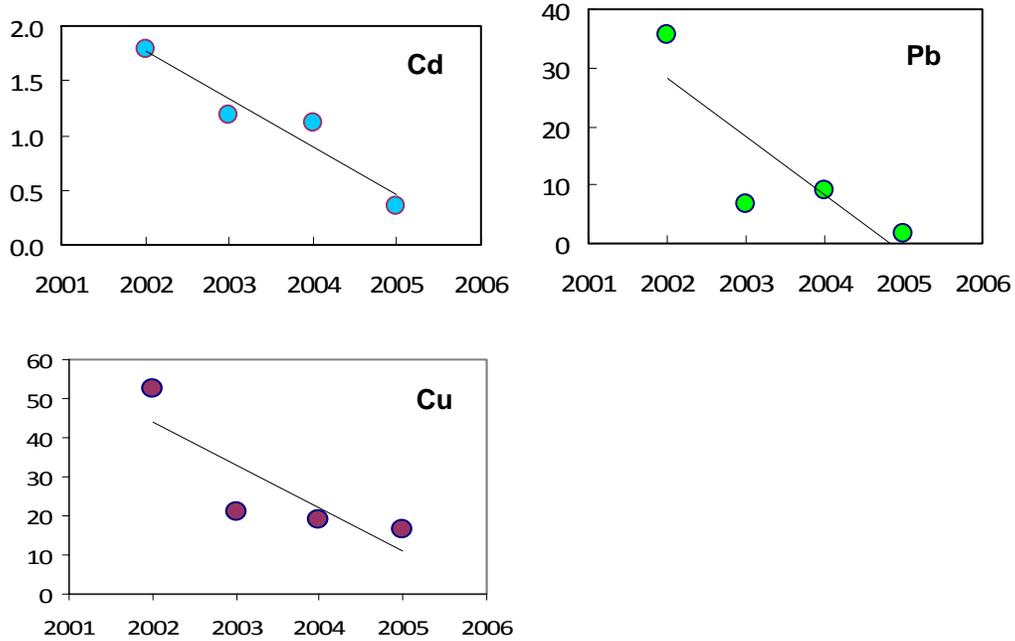
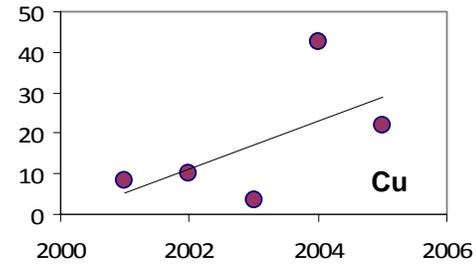
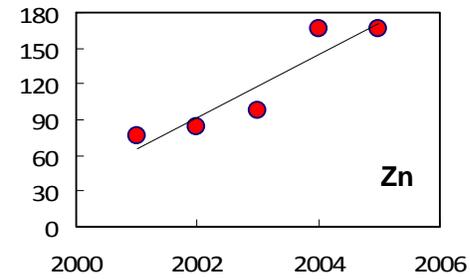
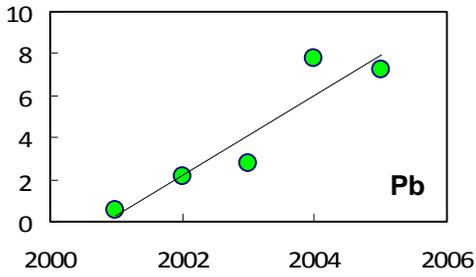
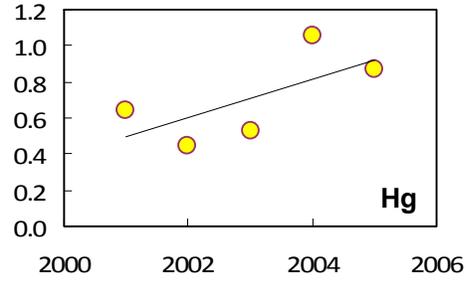
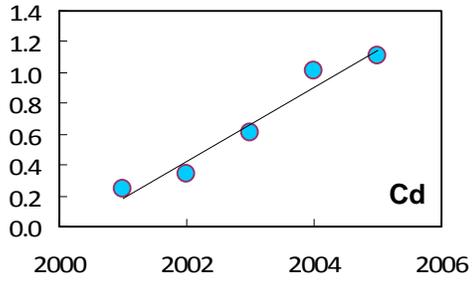
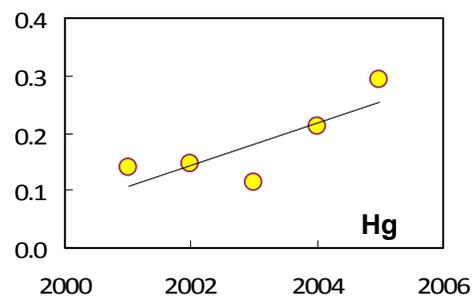
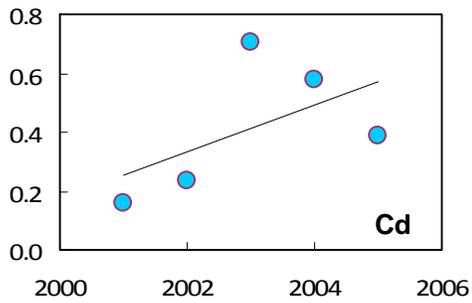


Figure 35. Temporal trends of trace metals in samples of *Mytilus galloprovincialis* from individual stations at the Italian coast.

Piombino



Cagliari (Sardinia)



Olbia (Sardinia)

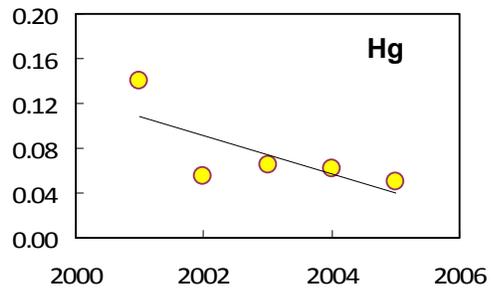
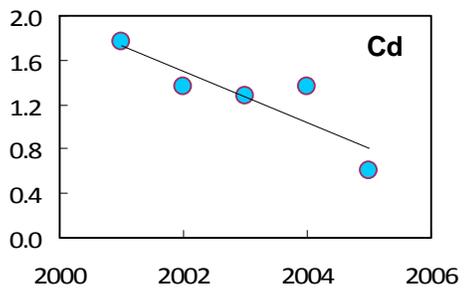


Figure 36. Temporal trends of trace metals in samples of *Mytilus galloprovincialis* from individual stations at the Italian coast.

Additional trends are reported in the literature. The longest temporal series existing in the region is probably the one derived from the French monitoring system (RNO). Data on trace metals for samples of *Mytilus galloprovincialis* collected in 21 stations along the period 1979-2006 are available and clearly show the general decline of concentrations, particularly of the higher ones, during this time span. Illustrative examples are given in Figure 37.

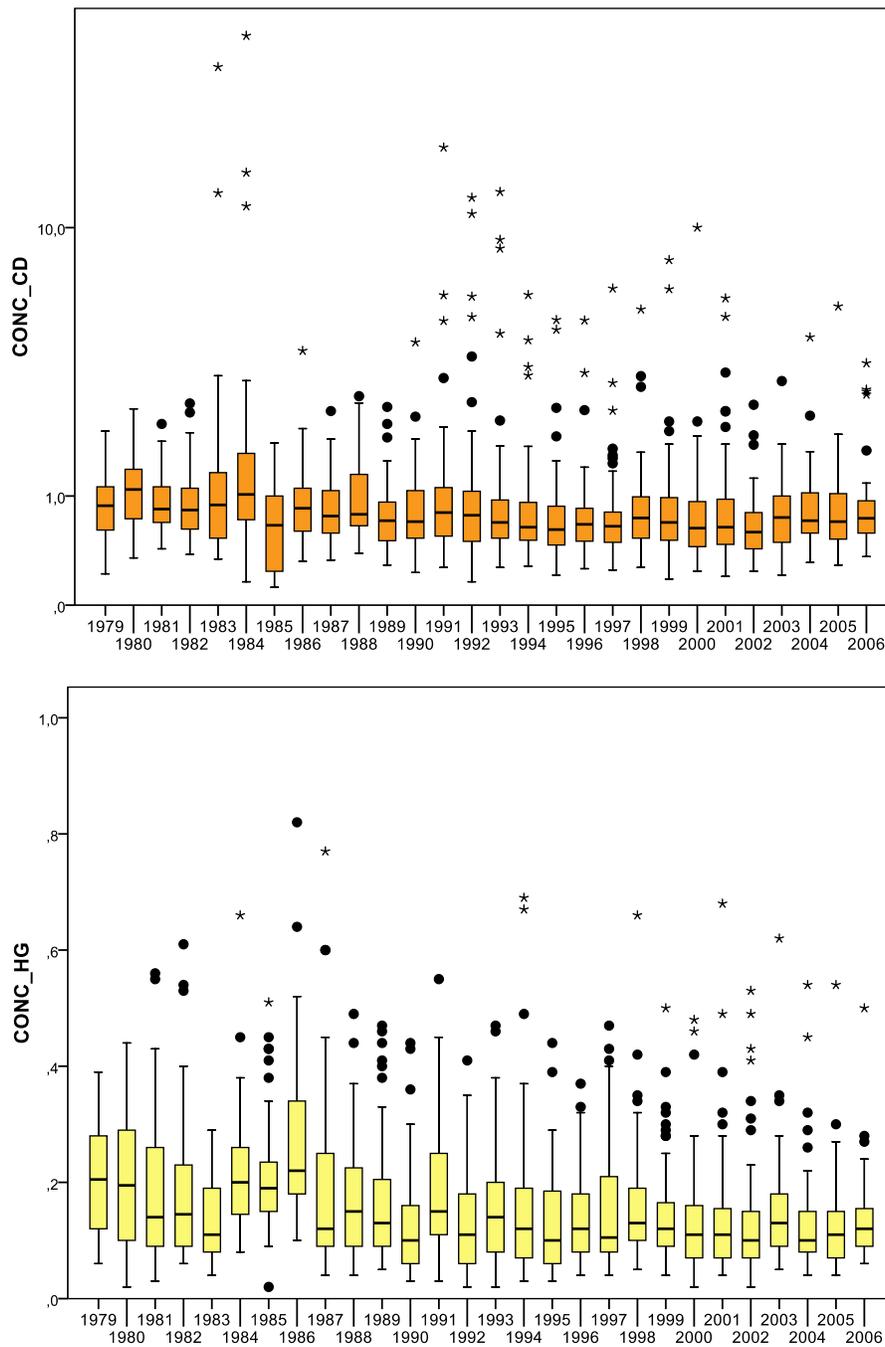


Figure 37. Temporal trends of Cd and Hg in *Mytilus galloprovincialis* from the RNO stations.

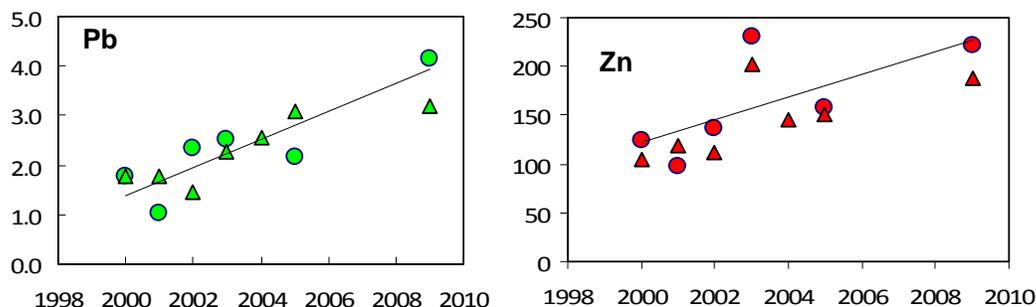
3.2.2.2 Adriatic

Temporal trends of pollution based on Eionet data (1998-2005) prepared by EEA show that concentrations of Hg in marine organisms are moderate in Northern, NE and NW part of the Adriatic Sea basin, and lower in the southern part of the basin, without any definite tendency. Concentrations of Cd in marine organisms show low concentrations and no significant trend in northern and eastern coast and decreasing trends along the western coast (EEA, 2010). However, in the stations holding high values for cadmium, there is a statistically general upward trend for this compound. This is the case for Rijeka and Kastela Bays (Croatia) that also exhibit upward trends for Pb, Zn and Cu (Figure 38).

On the other hand, Kljaković-Gašpić *et al.* (2007) monitored the blue mussel (*Mytilus galloprovincialis*) in the Mali Ston Bay, located on the eastern Adriatic coast, from 1998 to 2005. Analysis of temporal trends during the 7 years of monitoring showed that metal concentrations have not changed significantly over time.

A larger monitoring survey carried out during the 2001-2005 period in the Croatian coast determined that Pb and Hg were significantly elevated in the urban and industrial areas, while Cd was more uniformly distributed across the monitored sites. Metal concentrations did not change either during this five year-period (Kljakovic-Gaspic *et al.*, 2010).

Kastela Bay (Croatia)



Rijeka Bay (Croatia)

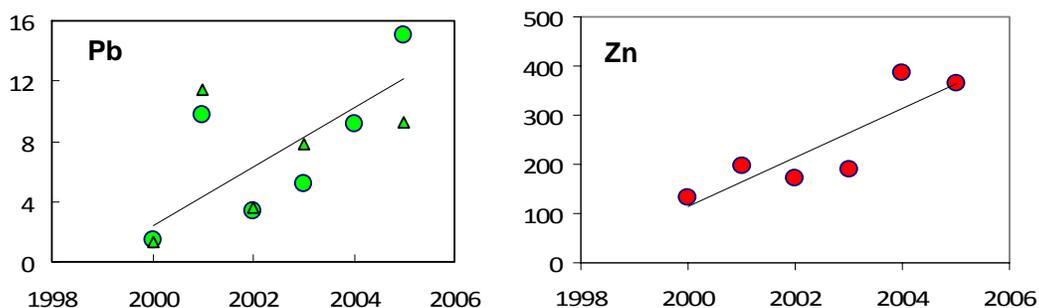
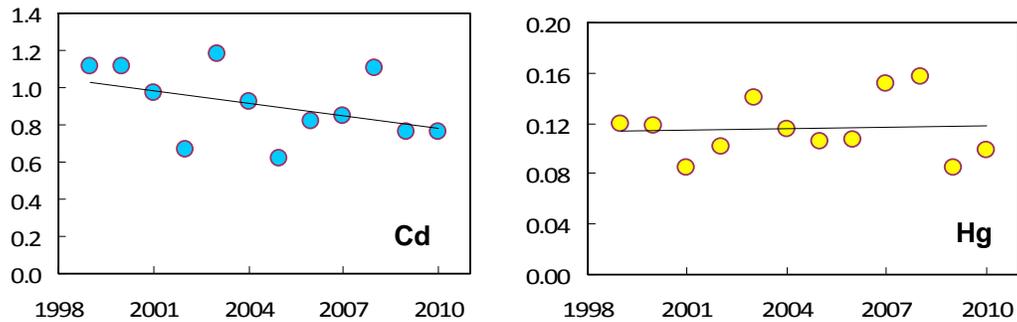


Figure 38. Temporal trends of Pb and Zn in *Mytilus galloprovincialis* from two stations of Croatia.

A long survey (1999-2010) has been performed by Slovenia in the Gulf of Trieste which has been considered chronically polluted by Hg and Pb. Consistent with the sediment data, the concentrations of Hg in mussels have not declined during this period (Figure 39A). However, a slight decrease has been observed for Cd as well as for Pb in the stations corresponding to the Italian coast (e.g. Bahia di Panzano).

Another site of concern is the area of Durres and Vlora Bay in Albania. As shown in Figure 39B, the concentrations of trace metals in mussels show a general increase for all of them during the last decade.

A - Gulf of Trieste (Slovenia)



B - Durres (Albania)

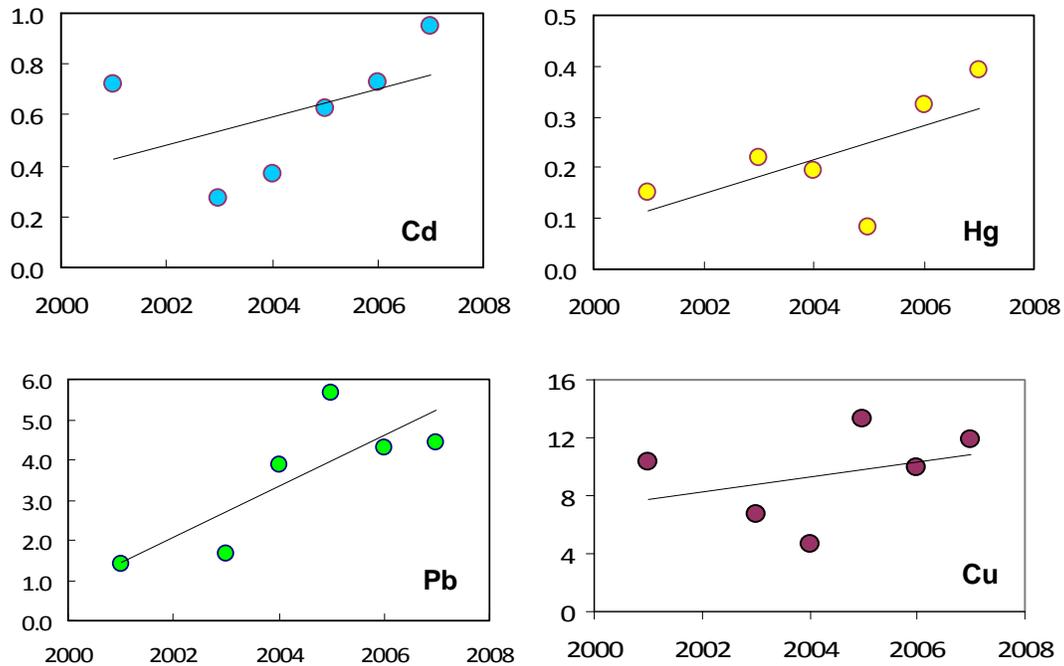


Figure 39. Temporal trends of trace metals in samples of *Mytilus galloprovincialis* from individual stations in the Adriatic.

As indicated above, and contrary to the Italian western coast, the Adriatic coast do not include areas of particular concern except those corresponding to costal lagoons (e.g. Grado and Venice) (see Figure 14). In general, the levels are not showing significant changes along the time. Particularly indicative is the decline of Pb in stations under urban influence. An example is illustrated in Figure 40 where the temporal trends of Cd, Hg, Pb and Zn in mussels collected in Lido Adriano (close to Ravenna) are shown.

However, Cd and Hg are still increasing in certain stations (e.g. Pescara, Grado Lagoon, etc.). The concentrations found in the Venice Lagoon do not follow a unique pattern. They are highly influenced by the characteristics of the site. Two contrasting situations are shown in Figure 41.

Lido Adriano (Italy)

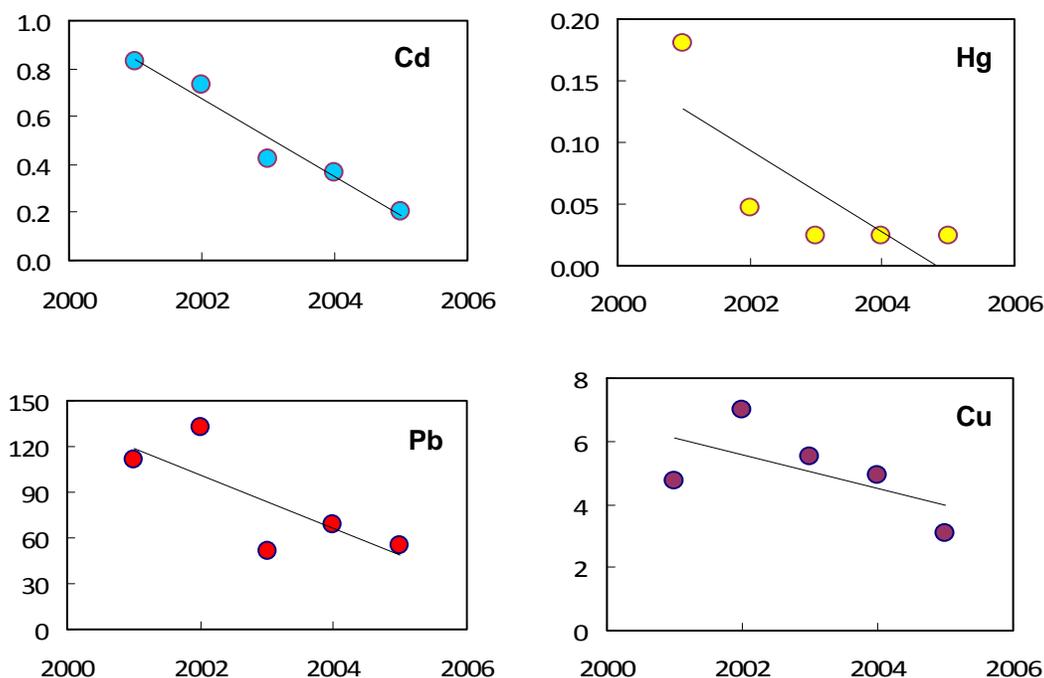


Figure 40. Temporal trends of trace metals in samples of *Mytilus galloprovincialis* from individual stations in the Adriatic.

Venice Lagoon (Italy)

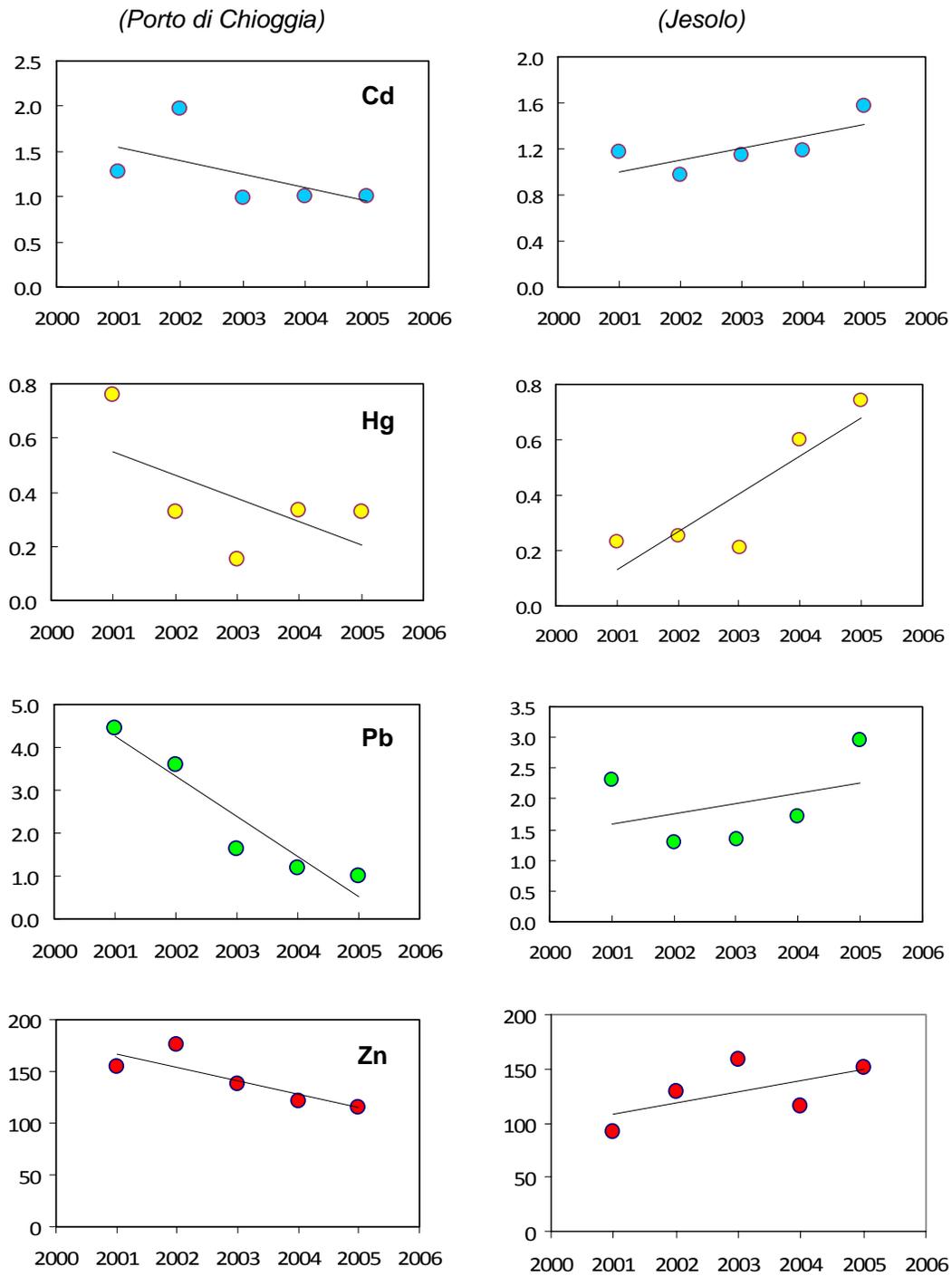


Figure 41. Temporal trends of trace metals in samples of *Mytilus galloprovincialis* from individual stations of the Venice Lagoon.

3.2.2.3 Central Mediterranean

As mentioned earlier, the coverage of this region is very limited and the only data allowing a trend assessment is that corresponding to the Bizerta Lake (Tunisia). As shown in Figure 42, Cd, Hg and Pb in mussels decreased during the period 2001-2008. Similar trends exhibit the clam *Ruditapes decussatus*, which has also been included in the monitoring of the area.

The stations belonging to the continental Italian coast (e.g. Taranto) and Sicily have been sampled less often and do not exhibit conclusive trends, either positive or negative.

Bizerta Lake (Tunisia)

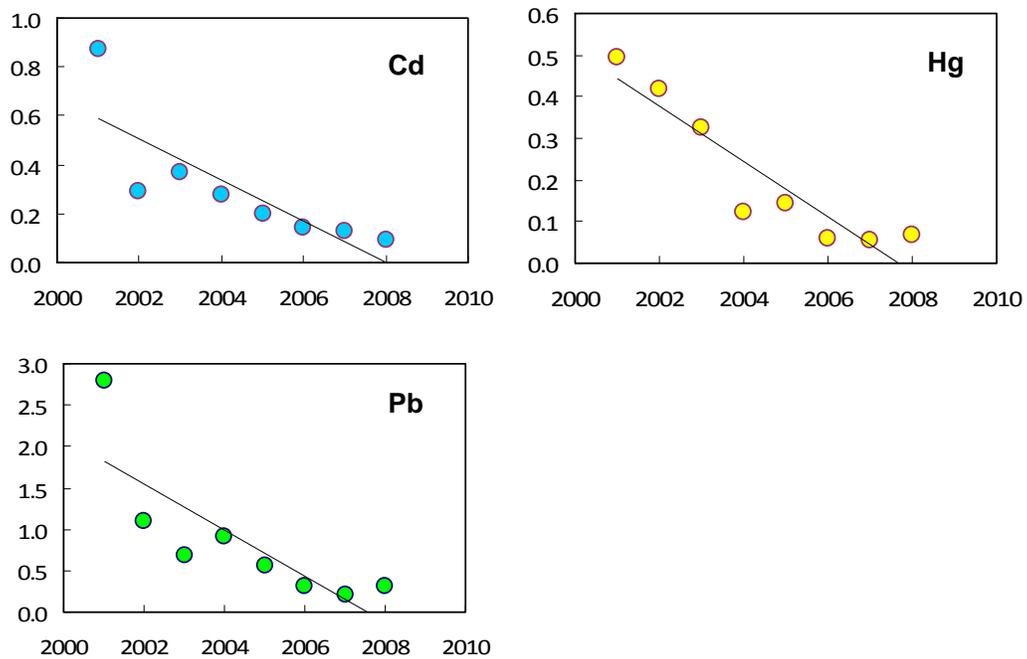


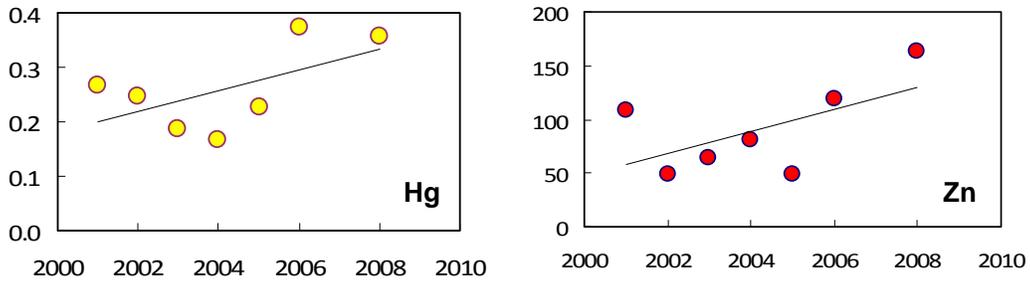
Figure 42. Temporal trends of Cd, Hg and Pb in samples of *Mytilus galloprovincialis* collected in Bizerta Lake (Tunisia).

3.2.2.4 Aegean-Levantine

In the region, there is no data on *Mytilus galloprovincialis* for assessing temporal trends. In Israel, *Mactra corallina* was used as indicator organism which exhibited relatively high values of Cd and Cu in the northern coast (Haifa). Despite the concentrations of trace metals in sediments revealed a consistent decline during the last 10 years (Figure 26), the levels found in clams show a significant increasing trend (Figure 43A).

On the other hand, In Turkey the red mullet *Mullus barbatus* was the selected indicator but only three stations in the south of the country were monitored (Mersin, Goksu and Tirtan). Here also the levels exhibit an upward trend (Figure 43B).

A - Haifa Bay (Israel)



B - Mersin (Turkey)

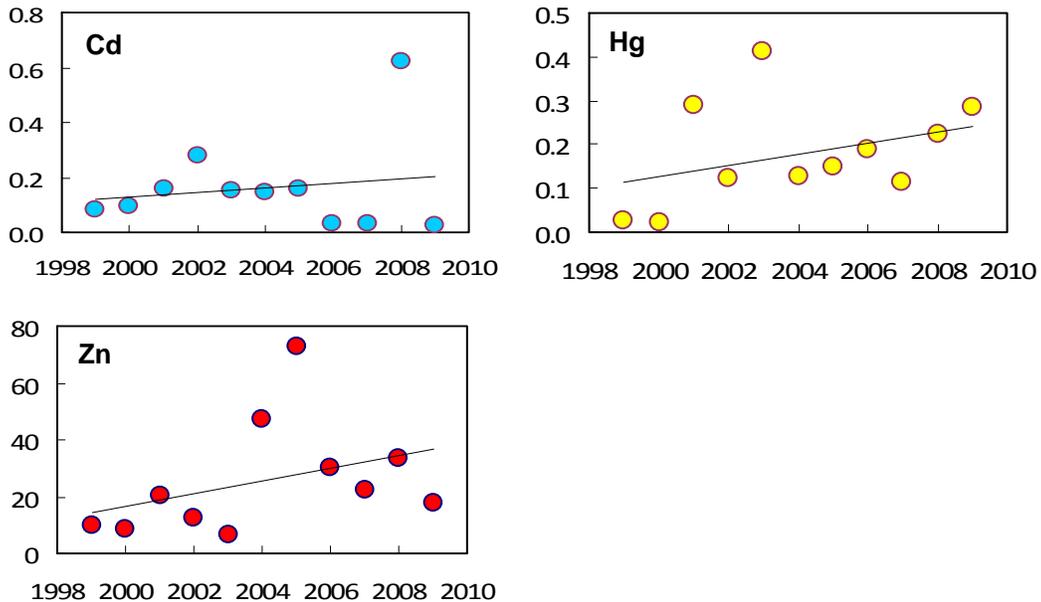


Figure 43. Temporal trends of Cd, Hg and Zn in samples of (A) *Mactra corallina* and (B) *Mullus barbatus* collected in Israel and Turkey, respectively.

3.2.3 Chlorinated pesticides in biota

DDT has been the most widely monitored pesticide. A general overview of the annual statistical data of concentrations of DDTs in samples of *Mytilus galloprovincialis* from various countries is shown in Figure 44. Two contrasting patterns are represented by Croatia and Albania. In the first case, there has been an apparent decrease of concentrations during the last decade whereas in the second it appears to be the contrary. However, in general, the overall trends are not so evident, although a drop in the number of stations exhibiting the higher levels is usually observed (e.g. France).

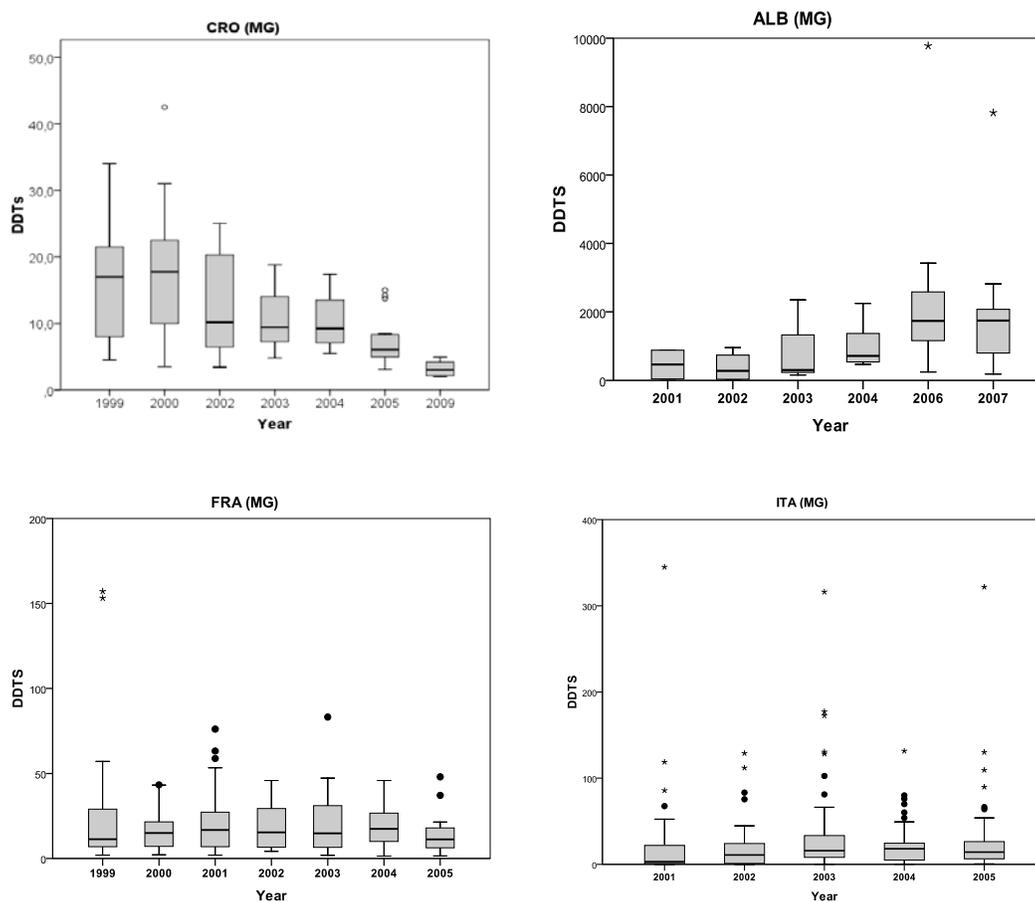


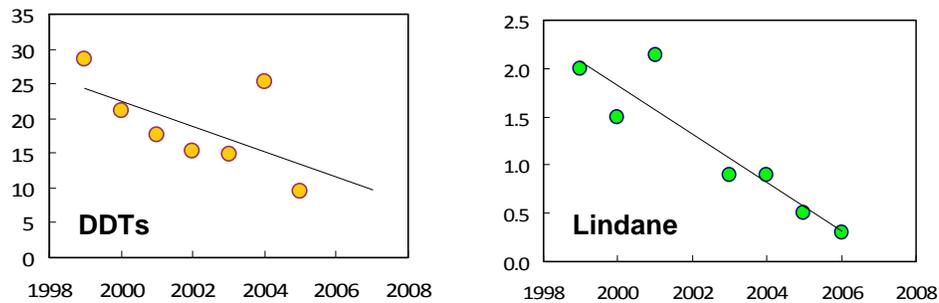
Figure 44. Temporal trends of overall concentrations of DDTs in samples of *Mytilus galloprovincialis* from various countries.

Data on the other pesticides (e.g. lindane and dieldrin) are scarcer but more clearly evidence a decline of concentrations over time. These features are better demonstrated at the level of individual stations for the different sub-regions.

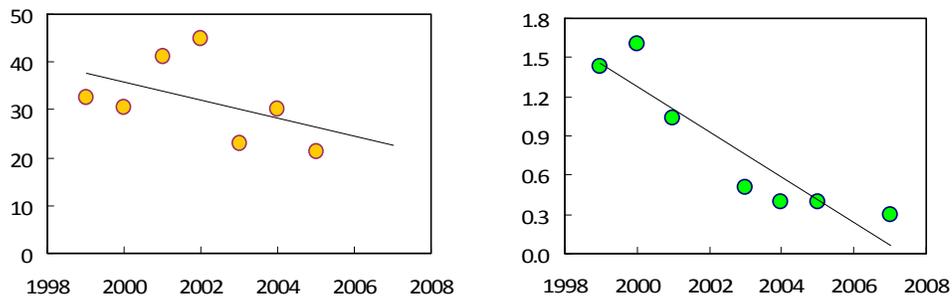
3.2.3.1 Western Mediterranean

A series of representative temporal trends for different stations of the Western Mediterranean is shown in Figure 45. As it can be seen, in all sites concentrations are decreasing, which is consistent with the banning of production and use of these compounds. In general, the decrease is also faster for lindane and the other chlorinated pesticides (not shown) than for DDT, that appears to be consistent with the higher long-life of the latter.

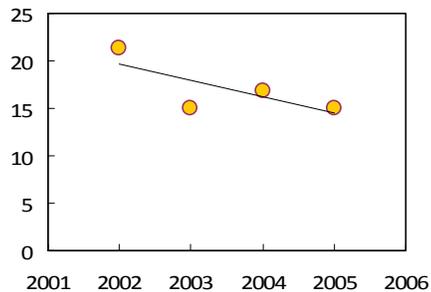
A - Etang de Thau (France)



B - Les Stes. Maries de la Mer (Camargue)



Fiumicino (Italy)



La Spezia (Italy)

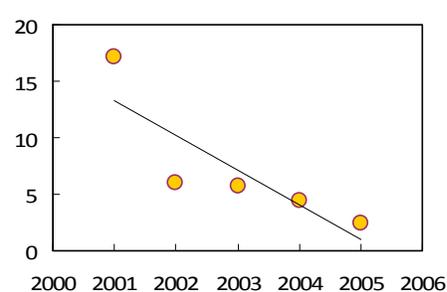


Figure 45. Temporal trends of DDTs and lindane in samples of *Mytilus galloprovincialis* from Western Mediterranean stations.

Similar trends were observed in the French monitoring network, which probably constitutes the most consistent data base for trends assessment (IFREMER, 2001). In general, it was found that during the period 1979-1998 the decreasing trends were in the order: Σ DDT > HCHs > PCBs. This may reflect that the regulation of the use of these chemicals and, consequently, of the contaminant inputs to the sea was more efficient for DDT and lindane than for PCBs. This is illustrated in Figure 46 where the concentrations of lindane in *Mytilus galloprovincialis* for the 23 stations of the Western Mediterranean, between 1982 and 2007, are displayed.

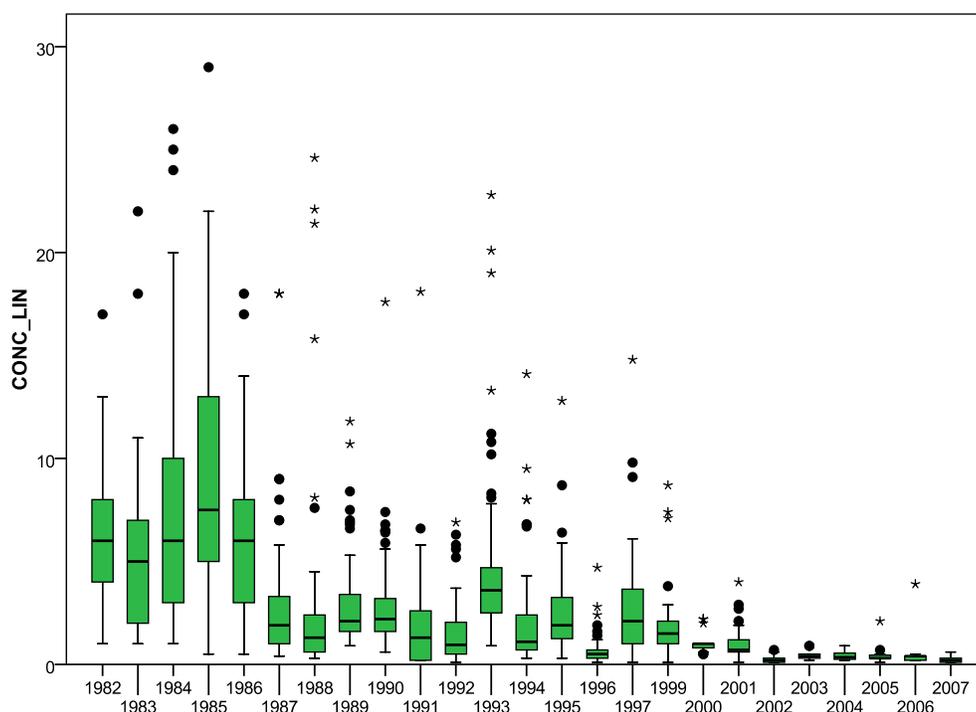
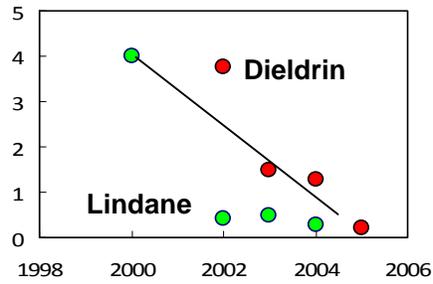
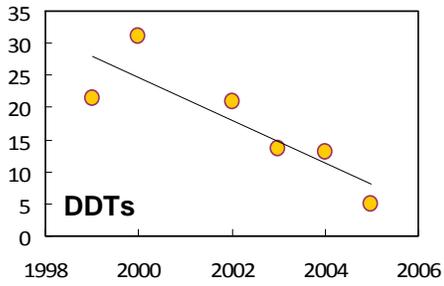


Figure 46. Temporal trends of lindane ($\text{ng g}^{-1} \text{dw}$) in *Mytilus galloprovincialis* from the RNO stations.

3.2.3.2 Adriatic

The Adriatic stations (e.g. Italy and Croatia) follow a similar trend, with also a more rapid decrease of concentrations of lindane, aldrin and dieldrin than DDTs (Figure 47). However, there are few exceptions such as the stations corresponding to the Albania coast, notably Durres and Vlora Bay, which is known to keep stockpiles of obsolete chlorinated pesticides. Notice also the extremely high levels found in mussels.

Kastela Bay (Croatia)



Vlora Bay (Albania)

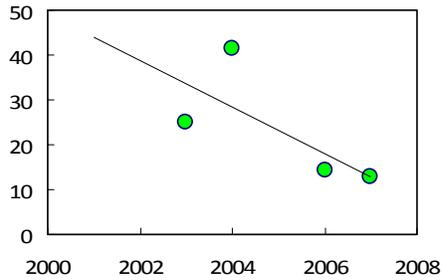
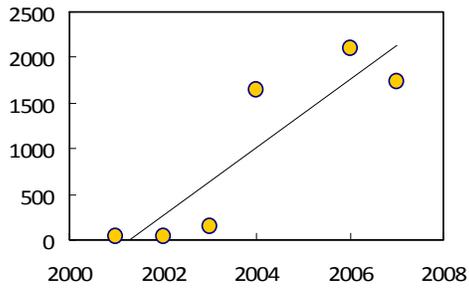


Figure 47. Temporal trends of DDTs, lindane and dieldrin in samples of *Mytilus galloprovincialis* from the Adriatic Sea stations.

3.2.3.3 *Central Mediterranean:*

Only few stations in the Central Mediterranean have been monitored over a period long enough to assess temporal trends. In any case, data show the same trends found in the previous regions as illustrated in Figure 48. Unfortunately, there are no data for the Southern coast.

Taranto (Italy)

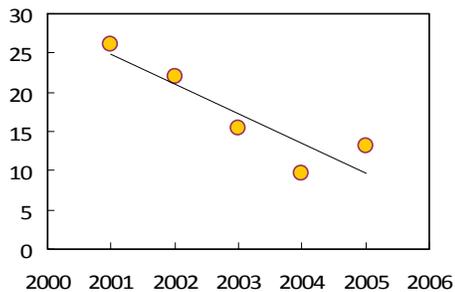


Figure 48. Temporal trends of DDTs, in samples of *Mytilus galloprovincialis* from a Central Mediterranean station.

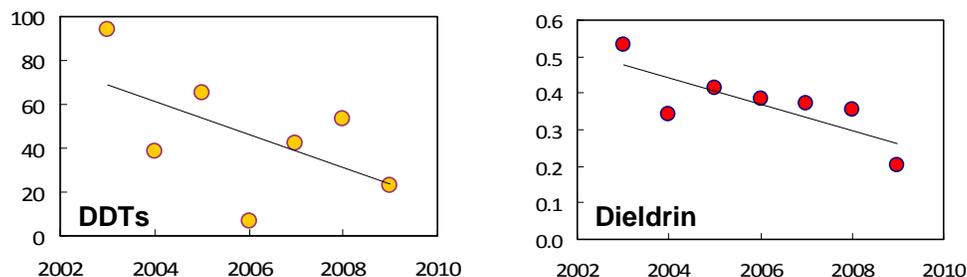
3.2.3.4 Aegean-Levantine:

In this region most data correspond to the red mullet *Mullus barbatus*, although only three stations at the southern coast of Turkey have been monitored. The profiles shown in Figure 49 indicate a downward trend of chlorinated pesticides during the last decade, although the variability of DDT data is rather large.

Conversely, the concentrations in mussels of the Ismir Bay, identified as a hot spot in the region, exhibit a significant increase, probably due to a less efficient management of the existing regulations concerning the stocking of these obsolete pesticides.

Examination of temporal trends for a period of 14 years (1988 – 2001) revealed that no significant differences in organochlorine contamination levels in fish and mussels from Hellenic waters existed during this time period, despite the banning of these compounds since the 1970s (SoHeIME, 2005).

Mersin (Turkey)



Ismir Bay (Turkey)

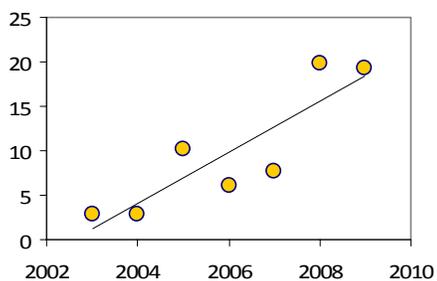


Figure 49. Temporal trends of DDTs and dieldrin in samples of *Mullus barbatus* (Mersin) and *Mytilus galloprovincialis* (Ismir Bay) from the Aegean-Levantine region.

3.2.4 PCBs in biota

The assessment of temporal trends of PCBs is more difficult because the lack of long-term consistent data mainly due to the change in concentration units (from Aroclor to individual congeners) during the last decade. The profiles of the overall concentrations shown in Figure 50 do not allow deriving any conclusion.

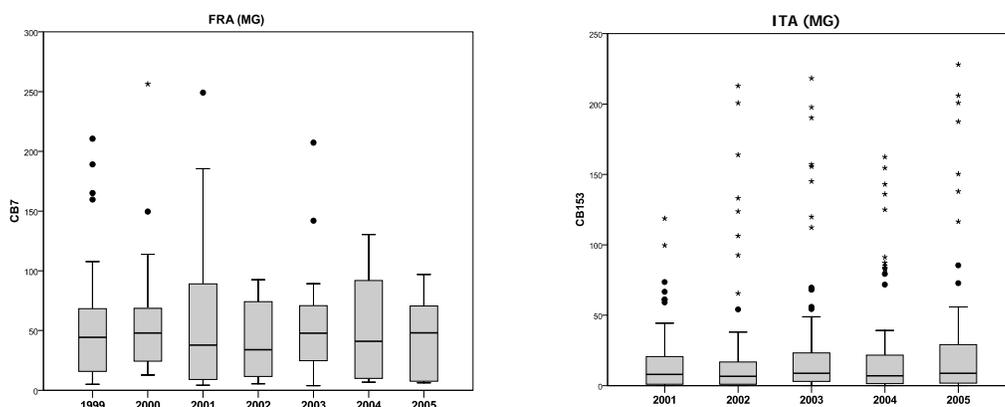


Figure 50. Temporal trends of PCBs in samples of *Mytilus galloprovincialis* from various countries

The temporal trends for some individual stations, shown in Figures 51 and 52, generally reveal conservative or even increasing trends of PCBs in mussels, probably reflecting an inefficient management of the existing regulations concerning their use and stocking, as was the case for DDT.

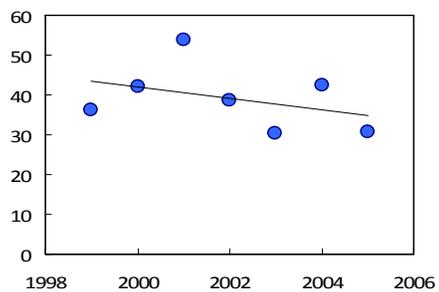
In the Western Mediterranean, the stations evidencing relatively high levels of PCBs are still exhibiting increasing trends whereas those more pristine seem to be stable or decrease (Figure 51).

For the other regions there is limited availability of data to allow drawing precise conclusions. A few stations show steady or decreasing trends whereas others the levels are still increasing in both mussels and fish (Figure 52).

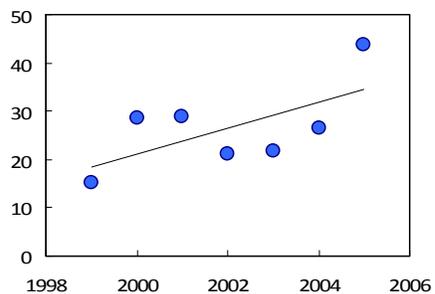
Temporal trends from the Greek UNEP/MAP-MED POL programme also indicate no reduction of pollutant levels despite the ban, indicating continuous inputs into the coastal environment (SoHeIME, 2005).

3.2.4.1 Western Mediterranean

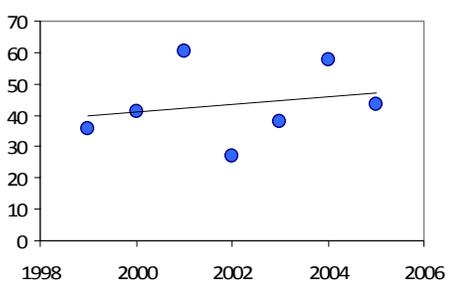
Les Stes. Maries de la Mer (Camargue)



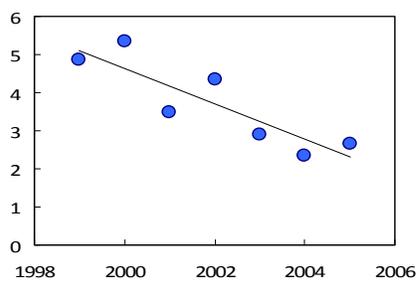
Etang de Thau (France)



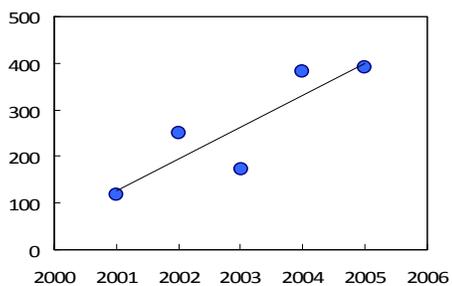
Golfe de Fos (France)



Corsica (France)



Vado Ligure (Italy)



La Spezia (Italy)

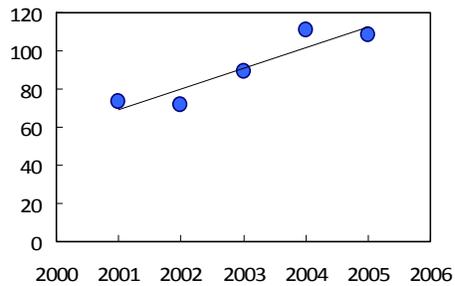
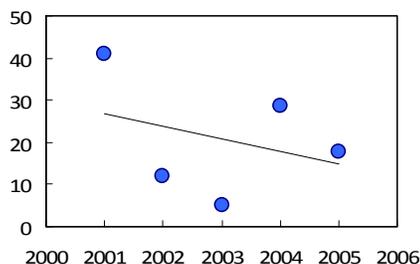


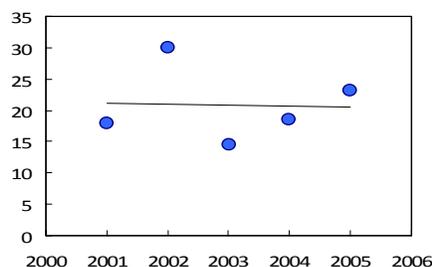
Figure 51. Temporal trends of PCBs in samples of *Mytilus galloprovincialis* from individual stations from different sub-regions.

3.2.4.2 Adriatic and Central Mediterranean

Bari (Italy)

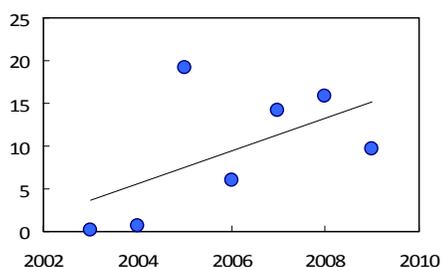


Taranto (Italy)



3.2.4.3 Aegean-Levantine

Ismir (Turkey)



Mersin (Turkey) – *Mullus barbatus*

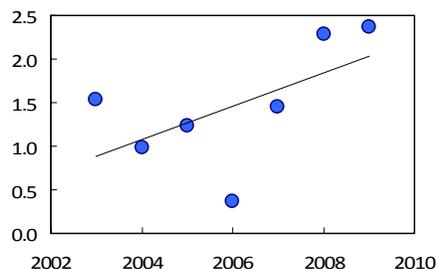


Figure 52. Temporal trends of PCBs in samples of *Mytilus galloprovincialis* and *Mullus barbatus* from different stations of the Adriatic, Central Mediterranean and Aegean-Levantine regions.

A long term study of PCBs and DDTs temporal trends in mussels conducted in the Saronikos Gulf during the years 1988 – 2001 did not exhibit a clear decreasing trend, despite the declining use and progressive elimination of these chemicals (SoHeIME, 2005). This stability of PCB concentrations over the years could not be explained only by the high persistence of these compounds and probably indicates that there are still continuous inputs into the coastal environment either by atmospheric deposition, which is considered as the most important recycling route, or by direct discharges. However, although the results of PCB analysis after 1988 are not directly comparable with previous measurements due to different analytical and calculation practices, it seems that a decline occurred during the 1970s and early 1980s. DDTs concentrations in mussels appear also relatively constant after 1990 while some unexpectedly high values were measured during 1996.

A recent assessment of the Mediterranean sediments contamination by persistent organic pollutants (Figure 53) (Gomez *et al.*, 2007) has also concluded that a decreasing trend is more evident for DDTs than for PCBs, indicating a steady input of the latter in the Mediterranean Sea and the need for an improved management of their potential sources. Previously, Picer and Picer (1991 and 1995) were also able to show different temporal trends for PCBs and DDTs in surficial sediments and benthic fish collected in the Adriatic from 1976-1990.

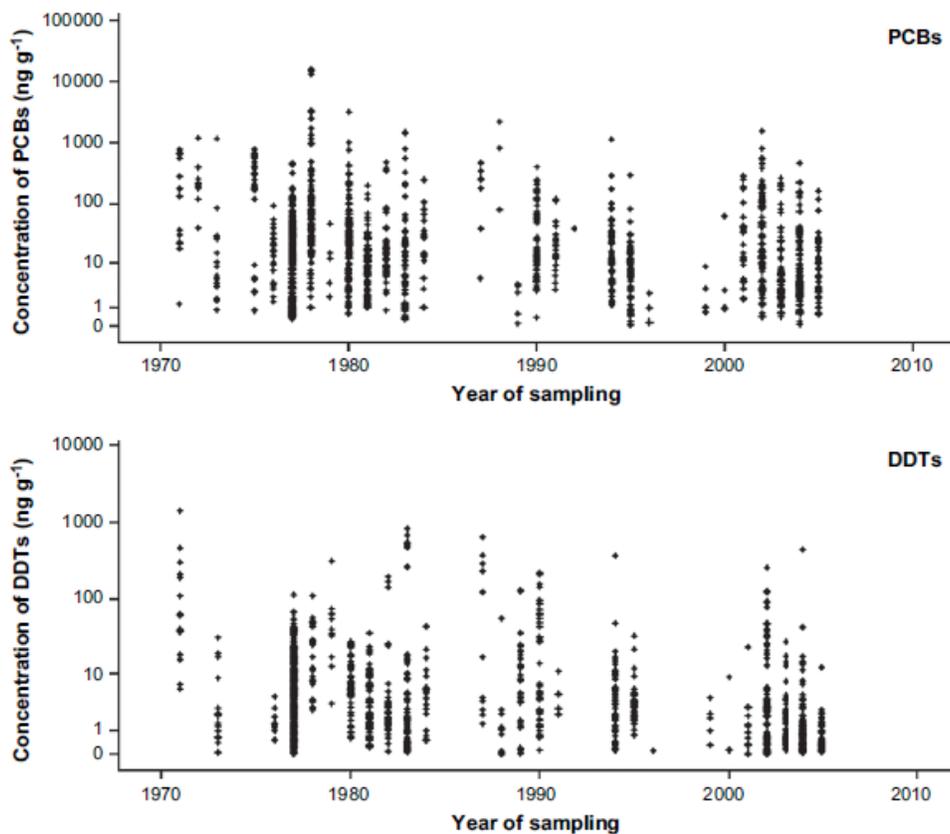


Figure 53. Concentrations of PCBs (Aroclor eq.), and DDTs in Mediterranean sediments with respect to the year of sampling (Gómez-Gutiérrez *et al.*, 2007)

4. Conclusion

The MEDPOL monitoring database (MEDPOL.mdb) and the inter-linked web version (<http://195.97.36.231/medpol/>) constitutes a relevant source of information for assessing the state of the Mediterranean Sea. The efforts made during the MEDPOL Phase III and Phase IV have been successful in building up and improving this essential instrument of environmental policy. Although at the moment is hosting monitoring data of only 14 Mediterranean countries, and the portion of data for each component and country is uneven, it constitutes the most comprehensive record of monitoring data for the whole basin. This, together with the available data from individual studies reported in the literature, represents the fundamental body of information for supporting the environmental policy in the region.

However, from the above analysis, it must be stressed that there is a need to establish monitoring programs in many countries, particularly from the southern riparian coast, to fill the geographical data gaps and ensure the continuation of existing temporal trend data. It should be taken into account that for evidencing and assessing significant temporal variations a sufficiently large time span of data is required (>10yr for sediments and >5 yr for biota).

These programs must be able to generate comparable and accurate data, taking into account the intrinsic variability of the environmental matrices considered. For example, the adoption of normalization procedures which could account for the differences in sediment characteristics (organic carbon or Al contents) as well as the implementation of quality assurance/quality control procedures are considered essential.

A useful outcome of the database can be the establishment of background concentrations for the target compounds in Mediterranean biota and sediments, which is necessary in order to have reference values for comparison with field data.

Moreover, with the improvement and development of analytical techniques, the identification and quantification of emerging substances of potential concern for the marine environment because of their persistence, toxicity and bioaccumulation properties, is continuously increasing. They are believed to be ubiquitous but information on their occurrence in the Mediterranean is limited and should be improved.

Finally, the conceptual approach of the MEDPOL Program, updated with the recent knowledge and experience generated by the scientific community needs to incorporate relevant assessment tools for hazardous substances in marine sediments and biota. Specifically, there is a need to establish environmental assessment criteria (EAC) for the hazardous substances included in the MEDPOL database: trace metals, chlorinated pesticides and PCBs.

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Annex I. Yearly and Country Statistics of MED POL Database

Yearly Statistics

<i>Year</i>	<i>Number of stations</i>	<i>Number of samples</i>	<i>Number of parameters</i>	<i>Number of values</i>
1999	167	1139	91	5683
2000	108	988	95	5188
2001	183	1257	109	9648
2002	189	1395	102	10845
2003	187	1327	102	11974
2004	445	2890	153	23990
2005	370	1721	149	17574
2006	237	1004	84	8280
2007	229	901	97	8008
2008	109	653	64	5366
2009	142	1287	61	9501
2010	1	9	6	54
Total	800*	14571	219*	116111

Country Statistics

<i>Country</i>	<i>Number of stations</i>	<i>Number of stations without coordinates</i>	<i>Number of samples</i>	<i>Number of parameters</i>	<i>Number of values</i>
Albania	7	0	91	29	795
Bosnia-Herzeg.	4	0	57	17	245
Croatia	72	5	1838	81	12472
Cyprus	97	0	886	43	4742
Egypt	10	0	39	10	163
France	21	0	673	59	6519
Greece	235	35	2068	66	14336
Israel	69	0	2557	26	14492
Italy	92	2	2113	50	26245
Morocco	35	0	122	28	909
Slovenia	28	1	2133	66	13334
Spain	32	0	633	40	11480
Syria	11	0	21	8	89
Tunisia	17	3	321	15	680
Turkey	70	0	1019	42	9610
Total	800	46	14571	219*	116111

*Only unique stations and parameters are counted

Annex II. Available data of hazardous substances in the MED POL Database

Number of samples analysed for trace metals in different matrices

Parameter	BIO	SED	AIR	RIV	WAT	Total
Cu	3446	810	175	133	58	4622
Zn	3244	856	173	156	50	4479
HgT	3188	809		129	16	4142
Cd	2675	387	175	131	41	3409
Pb	1702	754	175	129	46	2806
Cr	1558	819	175	90	51	2693
Ni	1008	690		98	45	1841
As	898	555				1453
Al	329	243	175			747
Co	20					20
Total	18068	5923	1048	866	307	26212

Number of samples for OCPs by matrix

Parameter	BIO	SED	RIV	Total
DDT	278	16	27	321
DDE	240	14		254
DDD	233	17		250
DDTs	172	20		192
ALD	260	22	27	309
LIN	268		27	295
HCB	260	19		279
DIE	214	17	27	258
END	156		27	183
HEP	55	7	27	89
HCH	31	14		45
ENDOS	4		27	31
DSRI	16			16
Total	2187	146	189	2522

Number of samples for PCBs by matrix

Parameter	BIO	SED	RIV	Total
CB28	386			386
CB52	412			412
CB101	377			377
CB105	346			346
CB118	346			346
CB138	418			418
CB153	398			398
CB156	258			258
CB180	416			416
CBs	52			52
PCB 1254	193	19		212
PCB 1260	159	19		178
PCBT		10	27	37
Total	3761	48	27	3836